**HIGH-LEVEL RADIOACTIVE
WASTE MANAGEMENT ALTERNATIVES**

SECTION 7: WASTE PARTITIONING
SECTION 8: EXTRATERRESTRIAL DISPOSAL
SECTION 9: TRANSMUTATION PROCESSING

May 1974

Battelle

Pacific Northwest Laboratories
Richland, Washington 99352

Prepared for the U.S. Atomic Energy
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FOREWORD

This report is a comprehensive overview study of potential alternative methods for long-term management of high-level radioactive waste. The study includes a compilation of information relevant to technical feasibility, safety, cost, environmental considerations, policy conflicts, public response and research and development needs for:

1. Disposal in terrestrial locations
 - a. In geologic settings on land
 - b. In the seabed
 - c. In ice sheets
2. Disposal into space
3. Elimination by transmutation (nuclear transformation of certain waste constituents into nuclides having less long-term toxicity).

The study is limited to the management of high-level radioactive waste from nuclear power by variations of these alternatives. Consideration of alternative types of electrical power generation are not within the scope of the study. In addition, evaluation of interim storage of radioactive waste in retrievable surface storage facilities is not part of this study. Disposal of waste in bedded salt deposits was studied extensively in other AEC programs, and the concept is included here as part of the overall matrix of geologic disposal techniques.

To complement these studies, investigations were also conducted on waste partitioning (separation of radionuclides in radioactive waste into different elements or groups of elements according to their long-term toxicity or suitability for different disposal methods), and systems methodology was developed to assess the effects of radionuclides from waste introduced into man's ecological cycle, assuming some failure of the primary waste containment.

Information pertinent to evaluating the various potential waste disposal techniques was developed without promoting any single disposal concept. The study is concerned with management of the waste and does not consider the potential for recovery of resources within the waste, including the heat. Concepts are developed only to the detail necessary to describe them for the overall investigation and in general are studied on a systematic, generic basis. This information can be used in comparing and assessing the various disposal concepts as a basis for decisions regarding their further study.

The evaluations of feasibility are not restricted to currently available technology. Rather, the study attempts to take into account technology which can be developed or is expected to be available at least within the next four decades. Indeed, most of the concepts studied are estimated to require 15 to 30 years for full implementation.

The study includes most currently known waste management alternatives, but is not considered to be all-inclusive. As new data become available, and as new or varied concepts become evident (e.g., disposal in rocks in permafrost areas, isotopic dilution of selected materials, etc.,) comparable follow-on studies will be carried out.

This investigation has been performed largely by a multiple-discipline technical staff at the Pacific Northwest Laboratory of Battelle Memorial Institute with significant input from a large number of consultants and outside contributors. This wide involvement of persons was an attempt to assure up-to-date and accurate coverage of the broad scope of subject matter, including areas where there are diversities of opinions.

This report is issued as nine major sections in four volumes:

Volume 1	Section 1	Summary ^(a)
	Section 2	Background and Data Base
	Section 3	Evaluation Methodology
Volume 2	Section 4	Geologic Disposal
Volume 3	Section 5	Ice Sheet Disposal
	Section 6	Seabed Disposal
Volume 4	Section 7	Waste Partitioning
	Section 8	Extraterrestrial Disposal
	Section 9	Transmutation Processing

Appendix material is included with its own respective volume.

In general, metric system units are used in this report. Conversion factors to English units are given in Appendix 1.A.

a. This section is almost identical to WASH 1297, High-Level Radioactive Waste Management Alternatives, US AEC Division of Waste Management and Transportation, May 1974.

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7.0 POTENTIAL FOR WASTE PARTITIONING

High-level radioactive waste generated in nuclear fuels reprocessing plants contains radionuclides having a wide range of half-lives--from less than 100 days to millions of years. Current practice is to treat the high-level waste as a single entity through storage, solidification and perhaps ultimate disposal. Such "total waste" will remain radioactively toxic for a time comparable to the time periods for major geological changes in the earth's crust.

An alternative approach in managing high-level radioactive waste involves separating the total waste into fractions of different half-lives. Short-lived fractions would then decay to become radioactively non-toxic in relatively short times--times short enough to be within man's ability to control the waste storage. Long-lived fractions--significantly reduced in mass, volume and radioactive decay heat output--could be considered for other treatment, e.g., transmutation to non-radioactive or short-lived nuclides, extraterrestrial disposal, or special terrestrial disposal. Extraterrestrial disposal and transmutation processing are discussed in Sections 8 and 9, respectively, in this volume.

The basic guideline for this study was to obtain a comprehensive, evaluative overview of separations technologies and their applicability for removing the actinides and long-lived toxic fission products from high-level waste. Major results sought from the review were identification

of separations concepts which show promise for high-level waste separations and identification of areas where research and development is required.

7.1 BENEFITS TO BE GAINED

It is potentially possible to segregate high-level radioactive waste into numerous fractions based on various properties such as half-life, decay heat generation, and mobility in man's immediate environment. Consideration of the various approaches to radioactive waste management being evaluated and discussed in this report, and the waste segregation needed to pursue these approaches, led quickly to the conclusion that a waste segregation based on half-life contributes toward these approaches. Separation of waste into two fractions, one containing nuclides which will decay to negligible radioactive toxicity in the order of 1,000 years, i.e., having nuclides with half-lives of less than about 100 years, and a second fraction containing the long-lived nuclides, would provide waste packages fitting the needs or capabilities of several of these waste management alternatives.

Initial considerations were based on the concept that the short-lived fraction of the waste should be decontaminated from the long-lived fraction to the extent that the short-lived fraction, solidified to a calcine or glassy form and aged for 1,000 years, would contain no more than 1 to 2 n Ci/g of any long-lived

nuclide. This short-lived fraction would, after about 1,000 years time, represent no significant radiological toxicity to man and could be released to man's immediate environment with no more control than that required from a chemical viewpoint. Storage of waste in manmade structures for this length of time is well within reason. Also, 1,000 years is a short time period compared to the times required for major naturally occurring geological changes in the earth's crust. Disposal of the short-lived fraction in the earth's crust prior to 1,000 years would present little likelihood that the waste would be returned to man's environment prior to decay to negligible radioactive toxicity because of naturally occurring geologic changes.

Emphasis was given to separating the actinide elements from the short-lived waste fraction since these elements have long-lived isotopes and a high toxicity to man. However, it became apparent that some elements other than the actinide elements also have radionuclides which present significant toxicity to man after the wastes have aged a thousand years. Significant among these are radionuclides of I, Tc, Sm, Sn, and Ni (Ni enters the waste as a result of some dissolution of non-core fuel element components). The decontamination factors required for removal of these elements from the short-lived fraction of high-level waste to meet the stringent criterion stated above range from about 100 for I to greater than 10^6 for actinide elements. By comparison, Purex fuel reprocessing

plants achieve a separation of U from fission products of 10^7 , a separation of Pu from fission products of more than 10^8 , a separation of U from Pu of more than 10^7 , and a separation of Pu from U of 10^6 . They achieve 99.5 to 99.9% recovery of U and Pu.

If the "dilution" of waste within a geologic formation, the low leach rates of many solidified waste forms and the sorption of radionuclides in the soil are taken into account, it may well be that the concentration of long-lived radionuclides in the short-lived fraction could be considerably higher than the above criterion without posing a significant risk to man through terrestrial disposal of the short-lived fraction.

Consideration of factors such as these is a part of the methodology for Failure Mode and Radiological Pathway Analyses discussed in Section 3 of this report series. Results of future analyses using these methodologies may be used to define better the separations requirements for any given waste repository concept; different separations requirements may be indicated for various repository concepts even if the same criteria are used.

The risk analyses have been completed for one sample case only and its use for determining partitioning requirements is not established. For the repository concepts analyzed to date, preliminary example use of the risk evaluation models indicated that only the actinide elements need to be removed from the short-lived waste fraction. Also indicated was that removal of the actinides from the short-lived fraction by a decontamination

factor (DF) as low as 100 may be adequate. Moreover, the primary actinide element of direct concern is Am, and U-233, U-234 and the daughters of these isotopes are the dominant risks for time periods greater than 100,000 years. Since Pu-238 is a major source of U-234, Pu partitioning may be required to reduce the risk from U-234.

7.2 PARTITIONING PROCESSES

An overall assessment was made of the technical feasibility of chemically separating high-level radioactive waste generated in nuclear fuel reprocessing plants into fractions with different long-term biological toxicity. Major emphasis was given to process concepts which would result in isolating the actinide elements from the remainder of the waste. This type of separation is called waste partitioning in this study. Less emphasis was given to the isolation of long-lived fission products (i.e., I, Tc, Sm and Sn) and Ni from the remainder of the waste. Separation of this kind is called waste fractionation. The reasons for this relative emphasis and the interest in removing these elements from the waste were developed in the Subsection 7.1. The results of this study are summarized here. Details of the study are presented in report BNWL-1776.⁽¹⁾

Considerations of the requirements for separating actinide elements from high-level radioactive waste identified three major problems or areas for study. One problem concerns the solids which are always present in

high-level waste and in the process streams in a fuel reprocessing plant. The extent to which such solids contain actinide elements requiring removal to meet partitioning objectives and the extent to which they interfere with the performance of partitioning processes must be defined and dealt with. A second problem is the choice of the most advantageous processing options to achieve actinide element partitioning needed for various waste disposal alternatives. Past experience and literature pertinent to the kinds of separations required in a waste partitioning program is very extensive. Consideration of this extensive background, its application to partitioning needs and choice of the most promising approaches comprise a major task. A third area concerns the requirements for analytical control of waste partitioning. The detection and measurement of very small amounts of actinide elements in the short-lived waste fraction with its very high beta-gamma radioactivity is a formidable problem; the question of whether adequate analytical control techniques exist or can be developed is pertinent.

The study sought to make as complete a review as possible of the technology pertinent to the three major problems identified above. An extensive literature review was made. Also input from recognized experts in areas pertinent to the problem was sought. Approximately 25 experts outside BNW were contacted during this study. Included were individual consultants, personnel from other AEC

installations and personnel from industrial firms with acknowledged expertise in specific separations techniques. The contributions of these experts are reflected in the content of the report.

Two basic concepts for achieving actinide element partitioning were defined. In one, the partitioning would be accomplished within the fuel reprocessing plant by suitable modification to improve the uranium and plutonium recovery and to isolate other actinide elements. In the second, the partitioning would be done on the waste as normally produced by the fuel reprocessing plant. The former is discussed as "repartitioning" and the latter as "adjunct partitioning" in reference 1.

Since waste composition will depend on many factors (e.g., reactor type, fuel exposure history, cooling time), the study sought to make the evaluations independent of waste composition. Separations processes will, in practice, have to accommodate waste of various types. However, most information currently available is based on waste from LWR fuel with relatively low burnup in comparison with exposures anticipated for the future. Although this study attempted to identify the problems which should be generic to all types of waste, the results are potentially limited by the possibility that present information cannot be extrapolated reliably to waste from the mixed nuclear power economy of the future.

The chemical separations technologies shown in Figure 7.1 were evaluated. Physical separation technologies (such as gaseous or liquid diffusion or gas centrifuging) which use the larger and heavier molecules of the actinides compared with those of the smaller fission products as a basis for separation, may also have some potential for partitioning, but study of these concepts was beyond the scope of this investigation. Figure 7.1 illustrates the fact that suspended solids or colloids in the waste are potentially a significant problem which could limit or define the feasible separations technologies. Solids are expected to be present in the aqueous waste; if treatment of them is needed, operations such as shown in Figure 7.1 would be needed prior to separations. As shown in Figure 7.1, the possibility that separations would have to be done on a solid waste phase introduces the need to consider pyrometallurgical, slagging, and other solids-based separations techniques within the scope of this study.

Rapid decisions on the feasibility of concepts were sought via consultation with the team of acknowledged experts mentioned earlier. Those concepts which appeared to have little chance of achieving the degree of separation needed for partitioning, which are inherently costly or which would require very extensive Research and Development for application to the partitioning problem were deemphasized in favor of the more promising concepts.

FIGURE 7.1. Basic Waste Partitioning Options

7.2.1 Solids

Solids will play a prominent role in the development of waste partitioning processes. Some of these solids are present in the dissolved fuel solution prior to solvent extraction in a reprocessing plant. Although the literature contains some information on quantities and composition of dissolver solution and high-level waste solids, most of the data are based on laboratory-scale studies. Very little data are available based on plant-scale operation with power reactor fuels. Plutonium has been identified in dissolver solution solids, but little is known about the occurrence of other actinide elements

on these solids. At present there is essentially no information useful to waste partition studies on the content of high-level waste solids. Research and Development on the content of dissolver and waste solids, on the separation of the solids from the liquid and on the processing of the solids to remove actinide elements is needed.

Potential procedures for partitioning waste solids (e.g., solidified waste or solids removed from waste slurries) as shown in Figure 7.1 were evaluated. It was concluded that such separations as slagging, volatility processes and pyrometallurgical processes present formidable problems

with relatively low probability of achieving the degree of separation needed for waste partitioning. The separations achieved in most of these processes are not clean. They present major problems in terms of high-temperature operation, control of volatile components, possible production of large quantities of contaminated waste and relatively high operating hazard. In general, major Research and Development including extension of basic knowledge would be required to develop feasible and effective partitioning processes. These processes were deemphasized early in the evaluation review.

If removal of actinide elements from solids separated from waste slurries is required (which remains to be determined from future studies on the actinide element content of such solids and the results of the Failure Mode and Radiological Pathway Analyses studies), development of procedures for accomplishing this will be required. Fusion-leaching procedures offer some promise. If adequate separation of actinides from the waste solids is impractical to achieve, the solids could be considered for combining with the actinide waste fraction. If further study shows long-lived radionuclide content of the solids to be adequately low, the solids may be added to the short-lived waste fraction.

7.2.2 Separations

In considering the application of known separations processes to partitioning of waste, it should be remembered that the objective is essentially the reverse of that for a fuel

reprocessing plant. In the latter a high degree of separation of U and Pu and perhaps Np from all other constituents of the irradiated fuel (and from processing equipment corrosion products) is sought. While high recovery of U and Pu (i.e., separation of these two elements from the remainder of the constituents) is desirable, it is not economically practical to achieve a degree of separation comparable to that required for waste partitioning. On the other hand, the objective of waste partitioning is to achieve a high degree of separation of all of the actinide elements from the remainder of the irradiated fuel constituents. Separations processes which have been developed for nuclear materials have sought a reasonable recovery and high purification of a desired constituent rather than a very high degree of removal of the desired constituent from the waste.

As shown in Figure 7.1, many basic liquid-phase partitioning concepts were considered. A consensus was quickly established, however, among the staff and consultants who participated in the study, that solvent extraction and ion exchange offered far greater promise for effectiveness and flexibility than the alternative concepts. This consensus was confirmed by communication with experts on the other concepts (e.g., flotation, dialysis, etc.). These other concepts are at this time not sufficiently developed, or are not sufficiently effective for use, or are beset by process problems (e.g., membrane degradation) that would preclude their use. For these reasons, major attention was centered on

solvent extraction and ion exchange. Use of these techniques for separations such as would be required by partitioning has been widely explored. Their use would therefore represent relatively straightforward extension of existing technology, maximum compatibility with present operations, and minimum investment in future Research and Development programs. A summary of separations feasibility conclusions is given in Table 7.1.

Detailed evaluation of problems which could limit practical application of solvent extraction and ion exchange (e.g., generation of large volumes of low-level liquid waste or contaminated waste ion-exchange resin) was beyond the scope of this work. This evaluation can be done with maximum cost effectiveness after actual partitioning requirements are better defined.

A large volume of literature on solvent extraction and ion exchange (abstracted and referenced in Reference 1) was reviewed with respect to application to the separations needed for a waste partitioning program. This survey of solvent extraction indicates that the approach can be used to accomplish the partitioning and/or fractionation that might be needed. Extraction of the actinides, Sm, Tc and probably Sn, can be achieved. Tracer experiments have shown that the concentrations in the remaining waste can be reduced to low levels at least for the actinides, and conventional techniques employing multistage extraction should accomplish adequate removal and separation. Nickel may be a special problem and every attempt should be made to keep it out of the feed. However, selectivity to provide a small package of

TABLE 7.1. Partitioning Feasibility Study Conclusions:
Adequacy of Existing Technology^(a)

	Actinide Separation (DF) ^(b)		
	10-100	1,000-10,000	10 ⁶ -10 ⁸
Solvent Extraction	Yes	Yes	Possibly
Ion Exchange	Yes	Possibly	No
Other Separation Techniques	Possibly	No	No
Analytical Capability	Yes	Possibly	No

- a. Existing separations technology needs adaptation to the objectives of partitioning.
- b. This study was concerned primarily with the adequacy of existing technology for obtaining adequate separation of actinide elements from the short-lived waste fraction. Adequate technology exists for obtaining needed purity of the separated actinide fraction.

these elements has not been adequately demonstrated. Complications from radiation damage to the solvent are of some concern, although for a ten-year-cooled waste these do not appear serious at first consideration. Capability of partitioning without producing major new waste streams either contaminated with the problem elements or so large in volume that they create new storage problems remains to be demonstrated. Research is required to determine the best solvents to make the separations at the desired points. A several-step process possibly involving more than one solvent may be required. Such an approach is shown in Figure 7.2. A logical approach is to follow one or more solvent extraction steps with another technique such as ion exchange.

This review has generally assumed that waste which has been stored in the (nitric) acid form would be the feed for solvent extraction. From a materials standpoint, this is the preferred situation, although other acid systems are not ruled out at this time.

Information reviewed in this study indicates that ion-exchange technology that can meet low-to-moderate partitioning or fractionation needs is available. Some experience has been obtained with each of the elements that might have to be isolated.

Process problems rather than separations effectiveness are most likely to restrict the practical use of ion exchange for partitioning or fractionation. The major problem is that lanthanides would be absorbed with Am and Cm with consequent high radiation

FIGURE 7.2. Partitioning Approach Using Solvent Extraction

dose to the exchangers and need for further operations to remove the lanthanides.

Thus, potential process problems include corrosion, addition of extra components and volumes to the waste, gassing of the resin bed, handling and disposal of contaminated waste resin, solids formation, short resin bed life, and a need to avoid high radiation levels. Storage of waste for up to ten years prior to partitioning appears mandatory if problems from high radiation levels are to be avoided.

Many of the ion-exchange techniques have not been demonstrated on a scale corresponding to process operations. Extensive scale-up development would probably be necessary. Also, it can be anticipated that highly precise process control would be required in practice.

Absorption or adsorption processes other than ion exchange in general have the same disadvantages as inorganic ion exchange. The selectivities of such absorbents or adsorbents are sometimes high, but their capacities are generally low, and they tend to be better suited to the removal of trace constituents from low ionic strength solutions than to partitioning of macro components from strongly electrolytic solutions (which is the situation for partitioning). A possible application might be the use of a solid absorbent such as activated carbon to remove iodine from either a solution or a gas stream after volatilization of iodine.

There are undoubtedly a rather large number of precipitation methods

that could be devised for partitioning of radioactive wastes. Considerable research during the Manhattan Project and for some time later was devoted to precipitation methods, both direct and carrier, for process-scale as well as analytical and research-scale separations. To a very large extent these methods have been replaced on the process scale by solvent extraction and ion-exchange methods. The reasons why this is so for large-scale processing, but not necessarily so for analytical and research applications is that recovery of the desired product is improved and waste volumes generally are smaller with extraction.

Virtually all known separations methods have been proposed and examined for the separation and/or purification of nuclear materials. Most have been considered for radioactive wastes. The methods include techniques such as: volatility, biological, flotation and electroflotation, molecular sieve filtration of metal complexes, dialysis, electrophoresis, and other electrochemical and pyrochemical methods.

All of these methods are deficient in one or more ways when considered for the present problem, although it is possible that some may have application to final separation of specific nuclides. Deficiencies for these methods (in comparison with solvent extraction and ion exchange) were not evaluated in detail during the course of this study.

It was concluded that achieving actinide element partitioning entirely by modifications to and

improved operating conditions in current and near-future Purex-type fuel reprocessing plants is not practical. Current flowsheets deliberately route the trivalent actinide elements to the first-cycle waste along with the lanthanides and other fission products. Extensive modifications would be required to separate these actinides from the total waste in a Purex process.

Some potential exists for improving U and Pu recovery in present Purex processes through modification of operating conditions and/or adding new columns, particularly if very high additional decontamination factors for these elements are not required. Coupling this approach with an adjunct facility for removal of trivalent actinides should be considered. The solvent extraction flow-sheet studied at Karlsruhe⁽²⁾ for recovering Am and Cm from high-level waste should be considered a candidate for this.

7.3 ANALYTICAL REQUIREMENTS

In practice, analytical measurements will be needed to confirm that decontamination goals required of the partitioning or fractionation process have actually been achieved. Low-level concentrations of α -emitting isotopes will have to be detected and measured with high accuracy in a strong β - γ field. Although not a requirement, these measurements should be made on-line (during partitioning operations) to avoid possible need for post-operations holdup (to make measurements) and recycle of out-of-specifications waste.

A detailed review of the analytical problems associated with a waste partitioning program was made both with respect to the general problems of sampling in heterogeneous systems and with respect to detection and measurement of specific elements of concern. This review indicated that the state-of-the-art for analytical measurements may not be equal to the requirements for partitioning, depending on the decontamination factors required. Developmental work may be required to provide the analytical measurements needed to assure control of partitioning processes and to verify whether the required separations have been accomplished. Certainly, further analytical development will be needed if high decontamination factors are required.

The presence of solids in the waste and the potential for other solids being generated during the partitioning processes present major analytical problems both from the standpoint of being able to obtain representative samples and of assuring that the analytical procedures properly account for the constituents of the solids.

In many cases, separation of the measured constituent from the bulk sample will be required. Developing required separations procedures and adapting them for on-line control will be major problems.

7.4 PARTITIONING COSTS

The costs to separate high-level waste into a long-lived and a short-lived fraction will, of course, depend on the degree of separation required, the number of elements which

must be removed from the short-lived fraction and the required purity of the long-lived fraction. A Delphi-type procedure was used to obtain an estimate of the cost to produce a short-lived fraction which will decay to negligible radioactive toxicity in about 1,000 years, i.e., removing the long-lived elements to very high decontamination factors. Several people knowledgeable in the costs of fuel reprocessing were asked to estimate the factor by which fuel reprocessing costs would be increased by the addition of facilities to accomplish this separation. The estimates ranged from a factor 1.25 to a factor of 4, with a mean factor of 2 corresponding to about \$35,000 per metric ton of fuel for partitioning.

Another cost estimate was made on separating 99% (DF=100) of the actinide elements only from high-level waste. This degree of separation is similar to that achieved in a process developed at Karlsruhe for removing Am and Cm from waste,⁽²⁾ and that process was considered in making the estimate. The volume of concentrated high-level waste to be treated per 1,000 kilograms of irradiated fuel processed is less by a factor of about seven than the volume of feed to the reprocessing plant. The basic reprocessing cost, ca. \$35,000 per 1,000 kg was reduced accordingly by using a $(x)^{1/2}$ scaling factor for the reduction in plant size. The resulting cost was further modified by comparing the number of process cycles for partitioning to the number required for fuel reprocessing. The

following total cost estimates for partitioning were derived.

	Cost/MT Fuel, \$
Actinides plus 1% of the Fission Products	10,000
Actinides less U + 1% of the Fission Products	15,000
Actinides less U + 0.1% of the Fission Products	20,000

The first case is believed to approach the need for transmutation processing (see Section 9). The latter two cases are for the major extraterrestrial disposal cases in this study (see Section 8).

7.5 RESEARCH AND DEVELOPMENT REQUIREMENTS

Data are needed on the accomplishments possible toward long-lived isotope removal and the cost involved. These will permit assessment of the technical and economic feasibility of the various approaches to managing the long-lived fraction as well as the overall economic impact on the nuclear fuel cycle. Hence, Research and Development work on partitioning (and fractionation) should not be delayed by awaiting completion of the alternative disposal studies.

The following summary of waste separation Research and Development requirements is based on the assumption that waste fractionation will not be required and that removal of actinide elements (partitioning) from the short-lived fraction to less than 1% (DF $\geq$ 100) of their concentration in fuel reprocessing plant waste will be required. The overall Research and

Development program for this basis is estimated to cost \$3 to \$5 million and to require about 5 years to complete.^(a) If further studies, as noted earlier, indicate waste fractionation or higher actinide decontamination factors are required, the Research and Development program must be modified accordingly.

It has been emphasized during these studies that solids in waste constitute a major problem in waste partitioning and that little is known about the amount and composition of such solids. A program to obtain and characterize waste comparable to that expected from fuel reprocessing plants is needed. Resolution of problems of adequate solid-liquid separation, determination of whether treatment of the solids to remove actinides is required and definition of processes for treating the solids would be part of this program.

Laboratory-scale studies are required to test conceptual flowsheets for attaining the required actinide element partitioning. Ideally these should be done with processing plant waste or process streams. However, because these are not readily available, work with simulated materials should be done initially to define

basic separations parameters; problems posed by plant-derived waste would be resolved as such wastes become available.

As conceptual flowsheets are tested and processes are developed, capital and operating cost estimates pertinent to the processes are needed both to guide the selection of a process or processes for further study and to provide cost input to other facets of waste management studies.

Pilot plant scale testing of the most promising flowsheets will be needed. This will entail facility design, construction or modification of a facility and operation of the facility to demonstrate the flowsheets, resolve problems posed by plant scale operation and provide more reliable cost data.

Development of analytical techniques should parallel flowsheet development studies. Specific analytical requirements will be defined in part by the flowsheets devised. These developments should be scheduled to permit testing of in-line analytical techniques during pilot plant testing of flowsheets.

An approximate schedule for the needed Research and Development program is shown in Figure 7.3.

a. It is assumed that the facility exists for pilot-plant testing, and costs for such a facility are not included in this estimate.

FIGURE 7.3. Schedule for Research and Development of Partitioning

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SECTION 8: EXTRATERRESTRIAL DISPOSAL

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8.0 EXTRATERRESTRIAL DISPOSAL

This study was conducted concurrently and cooperatively with a study by the National Aeronautics and Space Administration. NASA Lewis Research Center, Cleveland, Ohio, was the lead organization for the NASA study. Many NASA organizations as well as the Space Nuclear Systems Division of the Atomic Energy Commission contributed to their study. NASA has published several reports relating to the study. These are referenced or included as appendices. This study includes several direct extractions from NASA documents.

The approach to the study was to analyze space disposal of nuclear waste using existing technology as the base case and to consider advanced systems as potential improvements on the base case.⁽¹⁾

8.1 EXTRATERRESTRIAL WASTE MANAGEMENT SYSTEM

A preliminary conclusion of the study was that partitioning of the waste and shipment of only the trans-uranium elements to space is more likely to be practical than shipment of the total waste. From that initial observation, the study emphasis was on space disposal of selected, long-lived portions of the waste. However, with improvements in technology or with significantly lower disposal costs, space disposal of the total waste could become more practical.^(a)

8.1.1 Concepts Description

The basic concept of extraterrestrial disposal includes packaging waste materials in a safe manner and transporting the material by rocket or other means to a location off the earth. Several different trajectories have been considered. These include:^(b)

1. A high earth orbit on the order of 150,000 kilometers.
2. Transport to the sun.
3. A solar orbit other than that of the earth and planets.
4. Solar system escape.

Vehicles used in the analysis as current technology include existing space vehicles and the planned space shuttle and tugs. A shuttle-tug combination is illustrated in Figure 8.1. The manned space shuttle launch vehicle with an expendable external propellant tank is placed into a low-earth orbit, with lift-off assistance by two solid-fueled rocket motors. After firing, the solid-fueled motors are separated and dropped into the ocean where they are recovered. After use, the expendable external propellant tank is separated from the orbiting shuttle and then de-orbited by a small retro-rocket. The tug, with the waste payload, is deployed from the payload bay of the orbiter. For some space destinations (e.g., escape from the solar system), a second tug is required to provide the final stage propulsion; thus two

a. This point is not a NASA conclusion.

b. Placement of the waste material on an extraterrestrial body such as the moon, asteroids, and other planets was considered only briefly by NASA for reasons discussed later in this report.

FIGURE 8.1. Space Disposal System

space shuttle launches must be made for one waste package. The orbiting shuttle returns to earth at a prescribed landing site and would be ready for a repeat flight in about two weeks.

8.1.1.1 Waste Content and Disposal Priority

Space disposal is unique in that the cost of transportation is likely to be over \$2,000/kg of payload. Thus the weight of shielding creates an unusually large economic penalty. There are also unique requirements for capsule integrity to provide a reasonable degree of assurance of survival in the case of abort. The means of cooling are limited. Hence the heat content of the waste capsule is significant. The weight of the waste itself is also significant.

Because of the unusual requirements for space disposal, the determination of waste content and priority for disposal is particularly significant.

A primary consideration in establishing a priority for disposal is the potential long-term toxicity to man and the environment if the isotope is released. Many studies have been made of the relative harmfulness of different isotopes.⁽²⁻⁵⁾ A review of long-term hazards relative to these waste disposal options is included in Volume 1 of this report series. Most investigators generally conclude that the actinides are the most toxic in the long term. For this reason, this study suggests that transuranium elements (actinides minus uranium) receive first priority for extraterrestrial disposal.

Iodine-129 and technetium-99 are examples of fission product elements which may have long-lived toxicity comparable to that of the actinides. These fission products are considered by some to be almost nonradioactive, but there are others who feel that they are very toxic.

Several early investigators of space disposal or other permanent disposal methods considered primarily shipping either the fission products with high radiation levels or the bulk high-level wastes without partitioning.⁽⁶⁻⁹⁾ However, it seems reasonable to store fission products such as cesium and strontium, which have relatively short half-lives in the context of this study, in carefully designed manmade facilities which can assure no release to man's immediate environment during their lifetime. Radioactive cesium-137 and strontium-90 would decay by factors of several billion in one thousand years. Actinides as a group will decay by a factor less than 10 in one thousand years, and will be toxic for thousands of years.

Three different cases of waste content for space disposal were considered in this study:

Case 1. Package the bulk waste as it comes from the reprocessing plant in solid form. The package can include incorporation of the waste in forms such as glasses, ceramics or cermets. For this study the waste is assumed to be incorporated into borosilicate glass.

Case 2a. Remove nearly all fission products and uranium and thorium from waste, leaving only the transuranium elements for disposal. Only

1% of the fission products and 0.5% of the uranium are included with the transuranics. Transuranium elements contaminated with fission products will be in an oxide form.

Case 2b. Case 2a with 0.1% of fission products remaining with the transuranics for space disposal.

Case 3. Same as Case 2b with 99% of curium also removed from the transuranics to be disposed of into space.

A preliminary analysis of Case 1 indicated that it would have very high costs, high package integrity would be difficult to achieve, and the number of space flights would be excessive (in the thousands of flights per year in the year 2000). Therefore, this case received less detailed consideration than Case 2.

Cases 2a, 2b, and 3 would require chemical separation (partitioning) of the constituents disposed of extraterrestrially from the remaining constituents, which would require separate management. As the study progressed, Case 2a and 2b appeared to be the closest to the most practical from the standpoint of disposal into space, and these received the most emphasis for this study. In practice, the cost of additional partitioning and the additional weight which must be shipped would be balanced to achieve an optimum safety and cost.

Case 3 was considered only briefly. Curium-244 has a short half-life and provides up to 87% of the radioactivity and 93% of the heat in the LWR actinide waste. Thus removal of the curium could reduce the space package weight significantly. If the curium

were removed, the curium would probably be stored on earth for around 100 years until it decays to plutonium and other daughters. These longer-lived daughters with much reduced heat output and radioactivity could then be shipped to space separately at significantly lower cost.

For space disposal purposes the waste is assumed to be held for 10 years after reprocessing. This is primarily to allow decay of the hottest isotopes. In practice an optimum time would be arrived at based on

separations costs, encapsulation requirements, and space flight costs.

The weights of actinides and fission products pertinent to space disposal are shown for several reactor types in Table 8.1. The quantity of irradiated fuel sent to the reprocessing plants is summarized in Table 8.2. Based upon these values, quantities of transuranics for disposal and the space flight requirements are shown in Table 8.3 for several cases.

TABLE 8.1. Quantity of Actinides in Waste from Reprocessing Various Reactor Fuels 10-Year Decay

Reactor Type	Total Actinides Gms/MT Fuel Charged	Uranium + Th Gms/MT Fuel Charged	Actinides Less U+Th Gms/MT Fuel Charged	Curium Gms/MT Fuel Charged	Fission Products, Gms/MT Fuel Charged
PWR-U	5,480	4,774	706	23	34,400
PWR-Pu	6,980	4,685	2,295	472	35,100
HTGR, with Recycle Makeup	15,700	11,740	3,960	13	99,000
LMFBR-AI	6,360	4,327	2,033	12	38,100
LMFBR-GE	6,030	4,407	1,623	19	42,600

TABLE 8.2. Projected Quantities of Uranium, Plutonium and Thorium in the Spent Fuel Shipped to the Reprocessing Plant⁽¹⁰⁾

Metric Tons/Year						
Year	LWR-U	LWR-Pu	HTGR	AIF0	GEFO	Total
1980	2,388.6	268.9	5.8	0.0	0.0	2,663.3
1990	6,959.2	278.5	874.5	285.1	11.9	8,409.1
2000	4,845.2	0.0	2,754.2	446.9	6,804.5	14,850.8
2005	3,842.9	0.0	3,604.6	351.4	13,555.3	21,354.2

TABLE 8.3. Example Waste Transuranic Quantities and Mission Requirements for Space Disposal

Basis: Waste held 10 years before space disposal

Disposal Case	Year of Space Disposal								
	1990			2000			2010		
	Transuranics Shipped Per Year, Kg	No. of Space Flights Per Yr	No. of Shuttle Launches Per Yr	Transuranics Shipped Per Year, Kg	No. of Space Flights Per Yr	No. of Shuttle Launches Per Yr	Transuranics Shipped Per Year, Kg	No. of Space Flights Per Yr	No. of Shuttle Launches Per Yr
<u>Earth or Solar Orbit</u>									
Case 1. Total Waste ^(a)	--	700	700	--	2,500	2,500	--	6,000	6,000
Case 2a (288 Kg Transuranics/mission)	2,315	8	8	9,600	33	33	26,200	91	91
Case 2b (447 Kg Transuranic/mission)	2,315	5	5	9,600	21	21	26,200	59	59
<u>Solar Escape</u>									
Case 1 Total Waste ^(a)	--	1,800	3,600	--	6,500	13,000	--	15,600	31,200
Case 2a (113 Kg Transuranics/mission)	2,315	24	48	9,600	85	170	26,200	232	464
Case 2b (191 Kg Transuranics/mission)	2,315	12	24	9,600	50	100	26,200	137	274

a. Based on NASA TMX-2911, Feasibility of Space Disposal of Radioactive Nuclear Waste, December 1973. This is a different base than that used in the rest of the table. These numbers are used here only to indicate the order of difference with total waste and transuranics.

The actual quantities which might be considered for space disposal will vary with developments in the fuel cycle. Since parts of the waste stream are elements which have not been recovered because it was not economical to do so, the development of improved partitioning processes or the development of new uses for radioisotopes could reduce the quantities which are presently considered to be waste. Quantities discussed herein should therefore be considered only as a very preliminary example.

8.1.1.2 Waste Capsule

Encapsulation of the waste constituents is needed to prevent release of radioactive materials in event of a flight abort and to prevent harm to life. Containment is needed during surface transportation, during any phase of flight or abort, and during some lifetime in flight. Consideration must also be given for removal of decay heat, criticality control (for cases with actinides only), and for minimizing weight.

Based upon these considerations, the design of the capsule for total waste is shown in Figure 8.2 and for

FIGURE 8.2. Total Waste Capsule for Space Disposal

actinides only in Figure 8.3. For total waste, the borosilicate glass containing the bulk waste oxides is encapsulated in an inner storage container of stainless steel. An outer container of stainless steel is provided for impact resistance. Depleted uranium was used as shielding material between the two containers. (However, it has been noted that the depleted uranium may be unsatisfactory at the high temperature involved.)

The conceptual capsule design for shipment of the transuranics (Figure 8.3) includes transuranic oxide particles (with some fission product contamination) in the form of small spheres approximately 3.3 millimeters in diameter. These oxide spheres are encapsulated in tungsten to provide a capsule with high temperature capability. A void space is left around the sphere to allow for helium buildup as the radioactive material decays. The tungsten is coated with an oxidation-resistant coating such as aluminum oxide so

that the small capsule itself can withstand exposure to a high-temperature, oxygen-containing atmosphere.

These micro-capsules are mixed in a matrix of lithium hydride for shielding and aluminum or copper for thermal conductivity. This matrix material is compacted in the form of a sphere. The sphere, which consists of small encapsulated particles in a lithium hydride-aluminum matrix, is surrounded with additional lithium hydride and tungsten for shielding and is then encapsulated in stainless steel for impact resistance.

For protection in event of re-entry, an additional shell is added to the exterior of the waste capsule. A conceptual reentry shield, which surrounds the waste capsule, was designed to ensure stability and minimize the weight penalty and is shown in Figure 8.4.

8.1.1.3 Space Flight

The potential space destinations considered are listed in Table 8.4, along with the incremental velocity (ΔV) beyond low-earth-orbit velocity.

The conceptual launch systems for several destinations are shown in Figure 8.5. The first step is propulsion of the launch vehicle (a manned space shuttle) into a low circular parking orbit of about 370 kilometers. From this orbit, the upper stage or stages (Tugs) inject the waste package into its final destination. For solar or high-earth orbits, a single

FIGURE 8.3 Transuranic Waste Capsule for Space Disposal

FIGURE 8.4. Re-entry Shield and Transuranic Disposal Package for Solar Escape Destination

TABLE 8.4. Summary of Potential Space Destinations

	Delta V, (a) km/sec
High-Earth Orbit	4.11
Solar Orbits Via: Single Burn	
Beyond Earth Escape	3.65
Circular Solar Orbit	4.11
Venus or Mars Swingby	4.11
Solar System Escape: Direct	8.75
Via Jupiter Swingby	7.01
Solar Impact: Direct	24.08
Via Jupiter Swingby	7.62

a. Delta V is the sum of the velocity increments required beyond that of reaching low earth orbit.

tug which requires two fuel burns is used for each mission. For solar escape, two Tugs are required for each mission. Solar impact is not shown in Figure 8.5. Direct solar impact is not possible with these vehicles. However, solar impact using a swingby of Jupiter could be done using a single propulsion phase from the low-earth orbit. Similarly, swingbys of planets could be used for solar orbits or solar system escape. However, proper guidance and controls for all swingby missions would be most difficult to assure. The fact that missions utilizing swingby of other planets can be accomplished only at intervals of many months also makes utilization of such systems difficult.

The NASA studies indicate that transport of waste constituents to the sun or other extraterrestrial bodies (e.g., the moon or other planets) are technically feasible. However, these destinations were considered only briefly. (See Section 8.2.4.1).

8.1.2 Systems Requirements

System requirements for the space disposal concept are summarized in Figure 8.6. As illustrated in this figure, high-level liquid wastes would be conditioned, specifically by densification and encapsulation as solids, transported to the launch site and flown to a final trajectory.

8.1.2.1 Waste Treatment and Handling

The general system flow diagram shown in Figure 8.6 starts with high-level radioactive liquid waste from the reprocessing plant. The waste may be conditioned and stored retrievably for some period of time. Chemical adjustment (such as neutralization, which would not be expected in this case) and storage as liquid in tanks for a 5 to 10 year cooling time or encapsulation and interim storage are potential examples of these steps. The waste may then be chemically separated into an actinide or transuranium stream and one or more fission product streams. For space disposal the separation into transuranic streams is most likely. A further separation of the curium from the transuranic stream could also be practiced to reduce the heat and radioactivity and weight of the package being disposed of in space. In addition, it is possible that long-lived fission products could be separated from the waste stream and included for space disposal. The waste constituents to be transported to space would then be encapsulated into high integrity packages in preparation for transportation to the launch site.

FIGURE 8.5. Launch Systems for Solar Orbit, High Earth Orbits and Solar Escape

8.1.2.2 Site Preparation

It is assumed that existing Cape Kennedy facilities have the capacity and could be utilized for launch until about the year 2000. Some additions to the facilities would be required in the way of waste package handling, launch pads, and other buildings. Shortly beyond the year

2000, major additional launch facilities would likely be required to accommodate the higher capacities.

8.1.2.3 Transportation to the Site

Transportation from the reprocessing plant to the launch site will be in special railroad cars, trucks,

FIGURE 8.6. System Requirements for Managing High-Level Radioactive Waste by Extraterrestrial Disposal

or possibly barges. Because of the stringent packaging requirements for space transportation, additional packaging requirements for the land transportation from the reprocessing and encapsulation site to the launch site are expected to be relatively minor. Some minor additional shielding and special transportation restraints will be needed.

8.1.2.4 Operation of the Site

Operations at the site will consist primarily of unloading encapsulated material from land transportation vehicles in modest hot cell type of facilities, and assembling it in the space vehicles. Because the material will be encapsulated to survive launch pad accidents or re-entry from outer space and still maintain a radiation exposure of less than 1 rem at 1 meter from the surface, remote handling facilities will be minimal. Prior assembly, positioning, and fueling of the rockets will have been done. Final countdown and typical launch preparation procedures will be required. The manned shuttle and its appurtenances will be launched into a low-earth orbit, the payload bay will be opened and the tug with the waste capsule will be launched and programmed toward the final destination. The shuttle will then be returned to earth for re-use.

8.1.2.5 Final Monitoring

Final monitoring would involve position monitoring of each tug mission to assure proper final destination. This would require up to a few years for each mission. In addition, continuous monitoring would be

required on the contamination of space from failed capsules. Two trajectories seem most likely to assure true final disposal: impact with the sun (which is not likely with present vehicles) and escape from the solar system. Should there ever be any return of waste constituents from these two trajectories, the dilution of the waste constituents which might intercept the earth is expected to be so great that it would be undetectable by present day techniques. However, even with these disposal locations, continuous monitoring of the high-earth atmosphere for the next few hundred years may be desirable.

8.2 TECHNICAL FEASIBILITY

Key considerations in the technical feasibility of extraterrestrial disposal are: providing a reliable space flight system to assure that the waste will reach and remain at its proper destination; providing a high-integrity capsule to assure nonfailure during required residence times in space and to assure minimum spread of radionuclides in event of an abort; providing adequate provisions for cooling, shielding, and criticality control; and minimizing weight to keep the costs practical. Most of these considerations have been discussed briefly in the preceding descriptions but are covered in more detail in this section as they relate to technical feasibility.

8.2.1 Capsule Design Criteria

The basic design criteria used in the study for the above-described design factors are described in the following sections.

Radiation Shielding

A general criterion for radiation shielding was established that the capsule at all times must have radiation levels less than 1 rem per hr at 1 meter from the surface. Higher dose rates than the 1 rem at 1 meter were considered in some cases to determine the effect of higher dose rates on costs.

For shielding calculations used, it was assumed that the capsule shape is retained after impact following a postulated abort. For example, if a spherical capsule containing actinide waste impacts the earth, it is assumed that the capsule will retain its spherical shape and the resultant radiation level at 1 meter would not increase. In fact, it is likely that under some circumstances the capsule will be grossly deformed and actual dose rates could be somewhat different. Shielding calculations on distorted capsules are beyond the scope of this study but should be examined before space disposal is used.

Temperature

Temperatures within the capsule, from the center-line to the edge, should be sufficiently low that materials in the capsule will not decompose or react excessively with each other to result in no adverse effects on the capsule integrity. This stability is necessary in outer space, in re-entry circumstances, in a vacuum in the shuttle bay, and for accidental burial in earth or residence in a fire.

Consideration was given to establishing conditions such that the capsule be sufficiently cool that it can

be touched if it lands accidentally in an unknown location. However, it was generally agreed that the potential for severe harm to people touching a thermally hot capsule would be very low. Hence, this criterion was not used.

Containment

• Ground Transportation

A typical accident during ground transportation should not result in a serious hazard because a capsule designed to withstand flight abort and re-entry from outer space should withstand virtually any conceivable conditions during an earth accident. While the criterion of 1 rem per hour at 1 meter from the surface is consistent with the Department of Transportation regulation⁽¹¹⁾ for exclusive use vehicles of 1000 millirem per hour at 3 feet from the external package surface, some minor vehicle design features may be necessary to meet the requirements for radiation levels external to the vehicle and in positions of the vehicle which are occupied by persons.

• Flight Aborts

Launch Pad. The capsule should be able to successfully resist overpressure, high velocity fragments, fireball heat, impact, and residual fire of a launch pad accident.

Pre-orbital Abort. The capsule should be able to survive intact an accident during launch and prior to achieving orbital velocity. In general, a capsule which will withstand abort from orbit will withstand any condition that can occur between launch and orbit.

Abort from Orbit. The capsule should be able to survive re-entry

at a velocity of 11 km/sec with subsequent impact on earth.⁽¹²⁾ The impact conditions assume a hard granite surface at terminal velocity of the order of 300 meters per second. Elevated temperature must be assumed as a result of re-entry. Obviously, rough impact surfaces may be encountered.

Following impact, the capsule should be able to withstand burial in the deepest ocean trenches or burial in a variety of soils for reasonable time periods. A capsule landing in relatively soft sand is likely to be buried to depths beyond 6 meters. Hence, it may be very well insulated. If a capsule lands in the ocean, it is generally assumed that it will be recovered some day. However, there are real probabilities that it can never be found. Hence the ability to withstand seawater and the seabed environment for long time periods is highly desirable.

Long-Time Residence in Space

The necessary lifetime of the capsule during flight depends largely on the destination. If the destination is direct solar impact, then a containment lifetime of only a few months and a shielding lifetime only long enough to be removed from potential exposure to man may be all that is necessary. If the destination is a storage type of orbit, then a capsule lifetime of hundreds or thousands of years may be needed. If the

destination is solar escape, a lifetime of millions of years could be desired if no contamination of the universe is allowed, or tens of years if contamination of the universe is of no major concern. Thus capsule lifetime needs can vary greatly depending on the circumstances. At the present time, capsule design assuming a lifetime of a few hundred years may be the best that can be achieved.

Criticality

Criticality should not be reached under any foreseeable circumstances. These include loss of neutron-absorbing materials and dissolution of the capsule in water. Such considerations must take into account the fact that many of the actinides have very low critical masses.

Weight

Limitations of current rocketry in carrying payloads to certain destinations were used as the basis for sizing maximum capsule weights.

8.2.2 Capsule Design Details

Conceptual capsule designs for cases involving space disposal of total waste and transuranics only are presented below.

Capsule Design - Total Waste - Case 1^(a)

A conceptual design was developed by NASA Lewis⁽¹³⁾ for shipment of

a. Shielding calculations were based on fission products only.

total waste. The design was based on the incorporation of total waste oxides in mixtures of 30 mole% in borosilicate glass.

Consideration was given early in the analysis for all cases to reducing the shielding weight required. Possible methods considered were shadow shielding and reusable shielding.

With shadow shielding, areas in line with personnel or radiation sensitive components are more heavily shielded than those areas in which radiation would not have harmful effects. As an example, a waste container might have heavy shielding on the side next to the space vehicle and light shielding on the side exposed to space. The capsule would be transported on earth in shielded containers and installed in the space vehicle with special equipment and procedures. Such a system would be useful if successful flight could be assured. However, the potential hazards of such a container in event of unsuccessful flight indicated the need for extensive analysis beyond the scope of this study and was not considered further.

With disposable shielding, the package would be heavily shielded until some point early in the flight path where the heavy shielding would be separated. Only lighter shielding would be accelerated to the velocity necessary for disposal. For example, full shielding might be carried by a space shuttle to low earth orbit. The tug or tugs which accelerate the container to a subsequent trajectory would carry only a lightly shielded

package. The heavy shielding would remain with the shuttle and be returned to earth. The heavy shielding could be reusable. This system could require much less total energy for disposal. However, this concept was not considered further for the same reasons given for shadow shielding.

Calculations were performed by the NASA staff on containment vessels and shielding for total waste which would provide dose rates of 1 rem/hr, 10 rem/hr, and 500 rem/hr at 3 meters from the container centerline. In these calculations it was assumed that only the fission products contributed to the radiation dose. Although some differences would exist with actinides included, the differences should be unimportant. The NASA analysis of this capsule is included as Appendix B.

The container design was illustrated in Figure 8.2. The ability of this container to provide the impact resistance necessary for containment during any strenuous abort conditions is untested.

The NASA analysis concluded that the space transportation cost alone for disposal of the above containers shielded to 1 rem per hour at 3 meters from the surface would add to the cost of electricity at the bus bar approximately 4 mills per kW-hr for earth escape and 28 mills per kW-hr for solar escape. Increasing the dose rate at 3 meters from the outer surface of the package from 1 to 500 rem/hr would result in a factor of 3 reduction in cost.

The above costs were high and space flight requirements were extreme. The cost of providing greater package integrity than the simple cylinder would add still further to the complexity and cost. For these reasons the principal effort was applied to Case 2, transuranics in waste only.

Capsule Design - Transuranics only, with Fission Product Contamination - Case 2

Extensive developmental work on the use of isotopes in space and on aerospace nuclear safety is applicable to this case.

A preliminary capsule design for shipment of the transuranics in waste was shown in Figure 8.3. Analysis and testing of containers of this type have been done as a part of programs for the utilization of radioisotopes in space and aircraft reactor programs.⁽¹⁴⁻³¹⁾ An analysis of a capsule design was made by the staff at NASA Lewis. The following (up to "Criticality Considerations") is extracted completely from their report.⁽¹²⁾ Slight modifications have been made to conform to the format of this report.

Encapsulation of the transuranics with inclusion of small percentages (0.1 to 1%) of the fission products was studied in some detail to provide a conceptual design for determining the feasibility of the approach.

There are some differences in waste content as defined in the NASA studies and in the balance of this report. However, the differences are

minor and have no effect on the determination of feasibility of the process.

The amount of transuranic waste relative to the matrix was varied parametrically to obtain the maximum amount of transuranic waste products in a payload without exceeding the limiting temperature in the matrix.⁽³²⁾ The optimum transuranic waste content was approximately 8 to 10 volume percent of the matrix to maintain the temperature of the matrix below the prescribed temperature limit of 860°K. The temperature limit was established primarily by the elevated temperature characteristics of lithium hydride.

- Radiation Shielding

Shielding for the transuranic waste is required to reduce the external dose rate to acceptable levels. These levels are based upon acceptable levels for handling, acceptable levels for transporting in a manned shuttle, or accidental exposure levels to the general public. The value of 1 rem/hr at 1 meter from the external surface of the package was chosen as the base point (based on 10 CFR 71^[33] for transporting of radioactive waste) for accidental exposure to the general public. Effects of extending the dose rate to 100 rem/hr were examined for this study.

The shielding for gamma, neutron, beta and alpha radiation was examined for a weight optimization study.⁽³²⁾ Based on these calculations, a single layer of tungsten and a layer of LiH were used for capsule shielding. The

calculations showed that the main source to be shielded was the gamma radiation from the fission products and not the neutron or the alpha radiation from the actinides.

To minimize the shield weight, which is the major portion of the total payload weight, a spherical geometry was chosen for the waste capsule. The layer adjacent to the matrix material containing the actinides is composed of high-density gamma shielding material (tungsten). For safety purposes, in the event of a break in the outer vessels, a layer of stainless steel was added to the outside of the tungsten to prevent oxidation of the tungsten. The next and last shielding layer is the neutron shielding, or lithium hydride as selected for the study. The layers of material external to these were not considered as part of the shielding analyses.

- Impact Protection

The primary impact protection for the transuranic waste package is a spherical shell on the outside of the lithium hydride. The spherical shell selected, based on experiments and analyses, was 1.58 centimeters (5/8 inches) of 304 stainless steel. This was backed up by an additional 0.95 centimeter shell of 304 stainless steel between the lithium hydride and the tungsten shields.

To prevent any free hydrogen that could be present in the lithium hydride from reaching the outer stainless steel containment shell, a 0.127-millimeter (0.005 inch) layer of tungsten was assumed to be deposited on the inside of the stainless

steel shell. This should prevent hydrogen from diffusing into the stainless steel shell.

- Re-entry Shell

For re-entry protection, a re-entry shell must be added to the exterior of the transuranic waste capsule. The stable configuration selected to minimize the weight penalty was shown in Figure 8.4.

In re-entry, the heating rates on the package vary depending on velocity, angle and atmospheric density. At low heating rates the convective heat transfer away from the shell is most important, for which materials such as graphite function well. Appollo re-entries are an example of convective heating rates. For high velocities such as can be encountered in planetary entry following earth-escape velocities at steep angles, the radiative heating rate dominates and a reflective type of re-entry shell material is required.

One such material that has high reflective capability with multiple reflective sites for scattering and reflecting heat is a composite made of quartz fibers woven into a mat, similar to fiberglass, with a silica binder added. This material proposed by the staff at NASA/Ames Research Center results in a very good reflective barrier for the re-entry shell. Some of the thermophysical properties are:

Density	2.5 g/cm ³
Specific Heat	1.15 Joule/g-°K
Thermal Conductivity	2.5 x 10 ⁻² Joule/sec-cm-°K
Thermal Expansion Coefficient	5.6 x 10 ⁻⁷ /°K

This layer of composite silica fibers must be backed with a thin silver film followed by graphite to act as the re-entry heat shield in the slower re-entry mode. A pictorial description of these layers is presented in Figure 8.7. The insulation is added primarily to protect the stainless steel containment vessel from heating during the re-entry or in the event of a launch pad fire. On the back side of the re-entry shield, the thicknesses are reduced and the insulation has been removed to allow for the waste heat from the actinides and fission products to be conducted and radiated away. The re-entry shell weights represent approximately 13% of the total package weight.

- Single and Multiple Re-entry Packages

While the nuclear waste package is in the shuttle bay, the total package after having been precooled before launch, slowly increases in temperature. After separation from the low-earth orbiting shuttle, the waste package will come to an equilibrium temperature, based on the internal heat source (transuranics and fission products), the thermal conductivity through the layers of material and the heat sink temperature in space.

Steady-state temperature calculations were conducted for various sizes of waste packages. Using the temperature limit for the matrix material, the minimum number of packages per total payload was determined.

FIGURE 8.7. Typical Front Re-entry Shield Composite for Radioactive Waste Package

The temperature limit on the matrix would be exceeded for the total payload in a single sphere for the high earth orbit package with 0.1% fission products. Therefore this payload would have to be divided into two or more packages. For other Case 2 waste conditions, the design can use a single package per payload unless a safety assessment indicates multiple packaging is an improvement.

- Overall Package Configuration

An overall configuration was established using the previously-derived re-entry shield, impact protection and shielding against radioactivity. For high earth orbits the payload with transuranics and 0.1% fission products was divided into three equal packages, each with its own impact and re-entry protection. The largest single package has a diameter of 2.8 meters, with a 1.37-meter diameter stainless steel sphere containing the nuclear shielding and the waste-containing matrix. This package weighing 8400 kilograms contains 384 kilograms of actinides plus 134 kilograms of fission products. Other data for all the actinide packages considered are found in Table 8.5.

- Weight

The payload capabilities, assuming a shuttle as the basic launch vehicle, used for the basis of the package designs were 8480 kilograms for high earth orbit or solar orbit missions and 3270 kilograms for the solar system escape missions. These payloads may be designed as single waste packages or as multiple waste packages, each with its own re-entry shell.

- Total Packaging Weight Ratio Versus External Dose Rate

The above discussion and design has been primarily for an external dose rate of 1 rem/hr at 1 meter from the surface of the impact shell. If this dose rate could be increased (subject to safety assessment and acceptance), the amount of waste constituents could be increased per launch. The effect of allowable dose rate on the packaging weight ratio (total weight: waste material weight) is shown in Figure 8.8.

The choice of destination does not substantially change the package weight ratio. The greatest effect on the weight ratio is the reduced shielding for the higher dose rates. The predominant gain is obtained by raising the allowable dose rate to 10 rem/hr. Further increases to higher allowable dose rates are less effective in increasing the package ratio.

Capsule Design - Transuranics Only, with Fission Product Contamination, with Most of Curium Removed - Case 3

The amount of curium in the transuranic waste from several different reactor types is summarized in Table 8.6. Up to 93% of the heat and 87% of the radioactivity of the actinides is associated with the relatively short-lived curium from LWR's. However, curium content in LMFBR waste transuranics is much less than in LWR waste, and the curium content in HTGR waste is less than 6% of the total.

The removal of curium neutron radiation from the package permits a reduction in the amount of lithium

TABLE 8.5 Summary Data on Package Configuration for Transuranic Disposal to Space

DESTINATION	HIGH EARTH ORBIT & SOLAR ORBIT				SOLAR ESCAPE	
% Fiss. Products Included with Transuranics	0.1	1.0	0.1	1.0	0.1	1.0
Number of Packages/Launch	1	1	3	3	1	1
Dimensions of Package:						
Outside Diameter, meters	2.896	2.896	1.761	1.761	1.81	1.81
Thickness of (SS Shell), cm	0.1	0.1	0.1	0.1	0.1	0.1
Thickness of (Silica-Front), cm	4.0	4.0	4.0	4.0	4.0	4.0
Thickness of (Silica-Rear), cm	1.0	1.0	1.0	1.0	1.0	1.0
Thickness of (Silver Reflector), cm	0.01	0.01	0.01	0.01	0.01	0.01
Thickness of (Graphite-Front), cm	1.0	1.0	1.0	1.0	1.0	1.0
Thickness of (Graphite-Rear), cm	0.5	0.5	0.5	0.5	0.5	0.5
Thickness of (Insulation-Front), cm	2.0	2.0	2.0	2.0	2.0	2.0
O.D. Of Impact, Sphere, meters	1.43	1.37	0.973	0.923	1.036	0.98
Thickness of (SS-Impact Shell), cm	2.54	2.54	2.54	2.54	2.54	2.54
Thickness of (LIH Shield), cm	12.65	11.74	9.83	8.28	10.51	8.75
Thickness of (W Shield), cm	3.83	5.54	3.61	4.84	3.60	4.92
Off-Set of Internal Sphere Forward of Re-Entry Shield Center, cm	6.7	6.3	3.7	3.5	3.9	3.7
Weight of Package/Total Payload, KG	8400	8400	2800/8400	2800/8400	3270	3270
Weight of Transuranics Per Package, KG	634	384	149	96	191	113
Weight of Fiss. Products Per Package, KG	22	134	5.2	34	6.7	40
Weight of Re-Entry Shield Per Package, KG	1038	1038	370	370	415	415
Weight of Impact Vessel Per Package, KG	1295	1122	565	504	640	567
Weight of LIH Shield Per Package, KG	455	429	168	113	178	135
Weight of Tungsten Shield Per Package, KG	2780	3580	1032	1249	1190	1480
Weight of Matrix Per Package, KG	2080	1655	498	415	625	505
Dose Rate at 1 meter from Surface, R/hr	1	1	1	1	1	1
Number of Curies from Transuranics/Pkg ^(a)	1.52×10^6	0.92×10^6	0.356×10^6	0.23×10^6	0.46×10^6	0.26×10^6
Number of Curies from Fiss. Prod./Pkg	0.20×10^6	1.20×10^6	0.045×10^6	0.30×10^6	0.06×10^6	0.37×10^6
Thermal Power, KW/Package	44.0	30.12	10.36	7.59	13.25	9.26

a. Based on transuranics from LWRs.

Bases: Minimum number of packages
 Solar system escape mission
 2 shuttle launches per mission
 3270 kilograms total payload per mission

FIGURE 8.8 Total Package Weight Ratio for Different Exterior Dose Rates

TABLE 8.6 Thermal Power and Radioactivity of Transuranics in 10-Year-Old Waste

	LWR-U		LWR-Pu		HTGR		LMFBR-AI		LMFBR-GE	
	Thermal Power(a)	Radio-activity(b)	Thermal Power(a)	Radio-activity(b)	Thermal Power(a)	Radio-activity(b)	Thermal Power(a)	Radio-activity(b)	Thermal Power(a)	Radio-activity(b)
Total Actinides Less U	69.9	2,350	1,230	36,900	617	25,000	169	7,140	141	5,530
Curium	60.4	1,727	1,144	32,617	36.7	1,051	41.3	959.1	57.5	1,633
Percent of Total in Curium	85	73	93	89	6	4	24	13	41	30

a. Thermal power is in watts/MT of U + Th.
 b. Radioactivity is in curies/MT of U + Th.

and other shielding. In addition, the reduced heat load permits a reduction in the requirement for copper or aluminum heat transfer material and a reduction in the size of the thermal radiation surface required.

Thus, the volume, radius, and weight of the capsule can be significantly reduced. A detailed analysis has not been made of this case. However, it is expected that a reduction in space flight costs of 50% or more is within reason.

Materials Considerations (34)

A great variety of material combinations might be used for waste encapsulation. Some of the considerations are discussed below for possible future detailed evaluation.

Transuranics could be compounded in the form of nitrides or some other relatively stable compound which would be less subject to the α, n reaction encountered with oxygen-18 when the transuranics are in the form of oxides. However, transuranic oxides generally are more stable than nitrides or carbides. The α, n reactions could be reduced by the depletion of oxygen-18.

Lithium hydride is one of the best neutron attenuators per unit of weight because of the relatively light weight of the lithium and hydrogen atoms. However, other materials which are more chemically stable may be desirable. Samarium hydride has been considered because it reportedly has higher temperature stability and provides a favorable attenuator for neutrons.

A primary container of a material such as tungsten or molybdenum around the lithium hydride is desirable for hydrogen containment. At even slightly elevated temperatures there is a partial pressure of hydrogen over almost any hydride material. The hydrogen would probably diffuse out of a capsule of only stainless steel, thereby embrittling the stainless steel. For lifetimes of a few months or a year, this might be acceptable. For longer lifetimes, it could be a serious problem. Molybdenum is also likely to be more compatible with lithium and with lithium

hydride and less subject to corrosion from either of these materials than stainless steel.

Inconel or some other material of higher strength may also be considered as an alternative to the stainless steel casing for impact resistance.

Criticality Considerations (35)

Certain of the actinides produced in reactors during nuclear fission are fissionable. When these actinides are separated from the spent fuel and concentrated, they must be carefully controlled to avoid criticality.

An analysis was made of one container system for nuclear criticality safety. The principal conclusion is that an infinite amount of the material mix in Table 8.7 in the capsule configuration is well subcritical.

The criticality analysis was made on a container consisting of four concentric shells surrounding a sphere filled with actinide oxides embedded in a lithium-aluminum hydride matrix. The dimensions and compositions of the four spherical shells surrounding this center sphere are given in Table 8.8.

The estimated fraction of a minimum critical mass represented by each nuclide in one sphere is shown in Table 8.9. The table points out only that there is sufficient total fissionable material present for criticality if conditions are favorable. These estimates do not represent actual conditions since in each instance, spherical geometry and optimum water moderation are assumed.

TABLE 8.7. Composition of Transuranic Waste in Space Disposal Capsule for Criticality Analysis

<u>Material</u>	<u>Atoms/cc x 10⁻²¹</u>	<u>g/cc</u>	<u>Total g in Single Sphere</u>
Li-6	11.2	0.1120	6,325
Li-7	13.8	0.1610	9,092
Cu	18.9	1.9900	112,376
O	4.06	0.1080	6,099
Al	13.5	0.6050	34,164
H	25.0	0.0420	2,360
Np-237	1.57	0.6180	34,898
Pu-238	0.0124	0.0049	276
Pu-239	0.0552	0.0219	1,237
Pu-240	0.0409	0.0163	920
Pu-241	0.00600	0.0024	135
Pu-242	0.00391	0.0016	88
Am-241	0.112	0.0448	2,530
Am-243	0.185	0.0746	4,213
Cm-244	0.0440	0.0178	1,005

Note: Sphere volume = 56.6 liters

TABLE 8.8 Materials and Dimensions of Space Disposal Capsule for Criticality Analysis

<u>Shell</u>	<u>Material</u>	<u>Density g/cm³</u>	<u>Radius cm</u>	<u>Thickness cm</u>
--	Transuranics + Matrix	--	23.8	--
1	Tungsten	19.3	27.02	3.22
2	LiH	0.82	34.62	7.60
3	Stainless Steel	7.93	37.16	2.54
4	Carbon	2.1	38.16	1.00

TABLE 8.9 Estimated Fraction of a Minimum Critical Mass Represented by Each Transuranic Nuclide in One Sphere

<u>Nuclide</u>	<u>Approximate Amount in One Sphere, Kg</u>	<u>Minimum Critical Mass, Kg</u>	<u>Fraction of Minimum Critical Mass</u>
Np-237	34.9	43.1	0.8
Pu-238	0.3	4.5	<0.1
Pu-239	1.2	1.7 ^(b)	0.7
Pu-240	0.9	96.4	<0.1
Pu-241	0.14	0.26	0.5
Am-241	2.5	71.4	<0.1
Cm-244	4.2	14.2	0.3

a. Each estimate is based on most severe conditions.

b. Based on 30% Pu-240

With the lithium-aluminum treated as voids, the neutron multiplication factor, k_{∞} for the material described in Table 8.8 is 0.3. Since lithium is a neutron absorber, k_{∞} for the material would actually be less than 0.3. Consequently, an infinite amount of this material would be well subcritical.

The nuclear criticality safety analysis has been limited to the actinide waste package as designed. Possible accident conditions that could affect the nuclear criticality of the package during the fabrication, launch, or post launch were not considered for this study.

8.2.3 Encapsulation Processes

Proper encapsulation of the waste for space disposal is highly important and complex. Processes for achieving the encapsulation were reviewed, and summarized in this section.

The encapsulation process for containers for total waste, Case 1, consists primarily of welding existing containers inside secondary containers for shielding and impact resistance. For reasons given earlier, this case was not considered in detail; hence, a conceptual encapsulation process was not developed for this container.

The encapsulation process contemplated for transuranic waste is relatively complex to minimize weight and to provide a high degree of containment reliability. Encapsulation processes have been developed extensively for radioactive isotopic heat sources.⁽³⁶⁻⁴¹⁾ Steps in the transuranic oxide encapsulation process are shown in Figure 8.9. The process steps illustrated are "state of the art" and have been used for the preparation of cermetts of plutonium oxide, promethium oxide, and other radioactive materials.

The steps of the process are described briefly below:

1. Purification or Treatment.

This step might be the last one in the separation process or the first step in the encapsulation process. It consists of the processing necessary to ensure stability of the final product and container. It includes such activities as vacuum outgassing of the powdered oxide at high temperature to remove impurities such as moisture, trace chemicals, etc.

2. Powder consolidation. Transuranic oxide powders are pressed to small billets. This step provides an initial densification and puts the material into a form from which it can be crushed for sizing before final form making.

3. Crushing. The powder billets from Step 2 are crushed in much the same way that large rocks are broken up into gravel. This step further refines particles for sizing into the desired range.

4. Sizing. The particles from Step 3 are sized by passing them through different sizes of sieves.

5. Sintering. The particles from Step 4 are sintered to convert them to the final solid dense material. If desired, they may be passed through a plasma arc to make them spherical and further increase their density.

6. Recycle. Particles which have not attained proper size in the initial sizing are recycled back through the consolidation operation.

7. Coating Material Treatment. Materials to be used for coating are purified as necessary.

8. Sacrificial Coating of Magnesium. A thin coating of magnesium is applied to the actinide oxide particles by sputtering. This coating, when later removed, provides for a void in each final coated particle. Particles are placed on a tray and exposed to sputtered materials.

9. First Phase Tungsten Coating by Thermal Decomposition. Particles are thinly coated by thermal decomposition of tungsten hexafluoride in a fluidized bed. This coating provides a base for the subsequent coating after removal of the sacrificial magnesium.

10. Sacrificial Material Removal. The magnesium coating is removed by placing the particles in a high temperature vacuum furnace to evaporate the magnesium. This process leaves a void inside of the initial tungsten shell.

11. Decontamination. Coated particles are decontaminated by chemical treatment which may consist of etching in nitric acid, neutralizing in a stop bath, rinsing and subsequently drying in vacuum. Decontamination is required to effect final tungsten coating.

FIGURE 8.9 Steps in Transuranic Oxide Encapsulation Process for Space Disposal

12. Second Phase Tungsten Coating. The tungsten shell is finished by providing a heavy coating of tungsten by thermal decomposition of tungsten hexafluoride.

13. Aluminum Oxide Coating. A protective coating of aluminum oxide is provided by flame spraying.

14. Lithium Hydride Treatment. Lithium hydride is treated by chemical preparation and particle sizing.

15. Aluminum Treatment. Aluminum powders are treated by high temperature vacuum outgassing to remove impurities.

16. Lithium Hydride-Aluminum-Actinide Oxide Particle Mixing. The coated actinide particles are mixed with particles of lithium hydride and aluminum powder to provide a uniform dispersion.

17. Compacting. The powders are compacted into a solid mass by pressing at high pressure and subsequently sintering.

18. Cutting. The outer capsule for shielding and containment is formed by conventional means. The first step is cutting to size.

19. Forming. Forming of capsule halves is accomplished by conventional pressing.

20. Machining. The capsule is machined to the fine tolerances necessary for adequate welding.

21. Cleaning. The components of the capsule are cleaned chemically.

22. Assembly. The lithium hydride-aluminum-transuranium oxide is assembled in the capsule halves.

23. Welding. Welding together of the two capsule halves is accomplished by tungsten inert gas welding.

24. Shipment. The completed container is packaged for shipment.

8.2.4 Ground Transportation

Assuming that partitioning and encapsulation will take place at the reprocessing plant site, transportation of waste to the space transportation site should not present unusual problems. A capsule which is encapsulated for space flight should be suitable for earthbound transportation. If shadow shielding were used for space flight, additional shielding might be required for earth transportation.

For more unusual space flight systems such as particle acceleration, additional analysis of earth transportation requirements may be required.

8.2.5 Space Flight

Consideration was given to several different types of space flight. Concepts considered, some only briefly, include:

- Utilization of the energy in the waste for propulsion.
- Solar sails.
- Nuclear and ion propulsion.

- Acceleration of waste particles electrically from an orbiting platform.

- State-of-the art vehicles (this includes the space shuttle and space tug, which are advanced vehicles but will use existing technology).

The latter four variations appear to have potential for increasing the payload or providing increased propulsion velocity capability. Therefore, the differences in their use versus state-of-the-art vehicles are primarily in degree, as opposed to changing the overall conclusions from the study. (However, it is possible that other trajectories for final destinations--e.g., disposal into the sun--could become more practical with development of such concepts.) The non-state-of-the-art systems are much less developed and would require studies beyond the scope of this study to evaluate them fairly and uniformly. Therefore, while some warrant further study, it was concluded that the analysis using state-of-the-art vehicles provides a base point for evaluation of feasibility.

• Utilization of the Energy in Waste for Propulsion

NASA/Ames has analyzed the potential for use of waste-generated heat for propulsion. The analysis is included in Appendix C. This approach requires high heat density in the waste materials and appears promising only if short-lived isotopes such as strontium-90 are included in the waste package. The extra energy in the waste would, in turn, be compensated for by the increased shielding (and payload) requirements. The conclusion of that preliminary study was

that the approach warrants further considerations.

- Solar Sail

A solar sail vehicle utilizes a relatively large surface that is pushed upon by the sun's photon radiation, Figure 8.10. It can be utilized to propel a vehicle into the sun as well as away from the sun. To propel a vehicle into the sun, the sail must be placed at an angle such that the force of the sail tends to reduce the vehicle's solar orbital velocity. The solar force on the sail at the earth's distance from the sun is on the order of 10^{-4} dynes/cm². At speeds close to the earth's orbital speed, which is the maximum the vehicle would have, the drag is about 10^{-8} dynes/cm². This drag assumes a particle density of 10^3 particles/cm³.

Solar sail propulsion was studied by a group of students at Massachusetts Institute of Technology⁽⁷⁾ as a possible means of reducing the cost of space disposal. The conclusion was that: "The solar sail-powered mission offers such advantages over present propulsion schemes that it may be termed the only way to sunfall. It is basically unsophisticated, and no breakthrough in the state-of-the-art is necessary. It offers the inherent advantages of the direct sunfall mission: open launch window, low navigational requirements, definitive payload rate, and low time of flight. There are some uncertainties, however. Perhaps the main one is the final phase of flight, where solar radiation, even if simply reduced to infrared band, is likely to

melt down what is left of the spacecraft. Obviously, the velocity conditions at the moment must be such that free fall will result in sun impact."

The solar sail vehicle could possibly provide a realistic means of achieving direct solar impact. While there are a number of unanswered technical questions (such as the life of the sail), it appears to be a technical possibility.

- Nuclear and Ion Propulsion

High specific impulse vehicles such as nuclear rockets and ion thrusters have been mentioned in some studies, and the possibility of using such vehicles was considered briefly here. Use of such vehicles could ultimately make significant improvements in the economics of space disposal.

- Acceleration of Waste Particles from an Orbiting Platform

The brief study on waste particle acceleration is included in Appendix 8.D. The particle acceleration method contemplates transporting waste from earth to a permanently orbiting platform. On the orbiting platform the waste would be converted to the form of small charged particles. These particles would then be electrostatically accelerated to the desired velocity. One of the principal advantages of this system is the savings in energy requirements made possible by elimination of a container.

The remainder of this section will treat the technical feasibility of state-of-the-art space flight, which was the primary emphasis for this study.

The sail angle, θ , can be adjusted so the light force, F_s , on the sail and the sun's gravity force, F_g , interact to either increase or decrease the orbital velocity of the ship. A decrease in orbital velocity will cause fall into the sun.

FIGURE 8.10 Illustration of Solar Sail Vehicle

- State-of-the-Art Vehicles

State-of-the-art space flight includes vehicles such as the space shuttle and the space tug. These vehicles are not yet built, but they can be built with current engineering knowledge. The space shuttle is currently in the development stage.

The analysis below (through the section "Potential Space Transportation Vehicle Performance and Cost") was performed by the staff at NASA Lewis⁽¹²⁾ and is included here nearly verbatim. Minor modifications were made to conform to the format of this report.

The space destinations considered in this study include Earth orbits, solar orbits, solar system escape and solar impact. The space destinations are discussed in the order of increasing energy requirement. All launches are assumed to occur from the Eastern Test Range (ETR) in an easterly direction.

It is assumed that the launch vehicle will first launch into a low circular Earth parking orbit, although this is not always necessary nor advantageous. After parking in this orbit, the launch vehicle upper stage or stages will inject the waste package to its final destination. Mission energy is characterized by the mission Delta-V requirement which is defined as the sum of all the velocity increments that the launch vehicle has to provide after reaching low Earth orbit. In some cases the launch system alone can place or inject the waste package to its final destination. In other cases the waste package, after separation from the launch vehicle, will require subsequent trajectory (midcourse) corrections or propulsion upon reaching its destination. In these cases the waste package is part of an active spacecraft requiring propulsion systems and the addition of guidance, control and communications systems.

8.2.5.1 Potential Destinations

• High Earth Orbit

To achieve high circular final Earth orbits, starting from a low circular parking orbit, two propulsion maneuvers are required. The first is made in the parking orbit

and places the payload on an elliptical transfer orbit. After coasting along the transfer orbit to the desired final altitude, the second maneuver is made to circularize the final orbit. Both of these maneuvers are expected to be performed by the launch vehicle upper stage.

For high Earth orbits, in the event of a propulsion failure after reaching the parking orbit but prior to final placement, corrective action can be taken. The resulting orbit would have an adequate lifetime (several months) so that a second launch could be made to rendezvous with the waste package. The waste package would then either be sent to its final orbit or be retrieved. This discussion also applies to other destinations if the propulsion failure occurs prior to reaching Earth escape velocity.

For the disposal of nuclear waste it is not clear as yet which orbit altitudes are acceptable. Orbit lifetime is a primary factor. Orbit lifetimes of a million years or longer may be required if long-lived wastes are to be disposed of in space. At reasonably high orbit altitudes, above several thousand kilometers, atmospheric drag is negligible, but other perturbations such as solar pressure and solar, lunar, and planetary gravitational perturbations must be considered. Orbits near the Moon must be avoided to minimize lunar perturbations. High traffic regions or orbits important from a science or applications point of view (such as synchronous orbit altitude and some lower altitudes) should not be chosen.

Therefore, probably the best choice for Earth orbits would be those orbits lying between synchronous orbit altitude and the Moon. However, such orbits have the highest Delta-V requirement of the high Earth orbits, on the order of 4.1 km/sec.

Advantages of High-Earth Orbit

- The Delta-V required is relatively low in comparison to some of the other destinations.
- The waste package could conceivably be retrieved at a later date either to recover the waste material or to remedy some unforeseen problem.
- There is a launch opportunity any day.
- The waste package could be passive, requiring no guidance or propulsion capability since the second propulsive burn is performed by the launch vehicle upper stage.

Disadvantages of High-Earth Orbit

- The stability of high Earth orbits and hence orbit lifetime over a long period of time (on the order of a million years) is not well understood. To date the complexity of the multi-perturbation problem precludes rigorously verifying the stability of these orbits over these long-time periods.
- There is no assurance of the integrity of the relatively hot waste package when exposed to the space environment over these long periods of time.
- Eventually, the waste packages will be randomly located within a belt around the Earth. Gravitational perturbations cause orbits of the waste packages to vary slightly, with time, thus producing variations in orbits in this region. This region

would be regularly penetrated by future planetary spacecraft. However, because of the wide spacing between waste packages at such high placement altitudes, the probability of a collision would be extremely remote.

Since neither orbit stability nor waste package integrity problems are well understood, high Earth orbits cannot be considered a permanent disposal site for times on the order of a million years. Unless further studies can resolve these problems, Earth orbits should only be considered for hundreds or a few thousand years, with potential need for requiring further action at a later date.

- Solar Orbits

The solar orbits considered in this study are those achievable with relatively low Delta-Vs. These include (1) solar orbits achievable by injecting the waste package to Earth escape velocity or slightly beyond, (2) circular solar orbits slightly inside or outside the Earth's orbit about the Sun achieved by additional propulsion after escaping the Earth and (3) solar orbits achievable by swinging by Mars or Venus.

Earth Escape. The simplest method for achieving a solar orbit is to have the launch system inject the waste package to Earth escape energy. This can be done with a single propulsive burn from Earth orbit with a Delta-V of approximately 3.23 km/sec. The waste package would then be separated from the launch vehicle and after escaping the Earth's gravitational field would be in an orbit about the Sun. The waste package

would be in essentially the Earth's orbit about the Sun but in a different angular position.

With the waste package in this orbit, there is a high probability of the waste package re-encountering the Earth at some future time. Due to inherent limitations on injection accuracy and long-term gravitational perturbation effects--principally from the Earth--the waste package cannot be maintained at a fixed position from the Earth. As a result of these effects the waste package tends to drift with respect to the Earth, and preliminary calculations indicate a high probability of re-encountering the Earth within a few thousand years or less.

A better approach would be to provide somewhat more Delta-V than required for Earth escape (an additional Delta-V on the order of 0.42 km/sec for a total incremental Delta-V of 3.65 km/sec), so that the waste package would be in a slightly elliptic solar orbit with a small inclination to the ecliptic plane (plane of the Earth's orbit about the Sun). Initially, the orbit of the waste package would intersect the Earth's orbit at only one point. Furthermore, planetary gravitational effects tend to precess or vary the orbit of the waste package with respect to the Earth's orbit making an encounter even less likely. Preliminary calculations indicate that such is the case at least for a few thousand years.

Advantages of Elliptical Solar Orbits

- Of all the mission destinations or orbits considered except for some

Earth orbits, this requires the lowest Delta-V. The Delta-V required is approximately 3.65 km/sec which is slightly more than required to reach Earth escape velocity.

- Only a single propulsive phase from low Earth orbit is needed.
- There is a launch opportunity any day.
- The waste package could be passive, requiring no active spacecraft systems.

Disadvantages of Elliptical Solar Orbit

- There is no assurance that the waste package will not re-encounter the Earth for periods measured in thousands of years.
- There is an abort gap past Earth escape velocity which would preclude recovery. If the launch vehicle should fail after reaching Earth escape velocity, the waste package would be left in an unplanned solar orbit with subsequent Earth encounter possibilities. With the current state-of-the-art launch vehicle technology, it would be impractical to recover the waste package from these orbits.

There is no assurance that trajectories can be developed (and demonstrated analytically) which eliminate the possibility of reencounter with Earth for times on the order of a million years. Because of this uncertainty, Earth escape type of solar orbits cannot be established as a proven, acceptable destination for time periods greater than thousands of years at this time.

Circular Solar Orbits

In order to provide a positive separation between the orbit of the

waste package and the orbit of the Earth, the waste packages could be placed in circular solar orbits, either inside or outside Earth's orbit about the Sun. These circular orbits should be either inside 0.983 AU (astronomical unit, which is the mean distance between the Earth and the Sun, 149,000,000 kilometers) or outside 1.071 AU (the perihelion [the point nearest the Sun] and aphelion [the point furthest from the Sun] distances of the Earth's elliptical orbit around the Sun) to ensure that the waste package does not collide with the Earth. There is an incentive, however, for going no further than necessary since the required Delta-V increases with increasing distance from the Earth's orbit. For comparison purposes, a final orbit radius of 0.90 AU, which is inside the Earth's orbit, is used in this study. Starting from Earth orbit, two propulsive burns are required to reach the desired 0.90 AU circular solar orbit. The first burn requires 3.26 km/sec Delta-V from the launch system to inject the payload to slightly past Earth escape energy. After escaping from the Earth, the waste package is in an elliptical solar transfer orbit having the desired perihelion but with an aphelion still at the Earth's orbit from the Sun. The second burn adds 0.81 km/sec (for a total incremental Delta-V of 4.07 km/sec) and circularizes the orbit. This burn is performed by another propulsion stage, upon reaching perihelion after approximately a six-month coast.

Advantages of Circular Solar Orbits

- The Delta-V required is low in comparison to some of the other destinations. For the 0.90 AU circular solar orbit, a total Delta-V of 4.07 km/sec is required.
- There is a launch opportunity any day.

Disadvantages of Circular Solar Orbits

- The problem of assuring the stability of solar orbits for times on the order of a million years is unresolved, generally for the same reasons given for Earth escape and high Earth orbits. Presumably, the final orbit could be placed sufficiently far from the Earth's orbit to preclude a subsequent collision with the Earth over the times required.
- There is an abort gap past Earth escape velocity.
- In addition to the launch system, another propulsion system along with guidance, control and communications is required to perform the second burn. It is impractical to accomplish this burn with the launch system due to the long coast phase (about 6 months). This disadvantage could be diminished by performing the second burn for circularization with a simple spin stabilized, solid rocket motor.
- If the circularization burn should fail, the waste package would be left in an elliptic solar orbit, intersecting the Earth's orbit near aphelion. For these cases there is a high probability that the payload will eventually re-encounter the Earth. This

probability can be reduced by using departure trajectories similar to those suggested earlier for the Earth escape case.

Integrity of the waste package is an important consideration for this mission because its possible disintegration over long periods of time can influence the choice of an interior or exterior orbit. If the waste package should disintegrate, the Poynting-Robertson effect will tend to draw the smaller fragments into the Sun. If part of the waste package should vaporize, the solar wind could tend to move some of the material out from the Sun. If the integrity of the waste package cannot be assured over the long time period, these and other effects will have to be evaluated, not only in making the selection of orbit location, but also to establish the ultimate destinations of the waste material.

If the integrity of the package and the stability of the circular solar orbits (near Earth) can be assured for the time period of concern, circular solar orbits can be considered as a possible disposal destination. In addition, further study is required to evaluate the consequences of possible failure situations.

Solar Orbit via Venus and Mars

Another method for achieving solar orbits that do not cross the Earth's orbit is to swingby another planet, using the gravitational attraction of that planet to change the initial swingby trajectory. The resulting post-swingby trajectory does not cross the Earth's orbit; however, it

will periodically cross the swingby planet's orbit. Both Mars and Venus swingbys can be achieved with Delta-Vs only slightly higher than Earth escape. The total Delta-V consists of two Delta-Vs. The first Delta-V performed by the Launch vehicle injects the payload onto a target planet swingby trajectory, the second Delta-V, performed by another propulsion system after swinging by the target planet, places the waste package in the desired solar orbit. The above maneuver is performed to prevent a subsequent encounter with the swingby planet. The total Delta-V for either a Venus or Mars swingby missions is approximately 4.11 km/sec.

Advantages of Solar Orbit via Venus or Mars

- The total Delta-V required for either a Venus or Mars swingby mission is relatively low in comparison to some of the other destinations.
- With a properly oriented swingby the trajectory can be altered so that the post-swingby orbit will no longer cross the Earth's orbit.

Disadvantages of Solar Orbit via Venus or Mars

- For swingby missions the launch opportunity is limited. A launch opportunity to Venus occurs only once every 19 months and to Mars about once every 26 months. The duration or "width" of each of these launch opportunities can be about three to four months long without major increases in injection Delta-V (the wider the launch opportunity the higher the required injection Delta-V).

- The waste package will require a midcourse trajectory correction system (with currently achievable injection accuracies) to insure achieving a proper swingby position at the swingby planet.
- An additional propulsion system is required to prevent a post encounter with the swingby planet. This propulsion system and associated systems must perform reliably after a long coast phase (many months).
- The problems of long-time stability of the solar orbit and containment system integrity are unresolved, although these problems would be less important than for the previously discussed destinations which are closer to the Earth.
- There is an abort gap past Earth escape velocity.

Launch opportunities for either a Venus or Mars swingby appear to be quite limited. Such an operation would be expensive in terms of required Shuttle fleet size, number of launch facilities and use of ground crews. (For example, the reusable Space Shuttle is expected to have a two-week turn-around-time between launches.) These swingby missions offer no outstanding advantages over the 0.90 AU solar orbit (which can be launched on any day).

Solar System Escape

- Since both Earth orbit and solar orbit destinations have uncertainties regarding long-time orbit stability and containment system integrity, solar system escape and solar impact should also be considered as possible waste package destinations. Of the two, it takes less energy to escape the solar system, and this case will be discussed first.

Direct Solar System Escape. This can be achieved with a single propulsion burn from low Earth parking orbit with all the propulsion and guidance provided by the launch vehicle.

Advantages of Direct Solar System Escape

- The waste package is removed from the solar system.
- The waste package can be passive and requires no additional propulsion or astronics systems.
- There is a launch opportunity any day.

Disadvantages of Direct Solar System Escape

- An 8.75 km/sec Delta-V is required. This is high in comparison to the Delta-Vs required for high Earth orbits and solar orbits.
- There is an abort gap past Earth escape velocity.

There is a small variation in injection Delta-V depending on the launch day. The most efficient trajectories will be in or near the ecliptic plane of the earth's path and consequently will fly through the asteroid belt. There is no difficulty in targeting the trajectory to miss the outer planets. As a point of interest it takes approximately 20 years for the waste package to reach the mean orbital distance of Pluto (the planet in our solar system which is the farthest from the Sun) but it will take over a million years to reach the distances of the nearest stars. Solar system escape is the most attractive destination discussed thus far for assurance of long-term waste disposal.

Solar System Escape via Jupiter Swingby. Solar system escape can be achieved with a properly designed swingby of Jupiter using a single propulsion phase from low Earth orbit. As a result of using a Jupiter swingby, the Delta-V required to achieve solar escape energy is somewhat less than that required for a direct solar system escape mission.

Advantages of Solar Escape via Jupiter Swingby

- The waste package is removed from the solar system.

Disadvantages of Solar Escape via Jupiter Swingby

- The Delta-V required is approximately 7.01 km/sec, which is still high in comparison to some of the other destinations.
- The launch opportunity is limited, occurring only once every 13 months with perhaps a 60 to 90-day launch duration or "width."
- A midcourse trajectory correction capability is needed as was the case for the Venus and Mars swingbys.
- There is an abort gap past Earth escape velocity.

The Jupiter launch opportunity is sufficiently limited that many facilities and personnel would be required to support the anticipated number of launches required, and in general the Jupiter swingby could be more restrictive than the Mars and Venus swingbys. It would be simpler to use a direct solar system escape, even though the Delta-V is some 1.74 km/sec higher than for the Jupiter swingby.

- Solar Impact

A solar impact can be achieved directly or via Jupiter swingby.

Again the purpose in using a Jupiter swingby is to reduce the Delta-V.

Direct Solar Impact. This can be achieved with a single propulsion phase out of a low Earth orbit with all the propulsion and guidance provided by the launch vehicle. Enough Delta-V must be provided by the launch vehicle to cancel the Earth's orbital speed about the Sun, so that the waste package "falls into" the Sun. For a direct impact a Delta-V of approximately 24.08 km/sec is required. A grazing impact into the edge of the Sun could reduce the Delta-V requirement to about 21.34 km/sec.

Advantages of Direct Solar Impact

- The waste package is destroyed.
- The waste package can be passive.
- There is a launch opportunity any day.

Disadvantages of Direct Solar Impact

- The Delta-V required is extremely high.
- There is an abort gap past Earth escape velocity.

For this mission the Delta-Vs required are far beyond the capability of current or currently planned launch systems and therefore are considered impractical until more advanced systems are developed.

Solar Impact via Jupiter Swingby.

As was the case for the solar system escape via Jupiter swingby, a solar impact can be achieved with properly designed swingby Jupiter using a single propulsion phase from a low Earth parking orbit. By using a Jupiter swingby to achieve a solar impact, the Delta-V required is appreciably

less than that required for a direct solar impact. For this mission the Delta-V required is above 7.62 km/sec.

Advantages of Solar Impact via Jupiter Swingby

- The waste package is permanently removed from earth and is destroyed.

Disadvantages of Solar Impact via Jupiter Swingby

- The Delta-V is still high in comparison to some of the other destinations.
- The launch opportunity is limited, occurring only once every 13 months with perhaps a 30 to 60-day launch duration or "width."
- A midcourse trajectory correction capability is needed which increases mission complexity.
- There is an abort gap past Earth escape velocity.

The Delta-V required for this case is about 1.13 km/sec less than that required for a direct solar system escape mission. Many facilities and personnel would be needed to support the required number of launches during the limited launch opportunity.

- Other Destinations

Many other space destinations in addition to those discussed have been suggested. Examples include depositing the waste packages on the Moon, on planets, in orbits of other planets, on asteroids, at Lagrangian equilibrium points and so forth. These destinations were not considered in the detailed analysis although in some cases they could warrant further investigation. The general arguments used by NASA against these destinations include

(1) the regions are unexplored and/or are of scientific interest, (2) some of the regions could be of future value from an alternative applications standpoint, (3) launch opportunities are limited, (4) deep space propulsion is required, and (5) in many cases the retro Delta-Vs are high for soft landings.

Costs and energy requirements for placement on the moon or other extra-terrestrial bodies are expected to be between those for high earth and solar orbits and direct solar escape. It would be technically possible to use such repositories with state-of-the-art vehicles.

- Comparison of Destinations

To summarize the destinations discussed, Table 8.10. lists the typical Delta-V requirements for the various missions and their principal advantages and disadvantages. The Delta-Vs shown are representative for each destination, although there will be some variation depending on the particular launch opportunity and details of the mission profile. The Delta-V for high Earth orbits is an upper value for orbits between synchronous and lunar orbit altitudes. The Earth escape (solar orbit) Delta-V includes some provision for additional Delta-V in an effort to minimize the probability of a subsequent Earth re-encounter as was discussed earlier. The Delta-Vs for the other solar orbits include the Delta-Vs required by the waste package after departing from Earth. Passive waste package implies that it will require no special space propulsion, midcourse or associated astrionics systems. The abort possibility past Earth escape

TABLE 8.10. Summary of Potential Space Destinations

Destination	Delta-V, km/sec	Advantages	Disadvantages
High-Earth Orbit	4.11	Low Delta-V Launch any day Passive waste package Can be retrieved	Long-term container integrity required. Orbit lifetime not proven.
Solar Orbits Via:			
Single burn beyond Earth escape	3.65	Low Delta-V Launch any day Passive waste package	Long-term container integrity required. Earth re-encounter possible (may not be able to prove otherwise). Abort gap past Earth escape velocity.
Circular Solar Orbit	4.11	Low Delta-V Launch any day	Long-term container integrity required. Orbit stability not proven. Requires space propulsion system. Abort gap past Earth escape velocity.
Venus or Mars Swingby	4.11	Low Delta-V	Long-term container integrity required. Limited launch opportunity (3 to 4 months every 19 to 24 months). Requires midcourse systems. Need space propulsion or have possibility of unplanned encounter.
Solar System Escape:			
Direct	8.75	Launch any day Passive waste package Removed from solar system	High Delta-V Abort gap past Earth escape velocity.
Via Jupiter Swingby	7.01	Removed from solar system	High-Delta-V, Limited launch opportunity (2 to 3 months every 13 months). Requires midcourse systems. Abort gap past Earth escape velocity.
Solar Impact:			
Direct	24.08	Package destroyed Launch any day Passive waste package	Extremely high Delta-V. Abort gap past Earth escape velocity. Not possible with present vehicles.
Via Jupiter Swingby	7.62	Package destroyed	High Delta-V. Limited launch opportunity (1 to 2 months every 13 months). Requires midcourse guidance systems. Abort gap past Earth escape velocity.

Note: Delta-V is the incremental velocity required to leave a low-earth orbit.

An abort gap is a short time period wherein a controlled abort of the mission cannot be accomplished if the flight is off-course.

velocity is a disadvantage associated with all destinations beyond the Earth.

The conclusions reached thus far indicate the primary candidate mission destinations are direct solar system escape or possibly circular solar orbits for long-term disposal, or high Earth orbits for short term disposal, as shown previously in Figure 8.5. The capability of possible launch systems for each candidate mission destination is discussed in the next section.

8.2.5.2 Potential Space Transportation Vehicle Performance and Cost

- Basic Considerations

Only the larger current and planned launch vehicles are considered in this study. The vehicles considered are shown in Figure 8.11. The Titan IIIE/Centaur is the expendable booster that will launch the 1975 Viking mission to Mars. The Saturn V is the three-stage expendable Apollo booster. Its two-stage version was used to launch Skylab. The Space Shuttle is primarily reusable and is expected to be operational in the 1980's. It is planned as a replacement for virtually all the nation's space boosters in operation today.

One of the most important factors in assessing the feasibility of space disposal is cost. The costs presented in this section are those for the launch vehicles and their operations which were used for assessment of technical feasibility. Conversion of these costs to those used for

total waste management operating and capital costs is presented in section 8.5. These data can be used for comparative purposes for preliminary determination of the best launch vehicles and the most promising mission destinations.

- Expendable Launch Vehicle Performance and Cost

Performance and cost data for the Titan IIIE/Centaur and the Saturn V (and the Space Shuttle) are listed in Table 8.11 for the high earth orbits and solar orbits. Performance data are based on a launch due East from the Eastern Test Range (ETR) into a 185-kilometer parking orbit. The upper stage of the launch vehicle provides the Delta-V needed to accelerate the payload to higher energies from the parking orbit. The direct solar impact mission (24.08 kilometers per second) is not shown because it is well beyond the capability of current launch vehicles.

The costs of the expendable launch vehicles are highly use-rate dependent. The Titan IIIE/Centaur cost is about \$27 million at a production rate of four per year. At the higher launch rates expected for space disposal of radioactive waste, the cost would be expected to be considerably lower. For this study, it is assumed that the cost of the Titan IIIE/Centaur at high launch rates, can be reduced about 30 percent and its cost is taken at \$19 million as shown in Table 8.11. Similarly, the costs of the Saturn V and Saturn V/Centaur are taken at \$150 and \$155 million, respectively. Note that the costs used in this section include only the

FIGURE 8.11. Possible Launch Vehicles for Space Disposal of Radioactive Waste

costs of the launch vehicles and their operations. They do not include operational costs associated with handling the nuclear waste container at the launch site or the integration of the waste package with launch vehicle. These latter costs

are included with those for total waste management in section 8.5.

- Space Shuttle/Third Stage Performance and Cost

The Space Shuttle by itself can deliver payloads only to low Earth orbit. Missions beyond low Earth

TABLE 8.11. Space Launch Vehicle Performance and Cost
for High Earth Orbits and Solar Orbits

<u>Launch Vehicle</u>	<u>Payload, kg</u>	<u>Launch Cost</u>	
		<u>10⁶ Dollars</u>	<u>Dollars/kg</u>
Expendable Vehicles:			
Titan III E/Centaur	3,860	19	4,920
Saturn V	32,660	150	4,590
Saturn V/Centaur	35,290	155	4,390
Space Shuttle:			
Reusable Tug Current Size	4,170	12.25	2,940
Reusable Tug Optimum Size	4,670	12.25	2,620
Centaur Current Size	6,490	16	2,460
Centaur Optimum Size	8,480	16.3	1,920

Note: a. Delta-V = 4.11 km/sec.
b. For direct solar escape (Delta-V = 8.75), the payload for the Titan III E/Centaur and Saturn V = 0 and for the Saturn V/Centaur is 7480 kilograms.

orbit must be accomplished by having the Space Shuttle carry both a propulsion stage and the mission payload to Earth orbit in its cargo bay. The propulsion stage is generally referred to as a Space Shuttle third stage. After the third stage and payload are deployed in Earth orbit from the Shuttle Orbiter, the third stage will inject the payload to its destination. Existing expendable upper stages are currently being evaluated for early use as Space Shuttle third stages. These stages would be expended on each flight. However, it

is planned to eventually develop a new reusable Space Tug explicitly for use as a Space Shuttle third stage and having the capability of being recovered and reused. The Space Shuttle would launch the Tug and payload into low Earth orbit. After the Tug and payload are deployed from the orbiting Shuttle, the Tug will inject the payload to its mission destination. Following the injection burn, the payload is separated from the Tug and the Tug does a series of burns to return to the waiting Shuttle Orbiter for recovery and reuse.

Several Space Shuttle/Third Stage options were considered in this study. These include: (1) One of the reusable Space Tug concepts under study by NASA. It is designed to have the capability of performing a round-trip mission to geostationary (synchronous) orbit with a 1360-kilogram payload. It is a hydrogen-oxygen fueled stage with an engine specific impulse of 470 seconds and has a propellant capacity of approximately 24,040 kilograms. (2) A similar reusable Tug but optimally sized for the waste disposal mission. (3) The existing expendable Centaur stage. It also uses hydrogen-oxygen propellants and has an engine specific impulse of 444 seconds and a propellant capacity of about 13,610 kilograms. (4) A similar expendable Centaur stage but resized for the waste disposal.

With these various Space Shuttle/third stage options useful payloads are achievable to only high Earth orbit and solar orbit destinations. Their performance, which is based on a mission Delta-V of 4.11 kilometers per second, is shown in Table 8.11. The performance data are based on a Space Shuttle delivery capability of 29,484 kilograms into a due East 185-kilometer orbit, which is a Shuttle specification. Note that the optimally sized third stages have higher payloads.

For the high Earth orbits and solar orbits, the reusable Tug, at its current size, can deliver a payload of 4,170 kilograms, whereas the optimally sized Tug (about 20,870 kilograms propellant) can deliver a

payload of 4,670 kilograms. The current-size Tug performance is lower than the resized Tug because its propellant capacity is too large for the higher payload weight of waste disposal missions, and the Tug must be off-loaded. In the case of the Centaur, the propellant capacity is too small to utilize the full orbital capability of the Space Shuttle. The performance of the Centaur stage can be improved if its propellant capacity is increased. For the high Earth orbits and solar orbits, the current Centaur stage can deliver 6,490 kilograms. An optimally sized Centaur (about 17,240 kilograms propellant capacity) can deliver a payload of 8,480 kilograms.

It should be recognized that the higher payload capability shown for the Centaur stage is a consequence of its being expended rather than recovered. For the reusable Tug, a portion of its propellant is required to return to the Shuttle Orbiter waiting in low Earth orbit. For the expendable Centaur stage, all the propellant is used to achieve the desired mission Delta-V, and its payload is accordingly higher. If the Tug were expended, its performance would be comparable to that for the optimally sized Centaur stage.

The cost per Space Shuttle flight is currently estimated at approximately \$10.5 million. In addition, the cost per reusable Tug flight is assumed to be \$1.75 million, which includes operations, refurbishment and amortization of a unit production cost of \$20 million/Tug. Totaling

the two, the cost per flight of a Space Shuttle/reusable Tug is about \$12.25 million. The cost of the expendable Centaur stage at the high launch rates required for waste disposal would be about \$5.5 million. In total, the cost of a Space Shuttle/expendable Centaur launch is about \$16 million.

- Launch Vehicle Performance/Cost Comparison

Except for the Saturn V/Centaur, the launch vehicles considered thus far can only deliver useful payloads to high Earth orbit or solar orbit destinations. In order to provide an overall vehicle comparison for these destinations, the payload, cost per flight and cost per kilogram of payload delivered to a Delta-V of 4.11 km/sec were summarized in Table 8.11. These data should be used only for making preliminary comparisons since other factors will have to be considered in making a vehicle selection. For example, there are limits on the desired waste package size. Also, the nuclear waste is only a small fraction of the total waste package weight, and this fraction will vary with waste package size. These and other factors will influence the choice of a launch vehicle for a particular destination. Nonetheless, Table 8.11 shows that the Space Shuttle vehicles are more cost effective than the current expendable launch vehicles. The cost per pound of total payload delivered for the Space Shuttle vehicles is on the order of one half of that when using expendable launch vehicles.

For the shuttle launched missions, it appears worthwhile to resize the

upper stages for the waste disposal mission. The improved performance and cost effectiveness should readily justify the nonrecurring costs associated with resizing the stages. For the high Earth orbits or solar orbits the cost per pound of payload delivered for the resized Centaur stage is about 25% lower than for the resized reusable Tug. This indicates that an expendable Shuttle stage would be more cost effective than a reusable stage. This conclusion is sensitive to the required mission Delta-V. If the required mission Delta-V were below about 3.4 km/sec, a reusable Shuttle third stage (Tug) could be more cost effective than an expendable stage (Centaur). In addition to performance and cost, safety considerations and specific mission details can influence the final choice of a Shuttle third stage. From this reasoning, both reusable and expendable Shuttle third stages were considered for further evaluations.

- Multiple Space Tug Configuration Performance and Cost

The only launch vehicle considered thus far that has a useful payload capability for the direct solar escape mission is the Saturn V/Centaur. As shown in Table 8.11, it can deliver a payload of about 7,480 kilograms to this destination. At a launch cost of \$155 million, this results in a specific cost of 20,720 dollars per kilogram, or roughly an order of magnitude higher than for the Shuttle launched cases to high Earth or solar orbits. One possibility for providing a more cost effective solar escape capability is

to use several Shuttle/Tug launches to assemble a larger vehicle in Earth orbit. This same approach could also be used to provide higher payloads for the Earth orbit and solar orbit destinations.

The procedure would be to use several Shuttle launches to place several Tugs in low Earth orbit along with the payload. The Tugs, which have the inherent capability of being able to rendezvous and dock with each other, would be assembled in orbit to form a tandem vehicle. In performing the mission, the Tug stages will burn sequentially, and each stage, if it is to be recovered, will return to its waiting Shuttle Orbiter.

In this preliminary evaluation of a tandem vehicle, only the fixed size Tug concept was investigated. It is assumed to be available in both reusable and expendable configurations. The Tug and Shuttle performance parameters and costs are the same as discussed earlier. The one exception is the cost of an expended Tug. The expected unit cost of the reusable Tug is on the order of \$20 million. If the waste disposal mission required expending a Tug, the cost of the expendable Tug could be considerably lower. The production rate for an expendable Tug would be much higher than for a reusable Tug since each disposal mission would require a new Tug. The high use rate would probably justify development of an expendable Tug incorporating only the features necessary for accomplishment of the waste disposal mission. As an alternative approach, a modified version of the existing Centaur stage could be used as an expendable Tug.

An accurate cost for the expendable Tug cannot be established at this time, but for the purposes of this study it is taken as \$6.0 million per flight.

Several tandem Tug configurations can accomplish the direct solar escape mission, two of which are considered here for illustrative purposes. The first tandem configuration considered consists of two stages, a reusable Tug plus expendable Tug, and requires two Shuttle launches. The first Shuttle launch carries a full Tug to orbit and the second carries an off-loaded expendable Tug plus payload. The second tandem configuration considered consists of three stages, two reusable Tugs plus an expendable Tug, and requires three Shuttle launches. The first two Shuttle launches carry full reusable Tugs to orbit and the third carries an off-loaded expendable Tug plus payload. In both configurations the recoverable Tugs are the lower stages (burned first) since this is an optimum arrangement. It is assumed that the recoverable Tugs are brought back to Earth with the Shuttle Orbiters used to initially launch the Tugs. That is, no additional Shuttle cost is charged for returning a Tug.

The ideal payload performance capability to solar escape for the two- and three-stage tandem configurations is 3,900 and 6,080 kilograms, respectively. However, gravity losses will significantly reduce the actual performance of these multi-Tug configurations. The gravity losses have been determined for these configurations assuming the Tug has a thrust

level of 88,940 New . . . The actual payload capability of the two-stage and three-stage configurations for direct solar escape is 2,270 and 3,040 kilograms, respectively.

A higher Tug thrust level could be used to reduce the gravity losses, but it is not expected that the new Tug engine will have a thrust level higher than 88,940 Newtons. Another approach to reducing the gravity losses is to use a technique referred to as perigee propulsion. This is operationally more complicated and necessitates carrying the waste package once around the Earth in an elliptical orbit between Tug burns. However, using perigee propulsion increases the payload capability of the two- and three-stage configurations for direct solar escape to 3,270 and 4,400 kilograms, respectively.

An overall comparison of launch vehicles performance and cost for the direct solar escape mission is shown in Table 8.12. The expendable Saturn V/Centaur provides the highest payload weight, but at a cost of about \$20,700 per kilogram. The multiple Shuttle/Tug configurations using perigee propulsion achieve lower payloads but at a cost of about \$9,000 per kilogram.

- Space Transportation System Recommended

The currently planned Space Shuttle is more cost effective than current expendable launch vehicles by about a factor of two. The Space Shuttle will require a third stage to perform the disposal missions. Depending on the particular mission, this could be either a reusable stage,

such as the Space Tug, or an expendable stage such as a Centaur. In either case, the third stage should be resized for the selected disposal mission. In fact, the launch rates required for waste disposal are expected to be sufficiently high that it would probably be worthwhile to develop a version of the entire launch vehicle dedicated to providing maximum performance, higher reliability for the disposal mission, and lowest cost.

In this study, only current or planned space transportation systems were considered. It should be recognized, however, that the need for waste disposal is expected to extend far into the future and new space technology and systems development can be expected. These new developments should be reviewed periodically for their application to space disposal.

8.2.6 Energy Balance

The energy required to place waste material in a space disposal trajectory is compared on a fundamental basis here, with the beneficial electrical energy obtained from the production of the waste in the nuclear reactor.

The incremental velocity required to escape the solar system from a 100-nautical-mile earth parking orbit is 8.8 kilometers (28,700 ft) per second.⁽¹²⁾ The velocity required for the earth parking orbit is 7.8 kilometers (25,600 ft) per second. The total velocity required for solar system escape is thus 16.6 kilometers (54,300 ft) per second. With 30 kilograms of transuranium waste resulting

TABLE 8.12. Space Launch Vehicle Performance/Cost for the Direct Solar Escape Mission

Launch Vehicle	Payload, kg	Launch ^(a)	
		Cost, 10 ⁶ Dollars	Cost Dollars/kg
Saturn V/Centaur	7,480	155	20,720
(2,1,1) ^(b) Shuttle/Tug/Tug Configuration			
Without Perigee Propulsion	2,270	28.75	12,660
With Perigee Propulsion	3,270	28.75	8,790
(3,1,2) ^(b) Shuttle/Tug/Tug Configuration			
Ideal Delta-V	6,080	41.0	6,740
Without Perigee Propulsion	3,040	41.0	13,490
With Perigee Propulsion	4,400	41.0	9,320

Notes: a. Delta-V = 8.75 km/sec.

b. (--, --, --) represent the number of Shuttle launches, the number of expendable Tugs, and the number of reusable Tugs, respectively.

from the operation of a 1000 MWe reactor for 1 yr and a weight factor of 20 required for shielding, the energy required to place the waste in a

solar escape trajectory is approximately 3×10^{-6} times the nuclear electrical energy obtained in generating the waste.^(a)

a. The value was obtained as follows:

$$\text{Energy} = \frac{1}{2} MV^2 = \frac{1}{2} \cdot \frac{67 \text{ lb (30 kg) of waste} \times 20 \text{ shielding factor}}{32} \cdot (54,300)^2$$

$$= 6.2 \times 10^{10} \text{ ft-lb} = 8.5 \times 10^9 \text{ kg-m}$$

The electrical energy obtained from operation of the reactor is:
1,000 MWe x 365 days/yr x 24 hr/day = 1,000,000 kW x 8,760 hr

$$= 8.76 \times 10^9 \text{ kW-hr (Assumed 100% operating efficiency)}$$

$$8.76 \times 10^9 \text{ kW-hr} \times 2,655 \times 10^6 \text{ ft-lb/kW-hr}$$

$$= 2.32 \times 10^{16} \text{ ft-lb} = 3.2 \times 10^{15} \text{ kg-m}$$

The energy ratio is thus

Energy required for solar escape of Transuranics/Energy Produced

$$= \frac{8.5 \times 10^9 \text{ kg-m}}{3.2 \times 10^{15} \text{ kg-m}} = 2.7 \times 10^{-6}$$

8.2.7 Safety

The following analysis (Sections 8.2.7.1 through 8.2.7.5) of the safety considerations of nuclear waste disposal in space was developed in Reference (12). These sections are included as authored, with minor changes to fit the format of this report.

The fundamental philosophy of nuclear safety requirements for radioactive waste disposal missions in space can be stated as follows: Potential radiation exposure and harmful contamination of individuals, the population at large, and the ecology shall be negligible.

The statement in the previous paragraph includes also celestial bodies as required by the treaty to promote peaceful exploration and use of outer space which 60 nations, including the United States, have signed.

For operations during all phases of a nuclear waste disposal mission, the exposure and contamination values should be negligible for the population at large and within the permissible standards for the personnel involved in the mission. Criteria governing radiation exposure to humans during ground handling and transportation which may be applicable can be found either in 10 CFR, Parts 20 and 71⁽³³⁾ or in Reference (11).

The nuclear waste disposal missions discussed here are similar to other space shuttle/space trip missions carrying radioactive materials to synchronous earth orbit or to planetary orbits. The primary difference lies in the comparatively large

amount of radioactive material containing actinides being transported during the waste disposal missions.

The purpose of this analysis is to evaluate the response of the nuclear waste package to the potential accidents that could occur during the various phases of waste disposal missions. It was possible only to perform a qualitative evaluation because the mission hardware and systems and the mission proper are only in a preliminary state of definition. And furthermore, definitive design criteria do not exist for this particular type of nuclear waste disposal method.

8.2.7.1 Nuclear Safety Requirements

The nuclear waste contained within the waste package imposes certain nuclear safety requirements on the package design, its supporting equipment, and mission operations. Some of these waste package requirements are listed below:

- Sub-critical design under all conditions
- Encapsulation of waste material
- Shielding for external radiation
- Re-entry protection
- Impact protection
- Fire and explosion protection
- System to transmit location of capsule

Shuttle orbiter supporting equipment needs are

- Temperature Controls
- Monitoring Equipment

Operation requirement needs are

- Parking orbit altitude control with long decay time

- Recovery preparedness
- Future encounter avoidance control
- Orbital retrieval means.

8.2.7.2 Accident Model Considerations

In this section general potential accident cases are discussed qualitatively. Each of the cases includes a series of environments which could be present at various times during the accident. Development of probability numbers is beyond the scope of this study.

- Ground Handling

Provided proper procedures are established, the probability of an accident while handling the nuclear waste package, including installation within the Shuttle cargo bay, will be extremely small.

- Launch Pad Abort

The assumed potential accident would occur at or immediately after launch, causing the Space Shuttle to explode and burn, thus exposing the nuclear waste package to an adverse environment. The environment created by such an accident would be: blast overpressures, residual liquid and solid propellant fires. The residual fire of the solids could last about five minutes at a temperature of approximately 2600°K.

- High Velocity Impact

If a malfunction caused the shuttle to make a 180° change in direction soon after launch, a powered impact could occur. The resulting environment would be similar to those encountered during launch pad abort except the impact would be at high

velocity. Furthermore, a portion of the propellants would have been used, the residual fires would have shorter duration, and the fireball would be smaller. The presence of a crew on board would reduce the probability of occurrence to a very small number.

The actual impact velocity for the nuclear waste package itself would be lower than that of the Shuttle because of the cushioning effects of the shuttle structure.

- Failure During Ascent

Many ascent failures and malfunctions which would lead to catastrophic failure in an unmanned space vehicle would require mission changes or aborts, but not an accident. However, there remains a possibility, although small, that a catastrophic accident would occur on board the Shuttle. If the Shuttle remains intact, the result of the failure would likely terminate with relatively low impact velocity. If a major explosion occurs on board, the nuclear waste package would impact the ground at a high velocity. If the package impacts while still attached to the Tug, there may be an explosion with blast overpressure, fragments, fireball, and liquid propellant fire but at a considerably lower magnitude than during a launch pad abort because of the relatively small quantity of Tug propellants.

- Crash Landing

There is a possibility that the Shuttle Orbiter will make a crash landing. If there was insufficient time to dump the Tug propellants prior to the landing, overpressures, impacts of fragments, fireball, and propellant fires could occur.

- Uncontrolled Re-entry and Impact

The potential accident modes during re-entry considered here are those which might occur after the nuclear waste package has been deployed from the Shuttle. Basically there are then two possibilities: The package re-enters by itself or it is still attached to the Tug. The environment encountered by the package or the Tug/package (or Tug/Tug/package) depends on the entry velocity and entry angle into the atmosphere. (In this exploratory study only the vertical entry was analyzed). The nuclear waste package would be exposed to re-entry heating and thermal stress. If the heat shield of the package were to fail, the waste material encapsulation could fail also and expose the radioactive waste to the re-entry and impact conditions. (Note that the purpose of the heat shield is to preclude this latter failure mode.)

- Post Impact

After an intact impact of a nuclear waste package, it is exposed to environments which could cause overheating and melting, oxidation, and corrosion, which may eventually cause some release of the radioactive material and/or an increase in the external radiation dose. However, for any post-impact condition the external radiation dose represents a hazard. The dose for an intact package was assumed to be 1 rem/hr at 1 meter from the surface of the package. For any degree of breaching of the package the external radiation is increased, whether or not radioactive material is released.

8.2.7.3 Analytical Results of Accident Model

Only a qualitative evaluation was made of the possible release of radioactive materials and the potential hazards resulting from external radiation. A quantitative evaluation and the determination of probabilities can only be made after a more detailed hardware, system and mission definition has been made. Some of the analytical methods have been confirmed with experiments, such as large sphere impact tests of more than 300 m/s velocity and fragmentation tests.

- Nuclear Waste Package Response

The basic nuclear waste package design used in the analyses was the single package design for disposal of transuranium waste containing 1% fission products and shown in Figure 8.3. Where possible, the analytical methods checked against experiments that were closely related to the accident conditions.

- Overpressure

An accident at the launch pad which results in an explosion of the main liquid propellants could produce a blast overpressure of approximately 150 atmospheres (assumes a mixing mode yielding 20% TNT equivalent.)

In the analysis it was assumed the re-entry shell is stripped away, leaving the spherical stainless steel impact vessel to take the full overpressure. This assumption represents a conservative approach. The spherical shape is an ideal shape to withstand the pressure. The yield stress of the stainless steel is

2400 atmospheres (35,000 lb/in.²). An empty vessel with a radius of 0.68 meters and a 2.54-centimeters-thick wall can withstand an external pressure of 175 atmospheres without yielding. Since this is greater than the overpressure, the nuclear waste package will not be breached by the blast overpressure. However, because of the differences in compressive strengths of materials, the external pressure may cause the outer impact shell to shift relative to the internal waste, thus deforming the impact shell and reducing the shielding thickness.

- Fragments

During accidents of the explosion type, fragments of varying sizes and of various materials (predominately aluminum) could impact the nuclear waste package at varying impact velocities. An analysis was made assuming aluminum fragments (sharp and blunt) impacting at 1520 m/sec (5000 ft/sec). The results indicated the containment vessel would not be penetrated.

In addition to the analytical study an experimental test was set up with aluminum pellets fired at a stainless steel sphere with a wall thickness of 1.58 centimeters.

- Fireball

No calculations were performed for this condition. Comparison with other capsules involved in fireball tests indicated that because of the short duration (seconds) and the large mass of the nuclear waste package, no serious damage should result to the nuclear waste package due to the fireball.

- Residual Propellant Fires

Of the two types of fires, the solid propellant produces the higher temperature, 2300°C (4200°F). To evaluate the response of the nuclear waste package to the solid propellant, a heat transfer model was established which consisted of 72 nodes for the various layers of material. It was assumed that the re-entry shell had been stripped away by the overpressure.

Initially the surface heat flux is high since the temperature of the impact shell is low. As the surface temperature rises, the radiation effect from the propellant fire diminishes and the heat flux drops. The temperature of the impact shell approaches the melting point in about five minutes.

Because of the high temperature it is possible to breach the outer impact shell if the solid fire can last for five minutes adjacent to the package. This assumes the solid fuel burns from one side and that the full thickness of the solid fuel is in effect. Although it is possible to cause melting of the outer shell, other layers will still prevent release of the radioactive nuclear waste. However, the shielding (lithium hydride) could be lost and the external dose rate would be increased. This situation would require a shielded or remotely operated retrieval vehicle.

- Atmospheric Re-entry

In the event of an aborted mission, the nuclear waste package could return to earth in an uncontrolled manner (i.e., not on board

the shuttle). For this type of accident there are many combinations of velocity and angle of re-entry. The case which was analyzed and which resulted in establishing the required thickness for the re-entry shell was the vertical re-entry angle at 11 km/sec (36,000 ft/sec). This type of re-entry exposes the re-entry shell surface to a peak heat flux of 300 kW/cm^2 . Although the high heat flux lasts a short time it was felt that this represented a critical condition.

For the above condition the re-entry shell has sufficient thickness to prevent melting through to the impact shell. The calculations indicate that the impact shell temperature does not increase.

Because of the extremely high heat flux it will be difficult to perform a test of this kind to confirm the calculations. In addition, other re-entry conditions may impose other more severe conditions on the package (i.e., skip re-entry) although the probability for this type of re-entry will be very low.

- Impact

Following an uncontrolled abort (i.e., the nuclear package is not brought back with controlled Shuttle) the package will impact the Earth. Based on the design of the re-entry shell, the impact velocity should be 300 m/sec or less. This velocity would be exceeded only if there was an abort with a tug attached and thrusting in.

Based on the experiments and analyses it appears that in most impacts on Earth the package will be buried in the ground with relatively little damage to the outer shell.

However, if the package lands on a surface such as a solid, noncrushable one that does not absorb any of the energy, the nuclear waste package will probably be breached. This condition may or may not result in release of radioactive waste, since it is a low percentage of the matrix and is encapsulated with tungsten protection layers. It can be assumed that for the harder surfaces the nuclear waste package is deformed and possibly breached, and in either case the shielding will be reduced, producing the possibility of higher than designed for external dose rates.

- Post Impact

After an impact the nuclear waste package will either be intact or breached and either buried beneath the surface, partially buried, or on top of the ground. A series of calculations was performed on various degrees of burial. The results indicated that in the no-burial or partial-burial cases the vessel would not rupture within 23 days (approaching equilibrium condition). For deep burial, all cases except those containing waste that produced less than 2 kilowatts of thermal power, resulted in rupture of the impact vessel. This rupture was due to the increased pressure from helium released in the decay process and from dissociating lithium hydride. The rupture would in all probability be a minor crack to relieve the pressure.

No experimental data were available to determine whether or not the surfaces would melt. The calculations did not account for any material changes in the soil (conductivity was varied with temperature).

If the impact vessel remains intact there will be no oxidation and no corrosion for extended periods of time. If the outer vessel is breached, there would be some loss in the effectiveness of the shielding. The waste material already in the oxide form should not react with the surrounding environment. The radiation level in the immediate area would increase, due primarily to the loss of shielding. Because of this, it would be desirable to locate and retrieve the nuclear waste package as soon as possible.

8.2.7.4 Recovery of Nuclear Waste Package

It is desirable to locate an aborted waste package as soon as possible. For those aborts near the launch pad and prior to reaching a parking orbit the vehicle will be tracked and its location predetermined. For these cases recovery procedures can be worked out satisfactorily to recover the package without undue hazards to people in the vicinity. For those aborts which can occur approaching or past Earth escape Delta-V's, in which the nuclear waste package could re-encounter the Earth at some future date, knowledge of impact position may not be immediately known. It would be helpful if some kind of device for transmitting a signal could be incorporated in the package to help in locating the package prior to impact.

8.2.7.5 Summary of Safety Analyses by NASA

Within the framework of this exploratory study, nuclear waste disposal

missions, as described in this report, appear feasible. The waste package design concept with the various protective shells provides a means for preventing release of radioactive waste constituents in most hypothetical accidents. Additional testing would be required to confirm the concept.

The safety study on accident models and package responses point out certain key issues to maintain overall nuclear safety for space disposal of nuclear waste packages. These key issues are:

1. The waste package should be designed to maintain integrity without releasing significant amounts of its radioactive contents throughout all potential hazardous events.
2. The external radiation of the package, whether intact or damaged, has to be held to a minimum so that recovery can be accomplished without undue exposure to the population. (This might require development of location and retrieval means for Earth-impacted waste packages.)
3. Potential accident conditions that could lead to uncontrolled re-entry of the waste package have to be minimized. This would be accomplished by careful selection of trajectories, by use of highly reliable vehicles and by developing space retrieval means (vehicle) for retrieval of these packages from accidental orbits beyond Earth escape.

8.2.7.6 Space Contamination

One of the concerns expressed about disposal in space is the possible contamination of space. However,

the dispersion factors achieved in event of radionuclide release in space disposal are enormous.

As an indication of the magnitude of the dilution factor achieved, assume that waste material is dropped into the sun and subsequently forced back into the solar system. Assume that it is evenly distributed through-

out the volume of a sphere with a radius equal to the earth's distance from the sun (93,000,000 miles or 150,000,000 kilometers). The volume of this sphere is $1.4 \times 10^{40} \text{ cm}^3$. (a)

By the year 2000, the total accumulation of transuranics and fission products (obtained from Volume 1 of this report series) is:

Transuranics	1,770 M Ci
Fission Products	143,000 M Ci

If the total waste accumulated to the year 2000 were uniformly dispersed in this sphere ($1.4 \times 10^{40} \text{ cm}^3$ volume), the concentration would be $1.06 \times 10^{-23} \text{ } \mu\text{Ci/cc}$. (b)

The allowable concentrations in air of several elements, from 10 CFR 20 are

Pu-238	7×10^{-14}	$\mu\text{Ci/ml}$
Cs-137	2×10^{-9}	$\mu\text{Ci/ml}$
I-129	2×10^{-11}	$\mu\text{Ci/ml}$

Assuming an allowable concentration comparable to that of Pu-238, the actual versus allowable concentration would be

$$\frac{1.06 \times 10^{-23}}{7 \times 10^{-14}} = 1.5 \times 10^{-10}$$

For comparison purposes, assume that the following quantities of strontium-90, cesium-137 and plutonium-239 which have been put into the earth's atmosphere by weapons testing⁽⁴²⁾ are dispersed within one mile of the earth's surface (actually much of this is on the surface or in the oceans now):

Strontium-90	20,000,000 Ci
Cesium-137	34,000,000 Ci (c)
Plutonium-239	400,000 Ci
Total	54,400,000 Ci

The volume within 1.6 kilometer (one mile) of the earth's surface (both above and below) is about $5.3 \times 10^{23} \text{ cc}$. (d)

- a. Volume is calculated as follows:

$$\frac{4}{3}\pi R^3 = \frac{4}{3}\pi (93,000,000)^3 = 3.37 \times 10^{24} \text{ miles}^3$$

There are 161,000 cm in a mile or $4.17 \times 10^{15} \text{ cm}^3/\text{mi}^3$. The volume is thus:

$$3.37 \times 10^{24} \text{ mi}^3 \times 4.17 \times 10^{15} \text{ cm}^3/\text{mi}^3 = 1.4 \times 10^{40} \text{ cm}^3$$

- b. Concentration is calculated as follows:

$$\frac{1,770 \times 10^6 + 143,000 \times 10^6 \text{ Ci}}{1.4 \times 10^{40} \text{ cc}} = \frac{144,770 \times 10^6 \text{ Ci} \times 10^6 \text{ } \mu\text{Ci/Ci}}{1.4 \times 10^{40} \text{ cc}}$$

$$= 1.06 \times \frac{10^{17}}{10^{40}} = 1.06 \times 10^{-23} \text{ } \mu\text{Ci/cc}$$

- c. Taken as 1.7 times that of Sr-90

- d. Volume is calculated as follows:

$$\pi D^2 \times 2 \text{ mi} = \pi (8,000 \text{ mi} \times 161,000 \text{ cm/mi})^2 \times 2 \text{ mi} \times 161,000 \text{ cm/mi} \\ = 5.3 \times 10^{23} \text{ cc.}$$

The total hypothetical concentration in this volume is thus

$$1 \times 10^{-10} \mu\text{Ci/cc. (a)}$$

This hypothetical concentration compares with the space dispersion concentration as follows:

Present weapons test isotopes
dispersed in 3.2 kilometer (2
mile) thick layer around earth $1.0 \times 10^{-10} \mu\text{Ci/ml}$

Isotopes to year 2000
dispersed around sun $1.06 \times 10^{-23} \mu\text{Ci/ml}$

The present hypothetical concentration is thus 1×10^{13} times as great as the hypothetical space concentration. Thus, there is much room for concentration factors in the space disposal system before concentrations around the earth begin to approach the present status.

A more definitive analysis of the potential for space contamination is found in Appendix E.

8.3 RESEARCH AND DEVELOPMENT REQUIREMENTS

The following section summarizes the estimated Research and Development requirements for extraterrestrial disposal of radioactive waste transuranics. Key Research and Development milestones for a space disposal program (estimated at about 20 years duration) are summarized in Figure 8.12. Costs for research and development are estimated to be about 50 million dollars plus special space flight and vehicle development specific to waste disposal. This latter

cost element was very difficult to estimate and is not estimated by NASA. The BNW study staff feels that the cost is in the order of 100 million dollars (a figure not discussed with NASA). Another disposal Research and Development cost is that for the remaining fraction of the waste. From other disposal concept analyses in this report, these costs are expected to be in the range of 50 to 200 million dollars. Total Research and Development costs for management of radioactive waste are therefore expected to be in the range of 300 million dollars. Research and Development costs are highly preliminary estimates of near-low costs required for a viable program. A larger Research and Development program would probably be pursued to optimize costs and safety. The Research and Development costs shown here are only those related to transuranic waste disposal, and do not include the significantly higher costs required for general space vehicle development (\$6.6 billion estimated for first space shuttle flights^[43]), which are assumed to be part of the general space development program. Of the specific Research and Development tasks discussed in the following sections, the space flight studies (both those general to space flight development and those specific to waste disposal), are expected to be the most time and money consuming.

a. The concentration is calculated as follows:

$$\frac{5.4 \times 10^7 \text{ Ci} \times 10^6 \mu\text{Ci/Ci}}{5.3 \times 10^{23} \text{ cc}} = 1.0 \times 10^{-10} \mu\text{Ci/cc}$$

KEY MILESTONES

1. WASTE CONTENT ESTABLISHED	7. ENCAPSULATION PROCESS DEVELOPED
2. DISPOSAL PRIORITY ESTABLISHED	8. SPACE SHUTTLE FLOWN
3. PARTITIONING PROCESS DEVELOPED	9. SPACE TUG FLOWN
4. CAPSULE DESIGN COMPLETED	10. SPACE FLIGHT FULLY DEVELOPED
5. CAPSULE DESIGN TESTED	11. SAFETY ANALYSIS COMPLETED
6. ENCAPSULATION PROCESS ESTABLISHED	12. PUBLIC RESPONSE SATISFIED

FIGURE 8.12. Research and Development Program - Space Disposal of Transuranics

8.3.1 Waste Content

At present the principal code used to establish isotopic content of different wastes is ORIGEN. The accuracy of this code is suitable for the type of investigative work done to date. However, if extraterrestrial disposal is to be used for the actinide or transuranic wastes, a more accurate picture of true isotopic content is desirable than that obtainable in present codes.

Because of uncertainty and physics effects for extended exposure in different types of reactors, existing data on the isotopic content of spent

fuel is limited. A program to analyze spent fuel from several different fuel cycles is required. This analysis is something that should be done for almost any part of the Waste Management Program.

8.3.2 Disposal Priority

The purpose of this task is to put into more clear perspective the safety incentives and problems for space disposal of certain radioisotopes. With such information available, better choices can be made, regarding their disposal needs. Though a number of analyses of the

relative potential hazards of different isotopes exist, additional information would be advantageous for the analysis of space disposal. Past comparisons of the potential hazards in space of one isotope, such as Pu-238 with another such as Pm-147, have not covered the total range of variables for full evaluation of the incentives for space disposal.

Beyond the actinides, there are differences of opinion as to whether the very long-life fission products such as iodine-129 and technetium-99 are sufficiently hazardous to require exotic disposal techniques.

8.3.3 Partitioning

Partitioning is a fundamental requirement for execution of space disposal. Therefore, the timing and successful completion of partitioning process development is critical to space disposal. It is expected that suitable partitioning processes could be developed within about five years and that a commercial plant could be in operation five years from that time. Thus the availability of a commercial partitioning operation could be expected in about 10 years.

Partitioning, or separation of certain constituents from the waste for special management, is discussed in Section 7 and those costs for partitioning research and development (3 to 5 million dollars) are not included in the total research and development costs for space disposal. The current primary possibility considered most likely for the disposal of the long-lived isotopes in space is to separate the waste stream into fission products and transuranics.

Separating the transuranic and fission product stream could provide a waste fraction for disposal in space which appears to be in reasonable form. Nonetheless, it may prove desirable to remove the curium from this product to minimize heat problems for space disposal. It might also be advantageous to extract the neptunium for conversion to Pu-238 for other purposes. If long-lived fission products such as iodine-129 are truly shown to be a long-lived hazard, it might be beneficial to remove the iodine from the fission product stream and ship it to space with the transuranics.

8.3.4 Capsule Design

Flight qualified capsules have been designed and tested in periods ranging from 1 year to 3 years. With the existing background in capsule design and available test facilities, it is expected that a suitable, fully tested capsule design could be provided within 10 to 15 years. Higher priority could perhaps improve on this schedule if necessary.

It appears possible today to design a capsule which would satisfy stringent technical and safety criteria. However, there are a number of areas in which additional data and refinement of data and methods could improve the safety and reduce the overall cost of disposal. Typical of these are:

- Criticality and Radiation Neutronics Calculations

Additional code and experimental studies are required to refine criticality data and the radiation dose rates.

- Heat Transfer Analysis

Heat transfer under a wide variety of circumstances is one of the more critical aspects of design for space disposal. The capsule must be able to withstand such diverse circumstances as deep burial in sand, re-entry, impact in space vacuum, and explosion of the rocket on the launch pad. Optimization of materials, configuration and environment will require a great deal of refinement.

- Refined Structural Analysis

Some data exist on impact resistance of large capsules, but significantly more needs to be done to make this a practical system. Structure analysis starts with the containment of helium buildup within individual containers and extends through the impact resistance required to withstand re-entry, impact on solid materials, and extreme temperature conditions.

- Impact Testing

Testing for structural considerations, shielding, burial corrosion resistance, and any other possible circumstances will be required.

The Research and Development costs may be estimated by comparison of a somewhat similar past experience with radioactive heat sources for space. The design, fabrication, process development and testing of a single design of plutonium-238 isotopic heat sources for space missions has cost on the order of \$500,000.⁽⁴⁴⁾ However, because of the much larger size and quantities of waste capsules, and the consequently greater costs for

impact in the fabrication and testing programs, it is expected that the Research and Development costs for waste packaging will be greater by at least a factor of 10.

8.3.5 Encapsulation Process Development

Encapsulation processes are expected to be similar to those existing for space applications of radio-nuclide heat sources. Some of the key steps requiring development are:

- Particle Preparation

Processes exist for the preparation and micro-encapsulation of particles. In general, however, these processes are applicable to small scale production. Developmental work is required to provide production-type processes.

- Compacting of Matrix

The development of processes is required for compacting the matrix surrounding the fuel established in the capsule design.

- Containment

The containment shell surrounding the capsule will also require some process development studies. Extension of existing fabrication techniques could likely be used, particularly if the shell is stainless steel.

All of the required steps for an encapsulation process have been tested, although not necessarily on the same materials that will be used. The application of existing processes to new materials has been successfully demonstrated in the past in periods from a few months to a year. Pilot plants for encapsulation of

radioactive materials have been built in periods of 5 years or less.

As an example of the program costs and times incurred in this type of development, a radiopromethium heat source capsule was designed,⁽⁴⁵⁾ the fabrication process developed and the source fabricated and tested in less than one year for less than \$100,000. Testing for this capsule included rocket sled testing at terminal velocity and actual drop testing.

In Fiscal Years 1964, 1965, and 1966 a program for the development of cermet fuels which involved the spheroidization of uranium dioxide fuel particles, coating the particles with tungsten and consolidating them in the form of complex fuel shapes was conducted for NASA and the AEC. The fabrication process development portion of this work totaled \$780,000 over the three year period.⁽⁴⁶⁾

Again, because of the increased size, quantity, and complexity of transuranic waste capsules and the need for greater integrity, more extensive process development by factors of 5 to 10 than that required for smaller capsules is expected to be required for nuclear waste capsules.

8.3.6 Ground Transportation

While conventional ground transportation systems will probably be applicable, some minor studies will be required to confirm analyses.

8.3.7 Space Flight

As discussed in Section 8.2.4, there are vehicles which could be used today. Since the space shuttle

is a vehicle which is likely to be used for waste disposal, operational use of the space shuttle is anticipated. It is expected that the space shuttle will be operational in the early 1980's.

The space tug is required for economical space disposal. A firm schedule for an operative space tug has not been established. However, NASA personnel have indicated that it can be operable by 1985. It could probably be operational prior to this time if there was sufficient incentive.

Developmental work in this area is likely to be all under NASA. Research and Development costs or times for this category are difficult to estimate. However, it is believed that development costs will be in the order of hundreds of millions of dollars beyond the currently planned space shuttle development costs. Some of the primary studies are expected to be:

- Disposal Trajectory

The determination of a disposal trajectory which has no significant unknowns is one of the major technical needs of this total program. Development in this area might involve propelling several waste containers into the sun and monitoring for detectable transuranics which might return during the next 5 to 30 years. Another experimental program could involve sending capsules in an earth escape trajectory or a high earth orbit and monitoring and analyzing their course. If such a path could be followed for 20 to 100 years it could increase the reliability of predictions on the future of such capsules.

- Vehicle Development

NASA analysis to date has been based on existing concepts. Some development would undoubtedly be required to adapt and optimize present concept vehicles to waste disposal.

- Advanced Systems

Advanced systems such as particle acceleration and solar sails have been examined only briefly.

If space disposal is considered to be practical with current systems, it is probable that significant economic gains could be achieved through the development of advanced systems. While potential safety improvements are not apparent, there is some probability that additional safety could also be achieved through the utilization of advanced systems.

It is therefore expected that a much more detailed analysis of advanced systems would be pursued before space disposal can be implemented. If advanced systems show significant promise, they could warrant a complete and extensive development program comparable to that for conventional vehicles described above.

8.3.8 Economic Analysis

The economic analyses of space disposal alternatives will involve both a detailed engineering assistance study and a broad overview cost-benefit analysis. The engineering assistance work will provide continuous correlation of the economic effects of various technological approaches.

8.3.9 Safety Analysis

The principal constituent of safety analysis is the execution of

the space flight including potential accident considerations. Some other studies are required for the additional processing which must be done to put material in the form for flight.

The safety aspects of flight will include the effect of earthly accidents such as explosion on the launch pad, failure to achieve orbit, or re-entry from a partial orbit. Space flight effects will include establishing realistic probabilities and consequences for malfunctions or accidents.

Safety studies will also include estimation of effects of space disposal on our environment.

8.3.10 Public Response

Many arguments about nuclear waste are more emotional and philosophical than they are technical. A serious study of the emotional and philosophical aspects of space disposal of waste must be aimed at developing concrete conclusions for positive action. Action would involve communication interchange and education of the public and the technical community regarding the concerns of both.

If space disposal is to become viable, national and international policy must be examined and new policy established.

8.4 TIME REQUIREMENTS FOR COMMERCIAL OPERATION

The time required to establish space disposal as a commercial operation is dependent principally on the development studies discussed in the preceding section. Thus the time for commercial operation could be in the order of 20 to 25 years from the time

of actively starting Research and Development; this includes up to 5 years from full development to full operation.

Should experimental flights such as impact into the sun or monitoring a vehicle enroute to Pluto (20-year flight time) and beyond be considered necessary to the establishment of policy, then the time for commercial operation could be extended beyond 20 years.

8.5 CAPITAL AND OPERATING COSTS

The costs of transuranic waste disposal by extraterrestrial means include three main components--partitioning, encapsulation, and flight. Basic capital and operating costs in 1974 dollars for waste management are developed in this section, and are converted to units of nuclear electrical power and nuclear fuel in Volume 1 of this report series.

Partitioning costs summarized below are taken from the separate section of this report which discusses partitioning in depth. Encapsulation costs are discussed below. Flight costs were discussed previously in part 8.2 of this report (technical feasibility), and are summarized here. All additional cost data were developed in Volume 1 of this report and are summarized here.

8.5.1 Partitioning

Partitioning costs incremental to reprocessing costs without partitioning are estimated to be:

Transuranium Elements (U and Th removed) in a package along with 1% of the fission products cost \$15,000/MT of fuel.

Transuranium Elements (U and Th removed) in a package along with 0.1% of the fission products cost \$20,000/MT of fuel.

8.5.2. Encapsulation Costs

For a reference reprocessing plant capacity of 1825 MT/year of LWR fuel, the waste transuranic throughput was calculated to be 1,288 kg/year. To achieve a reasonable plant size it is assumed that the encapsulation plant will handle the transuranium waste from two 1825 MT/year reprocessing plants, or approximately 2,600 kg/year.

Using the process of Figure 8.9, preliminary encapsulation costs were developed and are detailed in Appendix F. The total cost of encapsulation and heat shield is estimated at \$4,700/kg of actinide waste, or \$3,300/MT of original LWR fuel. The estimate assumes 1% fission product contamination, with heavily shielded hot cells used for each process step. Since with either 1% or 0.1% of fission products in the transuranics, most of the work would require remote operation in hot cells, the costs would be similar for the 0.1% case.

For checks of the order of costs, the fabrication cost of reactor fuel elements containing high exposure plutonium has been estimated at \$52 to

\$55/kg of fuel material (uranium oxide and plutonium oxide) in a one-ton-per-day plant.⁽⁴⁷⁾ The cost of purifying and encapsulating cesium-137 at \$0.05/curie⁽⁴⁸⁾ is \$4350/kg.^(a)

Direct labor and equipment requirements are estimated to be, 17 personnel (total cost \$170,000/yr) and \$675,000, respectively. These are detailed in Appendix Table F. 2. The manpower and equipment requirements are based on experience in the spheroidization, coating and encapsulation of materials such as plutonium, promethium, and polonium. These costs are detailed in Appendix Table F.3, based on the material make-up of a capsule as given in Appendix Table F.4.

Direct material costs are estimated to be \$340/kg transuranic. The costs for materials could be somewhat higher than those estimated because of unique form or purity requirements. However, a great effect on the total encapsulation cost is not expected.

Indirect manufacturing expense includes items such as inspection, testing, and maintenance. This is estimated to be conservatively high at 100 percent of direct labor cost.

Overhead costs are calculated at 100% of direct labor plus indirect manufacturing expense.

Building capital costs are estimated by comparison with the Waste Encapsulation and Storage Facility (WESF) recently constructed at Hanford.⁽⁴⁸⁾ This facility is designed for the annual encapsulation of 30 megacuries each of cesium-137 chloride in 60,000-curie packages, and strontium-90 fluoride in 150,000-curie packages. This radioactivity amounts to about 340 kilograms of cesium-137 and 210 kilograms of strontium 90^(b) annually.

The completion was late 1973 at a cost of \$10.75 million.

Using a 0.6 power scaling factor to convert from 530 kg/yr of (Cs+Sr) to 2600 kg/yr of transuranics, would provide a cost for the transuranic plant of about \$27,000,000.^(c) Because of the additional complexity of the transuranic waste encapsulation process, a figure of \$40,000,000 is used for plant capital cost.

As a comparison of the size order of these costs, the capital cost of a one ton/day high-exposure plutonium mixed-oxide fuel plant was estimated at \$6,400,000 in 1966.^(49,50)

8.5.3. Costs for Space Transportation

This section was outlined by NASA Lewis⁽¹²⁾ and was taken from Section 8.2. Minor modifications in their report have been made to conform to the format of this report.

a. Using 87 curies of Cs-137 per gram.

b. Using 87 curies of pure Cs-137/g and 142 curies of pure Sr-90/g.

c. $\text{Cost } 2600 \text{ kg} = (\$10,750,000) \left(\frac{2600}{530} \right)^{0.6} = \$27,000,000.$

The vehicle costs, launch costs and operations costs are essentially a function of the number of vehicles, the launch rate, and the destination. Two representative destinations were estimated; high earth orbits and solar escape. The Kennedy Space Center was used as the reference launch facility. Beyond about the year 1990, significant additional space flight facilities will be needed.

The transportation costs presented below include technical support and operations.

8.5.3.1. Launch Costs

The most cost effective vehicles were selected from Table 8.11 for determining the space transportation costs.

- High Earth Orbit or Solar Orbit

The most cost effective vehicle was the Shuttle with a Centaur of optimum size (using approximately 17,240 kg of propellant). The launch cost for this vehicle is estimated at \$16.3 million (\$10.5 million for the reusable Shuttle launch and \$5.8 million for the optimum expendable Centaur). These costs are based on 100 total flights and not more than 40 flights per year. The Shuttle launch cost of \$10.5 million includes propellant costs and operational costs. If the flight rate is increased from 40 to 140 per year, the unit operational cost per flight would be reduced to approximately 75-90% of that for 40 flights/yr.

- Solar System Escape

This mission requires one Shuttle per Tug and at least two Tugs per

nuclear waste payload. Table 8.12 indicates that the most cost effective method for the solar escape destination involves perigee propulsion and consists of two Shuttles, a reusable and an expendable Tug for a total cost of \$28.8 million dollars per mission. The same conditions on launch rate apply to this cost as applied in the Shuttles for the high earth orbit destination above.

8.5.3.2 Ground Facilities Costs

Many of the existing facilities at Kennedy Space Center could be used on the basis of 20 flights per year. Beyond that, additional facilities would be required. In addition to launch and operations facilities, a new facility for receipt and inspection of nuclear waste packages and for storage of packages in a controlled environment would be required. Cost of this facility is estimated at approximately \$4 million.

As the number of launches per year increases past 20, additional facilities are needed. Projecting to 100 launches per year, an additional \$140 million ($\pm 20\%$) in mobile launchers, a new maintenance and checkout facility, new crawler and maintenance facility, a new launch pad and a new solid rocket booster disassembly facility would be needed.

After the year 2000 some other arrangement for launching facilities may be required since it would be difficult to handle more than 120 to 140 launches per year at Kennedy Space Center.

8.5.3.3 Total Space Transportation Costs

After the launch, other facilities come into use, such as tracking and monitoring stations and recovery teams and possible facilities for handling the Shuttle at other locations. Most of these facilities are assumed to already be in existence and would probably need only modifying. It would be expected that these modifications would be an order of magnitude less than the costs for the facilities discussed above.

The total transportation costs for both representative destinations are presented in Table 8.13 for launch rates of 40 per year. The facility cost per launch, based on a 30-year time period, increases from less than 0.1 to about 0.2 million dollars if the launch rate goes up by 100 per year, or a small amount.

8.5.4 Cost Summary

The estimated cost to put material into a high-earth orbit or solar orbit is \$1,950 per kilogram of gross payload, as shown in Table 8.14. The comparable estimated cost to escape the solar system to deep space is \$8,800 per kilogram of gross payload. Encapsulation will increase the weight of transuranic actinide wastes and thus the unit costs by factors of about 15 to 30.

Based on the above figures, approximate costs which might be expected for space disposal of transuranics with 0.1% fission product contamination are summarized in Table 8.15.

These costs convert to 0.2 mills/kW-hr_e for high-earth orbit and 0.5 mills/kW-hr_e for solar system escape. These costs, when discounted to the time of reprocessing and allowing 0.04 mills/kW-hr for disposal of the remaining waste fraction (fission products) are 0.15 Mills/kW-hr and 0.34 mills/kW-hr, respectively. When compared with costs for the production of electricity of about 10 mills per kilowatt hour, the cost of space disposal of waste transuranics is thus likely to be less than 5% of the cost of producing electricity. Additional costs would be incurred in the management of the remaining fraction of waste which is not disposed of into space. Based upon analysis of other potential means of disposal elsewhere in this report, this incremental cost should not exceed 1% of the cost of nuclear electricity.

8.6 PUBLIC RESPONSE, POLICY AND ENVIRONMENTAL CONSIDERATIONS

8.6.1 Public Response

Public reaction to the concept of space disposal is unknown but is expected to be mixed. The possibility of space disposal has been suggested many times in the past, but little factual information has been presented which the public could use in forming an opinion.

The fact that space disposal has the potential to remove certain radioactive waste constituents permanently off the earth (very distant from man's

TABLE 8.13. Space Transportation Cost for Disposal of Transuranic Waste

Basis: Reusable Shuttle 40 Payloads/Year

	<u>Cost/Payload</u> <u>\$Millions</u>
Facilities and Flight Preparation	< 0.1
<u>High Earth Orbit</u>	
Shuttle (1)	10.5
Centaur (Optimum) (1)	<u>5.8</u>
Total	16.4
<u>Solar System Escape</u>	
Shuttle (2)	21.0
Tug-Expendable (1)	6.0
-Reusable (1)	<u>1.75</u>
Total	28.8

TABLE 8.14. Estimated Space Transportation Cost for Disposal of Transuranic Waste

Basis: Space Shuttle is the base vehicle.
Transuranium elements contain 0.1
or 1% of the fission products.
Capsules are shielded for 1 Rem/hr
from surface of package.

<u>Percent Fission Product</u> <u>in Waste</u>	<u>High Earth Orbit</u> <u>or Solar Orbit(a)</u>		<u>Solar System Escape</u>	
	<u>1.0</u>	<u>0.1</u>	<u>1.0</u>	<u>0.1</u>
Total Payload/Transuranic Weight, kg/kg	8400/288	8400/447	3270/113	3270/191
Transportation Cost, \$/mission	16.4×10^6	16.4×10^6	28.8×10^6	28.8×10^6
Transportation Cost, \$/kg of Payload	1950	1950	8800	8800
Transportation Cost, \$/kg of Transuranics	56.8×10^3	36.5×10^3	254.0×10^3	150.0×10^3

a. Transuranic waste is divided into three equal packages.

TABLE 8.15. Cost for the Disposal of Transuranium Elements in Space

	<u>Cost, \$per kg Transuranic</u>		<u>Cost, \$per MT fuel</u>	
	<u>High Earth Orbit</u>	<u>Escape from Solar System</u>	<u>High Earth Orbit</u>	<u>Escape from Solar System</u>
Partitioning	14,000	14,000	20,000	20,000
Encapsulation	4,700	4,700	3,300	3,300
Space Flight Cost (payload= 20 x actinide wt)	<u>36,500</u>	<u>150,000</u>	<u>26,000</u>	<u>107,000</u>
Total	55,200	168,700	49,300	130,300

environment or population) is expected to be a strong positive factor.

The principal objections of critics of space disposal appear to be concerned about high cost, unfavorable energy balance, and safety during launch and space flight. Questions regarding cost and energy are relatively straight forward to determine, and therefore could be amenable to general public understanding. Questions regarding safety are much more complex and emotional, and therefore would be expected to be more difficult to achieve relatively uniform favorable public response.

Other items which we might expect to be of concern to the public are the proof of high certainty that events will proceed as planned and philosophical-technical questions regarding sending materials into "unknown" areas.

8.6.2 Policy Conflicts

International and National policies which could impact extraterrestrial disposal are discussed in section 3 of this report series and are reviewed briefly here.

Any of the extraterrestrial disposal concepts would come under the provisions of the International Treaty on Outer Space, which was ratified by the United States in 1967. Article VII of this treaty provides that "Each State Party to the Treaty that launches or procures the launching of an object into outer space, including the moon and other celestial bodies, and each State Party from whose territory or facility an object is launched, is internationally liable for damage to another State Party to the Treaty or to its natural or juridical persons by such object or its component parts on the Earth, in air space or in outer

space, including the moon and other celestial bodies." Any launch operations or mission aborts could entail international liabilities under this treaty. An orbit degradation which caused a capsule to eventually return to earth might also be subject to these provisions.

Liability for damage from any of the space concepts could be incurred under the terms of the 1972 United Nations' Convention on International Liability for Damage caused by Space Objects. The United States has not yet ratified this treaty, which defines in some detail the responsibilities of countries launching objects into space. For example, Article XII of the Convention specifies "...such reparation in respect of the damage as will restore the person, natural or juridical, state or international organization on whose behalf the claim is presented to the condition which would have existed if the damage had not occurred." While this convention will not materially extend the liability already accepted under the Treaty on Outer Space, it will, if ratified, mean acceptance of a precise method for defining, assessing and paying for space-related damages.

Article 25.1 of the International Convention on the High Seas provides that "Every State shall take measures to prevent pollution of the seas from the dumping of radioactive waste, taking into account any standards and regulations which may be formulated by the competent international organizations." Any radioactivity which reached the ocean from a failed space flight might be deemed in violation of this convention.

Any contracting party to the convention can request a revision five years after it has entered into force. The request for revision is considered by the General Assembly of the United Nations.

The International Nonproliferation Treaty, to which the U.S. is a party, could conceivably affect any ultimate disposal concept. The basic objective of the treaty is the limitation of nuclear weapons through the effective control of the flow of source and special fissionable materials.

Article III of the treaty states in part: "Procedures for the safeguards required by this article shall be followed with respect to source or special fissionable material whether it is being produced, processed or used in any principal nuclear facility or is outside any such facility. The safeguards required by this article shall be applied on all source or special fissionable material in all peaceful nuclear activities within the territory of such State, under its jurisdiction, or carried out under its control anywhere." While mixed fission products and transuranic elements might be considered outside the scope of this treaty on the technical basis of cost and risk of separation of the fissionable materials, any concept dealing with separated transuranic element disposal would almost certainly fall under the agreements of this treaty. It would seem, then, that those procedures and operations would have to conform to International Atomic Energy Agency safeguards. Likewise, they would be subject to the appropriate international inspection and observation.

Any amendment to the treaty must be approved by a majority of all parties to the treaty and by all nuclear weapon states party to the treaty. Twenty-five years after the treaty is in force a conference is to be held to decide whether it should be extended indefinitely or renewed for a fixed time period. This decision will be made by a majority of the parties to the treaty.

Each party to the treaty has the right to withdraw if it decides that extraordinary events have "jeopardized the supreme interest of its country."

8.6.3 Environmental Considerations

The principal environmental considerations of the extraterrestrial disposal system will be in regard to the space flight operation. There will be some side effects such as the additional processes required by partitioning and encapsulation. There will be additional fabrication of space flight vehicles and additional production of rocket fuel beyond that required without space disposal of nuclear waste constituents. There will be additional transportation requirements. If a disposal plan which involves only the disposal of transuramics is used, disposal would still be required for the fission products in the waste. However, it is possible that the fission products would be handled in a manner different from the combined waste since the lifetime requirements are much different.

The incremental environmental effects with reprocessing plants, the

rocket fabrication plants and fuel production plants should not be unique.

The principal effects considered for the environmental impact of space flight are land, resource use, water, air, ecologic impact, aesthetic impact and transportation impacts.

Land

The existing Kennedy Space Center site should be adequate until about the year 2000, assuming disposal of 10-year-old waste. Beyond that time, additional land and facilities roughly comparable to those at the Kennedy Space Center will be required about every 10 years. The expected effect of the Shuttle system on the Kennedy Space Center has been evaluated by NASA. Much of the discussion below is extracted from their preliminary analyses.⁽⁵¹⁾

Large areas of land surrounding the launch facilities are required for supporting services and for a buffer between these activities and the surrounding community. At the Kennedy Space Center, maintenance of environmental stability and planned multiple land use have been stressed. For instance, under an agreement with the Bureau of Sport Fisheries and Wildlife, the boundaries of the Merritt Island Wildlife Refuge and the Kennedy Space Center are now co-extensive. This agreement provides that the Bureau, subject to certain conditions, exercise primary administration over all property (except the Space Program facilities) for all purposes unrelated to the Space Program. Legislation has also been introduced which, if enacted, would allow the

joint use of the Space Center area north of the Haulover Canal by the National Park Service as a National Seashore Park (rather than as a part of the present Wildlife Refuge). The multiple land use was considered in the evaluation of candidate Space Shuttle launch and landing sites, and activation of the selected building sites.

Future activity will continue to stress land-use patterns compatible with the use of the area as a Wildlife Refuge and/or a National Seashore Park.

Resource Use

With space disposal, there would be potential adverse effects from the ultimate loss of some materials from the Earth. For example, the world reserves of tungsten are 1,200,000 metric tons, with a potential world resource of 51,000,000 metric tons.⁽⁵²⁾ At a launch rate of 100 capsules/year, over 100 metric tons per year of tungsten would be irretrievably lost to space.

Water

In the space flight system, with planned recovery of all elements of the Space Shuttle except its fuel tank, the potential impact of the program flight aspects on water quality is related to:

- On-pad accidents and propellant spills which may result in run-off of propellants to local drainage systems.
- In-flight failures which may result in vehicle hardware and propellant landing in the ocean.
- Controlled re-entry of spent booster and Shuttle hydrogen and oxygen tanks.

- Construction and operation of facilities.

Provisions such as dikes and catch basins are made for containing on-pad spills and disposing of the spilled propellant without major contamination of the water (or air) environment. Infrequent on-pad vehicle failures would normally be expected to result in a fire that consumed most or all of the propellants. Thus, they are discussed in the section on air quality. Any unconsumed propellant would be treated in the same way as a spill.

Potential sources of pollutants to the marine environment and the major pollutants would be: hardware, solid propellants, liquid propellants, lubricants, and hydraulic fluid-hydrocarbons.

Possibilities of water pollution are primarily associated with water soluble, toxic materials which may be released to the water environment. Rocket propellants are the dominant source of such materials. Impact of the Shuttle fuel tank would release liquid hydrogen and liquid oxygen which would burn or evaporate rapidly into the atmosphere. Toxic materials contained in the Shuttle would be returned to the launch site. However, if the Shuttle were forced to abort to a water landing, these materials would enter into the water. The quantities from these infrequent occurrences would dilute to non-toxic levels of concentration within the area affected by the emergency landing.

The ammonium perchlorate in solid propellants is mixed in a rubber binder and would thus dissolve slowly in

water. Toxic concentrations would be expected only in the immediate (within a few feet) vicinity of the propellant, if they occur at all.

Oils and other hydrocarbon materials which are essentially immiscible with water, if released, may float on the surface of the water. Quantities of hydrocarbons used are small.

Jettisoned or re-entered hardware will corrode and thus contribute various metal ions to the environment. The rate of corrosion is slow in comparison with the mixing and dilution rate expected in a marine environment, and hence toxic concentrations of metal ions are not expected to be produced. The miscellaneous materials (e.g., battery electrolyte, hydraulic fluid) are present in such small quantities that, at worst, only extremely localized and temporary effects would be expected. In the immediate off-shore areas at Kennedy Space Center there is ample current to ensure dispersion of these materials.

The ground water sources in the area have been examined; no adverse effects are anticipated on the ground water.

In 1970, Kennedy Space Center used approximately 3.8 million liters (one million gallons) per day out of a peak of 100,000,000 liters (26 million gallons per day supplied by the Cocoa Municipal Water Supply. In 1980, it is predicted that Kennedy Space Center will require about 3-4 million liters/day (0.9 million gallons/day) out of an estimated output of 130 million liters/day (35 million gallons/day). The fresh water requirements of Kennedy Space Center for the operational period are pro-

jected to be less than that required for the Apollo program and will amount to only a relatively small portion of the overall demand for other resources.

The chief potential for pollution on surface water is the propellants. In a normal launch essentially all propellants or propellant products are injected into the atmosphere, and the hardware, except for the propellant tank, is recovered. In the case of an in-flight failure in the early stages of flight, the booster and propellant tank would probably impact intact. The shuttle would be expected to separate intact and return to the launch site.

Handling of propellants at the Kennedy Space Center will follow the same procedures established and proven by the successful operations for the Apollo and other program efforts. The Kennedy Space Center has not had a fuel or oxidizer spill in excess of 3.8 liters (one gallon) except for a spill that occurred in 1969 during preparation for launch of Apollo/Saturn 505. Approximately 20,000 liters (5,300 gallons) of propellant were captured in the spill pond provided for the purpose, collected and turned over to the Air Force to dispose of in accordance with accepted and established procedure.

The construction of Shuttle facilities and their operation is not expected to affect aquatic areas.

Air

The quantity of combustion products released during a flight is shown in Table 8.16. The result of most importance is the history of the

TABLE 8.16. Combustion Products of Concern Emitted by the Space Shuttle into Selected Atmospheric Layers

Atmospheric Layer	Altitude Range Kilometers	Combustion Product	Single Mission Quantity Emitted, kg	
			Solid Rocket Motor	Shuttle
Surface Boundary Layer	0-0.5	CO	37,200	
		CO ₂	6,600	
		HCl	31,900	
		Cl ₂	92	
		Al ₂ O ₃	43,300	
		H ₂ O	15,900	19,500
Troposphere	0.5-10	CO	113,000	
		CO ₂	20,100	
		HCl	96,900	
		Cl ₂	278	
		Al ₂ O ₃	132,000	
		H ₂ O	48,100	62,200

concentration of the pollutant at ground level downwind of the launch point should wind currents move a portion of the cloud to the ground. In all normal launch cases, the peak concentrations are well below the applicable maximum allowable 10-minute concentration levels to industrial workers.

Exhaust cloud concentrations of CO, HCl, and Al₂O₃ have been calculated as a function of distance downwind of the launch pad for an abort with burning of solid rocket propellants. Peak concentrations are about 5 to 10 times larger for this case than for the normal launch, but would still be below the 10-minute maximum allowable

concentration levels to industrial workers within the controlled area.

At some locations downwind the recommended limits for 10- and 60-minute exposures to hydrogen chloride for the general public may be exceeded as the result of a pad abort or fire. The time dependence of the concentration at these locations is such that the time-averaged concentration is less than the recommended limits. The action taken to assure protection of the public will depend on exposure measurements within these zones.

The National Academy of Sciences/ National Research Council Report summarizes the known effects of HCl on wildlife. There would be no effects

of even the predicted peak ground level concentrations of about 9 parts per million for a pad abort.

NASA has analyzed the lower atmospheric effects and upper atmospheric effects on rain; no adverse analyses of pollutant effects of Shuttle operations are foreseen.

During rain possible precipitation scavenging of hydrogen chloride from the solid rocket exhaust cloud has been analyzed by NASA in an over-land trajectory. If possible harmful effects of rain containing hydrogen chloride after the launch were anticipated, the launch would be postponed.

Ecologic Impact

Except for the effect of noise on birds and animals, the major impacts on the ecology have already been suffered during construction of the Kennedy Space Center. During additional construction comparably small ecologic effects would be expected for each comparable construction addition.

Aesthetic Impact

The visual impact on the community will be small since the facilities are in a reservation. The high visual impact occurring at launch is of short duration.

Noise

The major source of noise associated with the Space Shuttle program will be the noise generated by the rocket engine exhaust flow during engine tests and launches and that of the sonic boom. The nature of the rocket engine noise may be generally described as intense, of relatively short duration, and spectrally composed of predominately low frequency energy. Noise pollution as a result

of construction projects is not anticipated to be a problem, because all projects will be carried on within the boundaries of the Kennedy Space Center or equivalent, which includes a large buffer zone for rocket launches.

Space Shuttle operational personnel within the launch area will be protected by personnel protective equipment or by isolation so that reasonable limits will not be exceeded. Throughout the Apollo/Saturn V Program vehicles generated frequencies and intensities of the same order as those predicted for the Space Shuttle. Operational Observers are stationed 3,500 meters (11,500 feet) from the launch pad in a small enclosure, and emergency crews are located approximately 550 meters (1,800 feet) from the launch site in standard armored personnel carriers. None of these personnel has suffered injury.

Structural damage is possible with low frequency, high-intensity noise. Therefore, structures within the controlled area will be designed to withstand the noise environment to which they are to be exposed.

For uncontrolled areas, a general noise exposure criterion of a maximum overall sound pressure level of 115 decibels, referenced to $0.00002 \text{ Newtons/m}^2$, for both man and structures has been established by the Launch and Landing Site Review Board with respect to rocket engine noise. Normally the acoustic energy which propagates into this region is of low frequency content, i.e., 100 Hertz and below.

The Kennedy Space Center launch site meets the above criterion. The center has existing land area that is adequate for the Space Shuttle zones, and the noise generated will not affect the local area populace to any higher degree than previously experienced.

The runway orientation for Shuttle and logistics aircraft landing is such that overflights of populated areas and interference with existing civil air routes are minimized. Noise pollution affecting neighboring communities is estimated to be significantly less than that emitted from commercial jet carrier operations at the neighboring Titusville/Cocoa Airport. The Kennedy Space Center buffer zone extends well into the Indian River west of the proposed new runway where the Shuttle is planned to land using commercial-type jet engines.

Space Shuttle sonic booms occur during the ascent after launch, during booster descent after separation, and during Shuttle descent after re-entry from orbit. The most severe booms result when a vehicle engages in certain types of maneuvers that tend to amplify the overpressures. These maneuvers cause focusing of the sound pressures over a very small but predictable area on the surface. Focus on populated areas can be avoided by properly programming the flight maneuvers of the vehicles.

Depending on mission orbit, return opportunity, and maneuverability, Shuttle re-entry sonic boom, however, may occur over land. The sonic boom characteristics for the returning shuttle have been calculated based upon extensive analytical work throughout NASA and on an exhaustive experimental program conducted by the Ames Research Center.

In summary, the low overpressures, infrequent occurrence, and public awareness of sonic boom resulting from Shuttle return to the Kennedy Space Center lead to the conclusion that no significant environmental effect to people, structural and natural condition results. Some disturbance to eagles, ospreys and other wildlife may occur. The degree of disturbance is expected to be minor, but can be evaluated more fully and accurately when Space Shuttle launchings begin in the existing planned space program.

Transportation

The transportation effects would be considerably greater from the large space launch components than from the "payload" capsules carrying the transuranic waste constituents. In this environmental consideration, as in all others, it should be remembered that only the disposal of the transuranic elements in waste are being considered here. If space disposal is used, all of the environmental considerations discussed for disposal of the remaining waste constituents will also be incurred.

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SECTION 9: TRANSMUTATION PROCESSING

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9.0 TRANSMUTATION PROCESSING/ELIMINATION

A possible approach to the management of radioactive waste is the use of nuclear processes themselves to change (transmute) the toxic long-lived radioactive constituents in high-level waste into short-lived radioactive or nonradioactive isotopes. Transmutation is generally defined as any process whereby a nuclide absorbs or emits radiation and is thereby changed into another nuclide. It is proper that in the study of waste management alternatives consideration be given to the possibility of eliminating the waste by transmutation. Since many of the toxic materials were formed from transmutation processes, it is conceivable that they can be destroyed by the same processes. More practically, the transmutation process can accelerate the decay rate of radioactive waste by converting long-lived radioisotopes to other isotopes which have shorter decay times. If this can be achieved, the quantity of waste containing long-lived radionuclides could be reduced significantly and the time required for safely storing treated radioactive waste may be substantially shortened.

The transmutation concepts considered in this study were limited to managing high-level radioactive waste from nuclear power. All constituents of high-level waste for example, short-lived fission products, are not amenable to treatment by transmutation processing/elimination (hereafter referred to as transmutation).

Therefore, this study mainly focused on the problem of reducing the long-term toxicity of high-level waste.

This section of the report describes known processes which could potentially be used for transmuting toxic isotopes into nontoxic isotopes. The study is extensive enough to determine which fundamental processes are theoretically not possible in light of current technology. At the same time the study was not exhaustive in that little or no optimization of possible processes was attempted. Thus, these initial conclusions of technical and/or economic infeasibility of transmutation should not dismiss future research entirely since it is the only method that has the potential for accelerating natural aging processes of eliminating waste.

9.1 TRANSMUTATION WASTE MANAGEMENT SYSTEMS

On the basis of an extensive literature review and private communications with other professional investigators the various ideas proposed for transmuting nuclear waste were collected. The waste management transmutation ideas were divided into four categories based upon the type of physical device conceived for accomplishing the transmutation. A tentative description of each concept system was developed and criteria were developed as a basis for judging technical feasibility. The four concepts are the use of:

- 1) Accelerators
- 2) Thermonuclear Explosives
- 3) Fission Reactors
- 4) Fusion Reactors

Waste transmutation processes considered in concept (1) involve the use of several different nuclear and photonuclear particles as sources for accomplishing transmutation. In particular, charged particle bombardment employing both protons and electrons has been studied. Photon radiation, both bremsstrahlung and stimulated emissions, has also been investigated. Finally, neutron radiation is a candidate transmutation source. The study of concepts (2) through (4) were considered only on the basis of using these devices as neutron sources.

9.1.1 Overall Criteria

To establish the relative merits and specific technical feasibility of the various approaches, a number of criteria were developed and applied to the proposed concepts. These include : overall energy balance, overall waste balance, specific transmutation rate, and total transmutation rate. Throughout this section, the merits derived from transmutation are viewed in terms of toxicity indices,^(a) permitting easy comparison of alternatives.

9.1.1.1 Overall Energy Balance

Overall energy balance is a fundamental criterion which applies to all advanced concepts. For a process to be practical it must not consume more energy than was originally obtained when the waste was created. This criterion is strictly valid only so long as fission power reactors are contributing significantly to the energy economy. If fission reactors are furnishing the energy for the transmutation process and if this criterion is not satisfied, as much waste is being created as destroyed. If fission power were no longer necessary, this criterion is no longer one of feasibility but of practicality. It might then be acceptable to the public to violate this criterion if the energy source were sufficiently plentiful and environmentally clean. In considering two processes, the one which consumes the least energy per transmutation is the more attractive for this criterion.

9.1.1.2 Overall Waste Balance

The second criterion applied to the various transmutation concepts was the overall waste balance. For a process to be feasible it must remove more long-term toxic waste than it creates. This criterion is of

a. Toxicity index is defined as the base 10 logarithm for the amount of air or water in cubic meters required to dilute the present amount of a given isotope to levels defined in the Code of Federal Regulations (10 CFR - Part 20) as the maximum permissible concentration. The toxicity index provides only an approximate comparison of radiological risk, since it does not allow for accumulation or reconcentration of a nuclide in environmental media, nor for the total impact of a number of nuclides. For limited comparisons, it is an acceptable alternative to dose calculations if used with caution.

particular concern in transmutation because the process is similar to that which originally created the waste.

The assurance that this criterion is met depends upon a rather detailed analysis. Some of the elements necessary for this analysis are knowledge of the nuclear reactions used and energy and type of source nucleon. In addition, detailed knowledge of the proposed mother-daughter chain must be available. In summary, it needs to be demonstrated that selection of a particular transmutation path does not result in the creation of descendant isotopes which are ultimately of comparable toxicity to the waste itself.

9.1.1.3 Specific Transmutation Rate

The third criterion applied to the various concepts was the specific transmutation rate of the process. Any isotope in waste which is a candidate for transmutation has a natural rate of decay. Any transmutation process has a rate of transmutation which is the product of the nucleon flux times the probability of the specific transmutation reaction. (The latter parameter is known as the cross section of the reaction.) For a transmutation process to be feasible the transmutation rate must be of the same order as the natural decay rate so that the actual removal rate is increased. To be a highly successful process, the transmutation rate should be several times that of the natural decay rate.

9.1.1.4 Total Transmutation Rate

For a process to be feasible it must be capable of transmuting a significant fraction of the expected inventory of radioactive waste. In addition, adequate transmutation sources must be available in sufficient quantities over the total proposed history of the process for the process to be feasible. Here, the total rate of the power industry to transmute its own waste is considered. For example, if neutron transmutation is the process under consideration, this criterion is concerned with the available neutron supply as a function of time.

Application of this criterion requires the examination of the available sources for transmutation as a function of time. If alternative sources cannot be shown to be available in sufficient quantities when primary sources are no longer available, then transmutation will not be feasible.

9.1.2 Description Of The Concept Systems

As mentioned above, the ideas proposed were categorized in terms of accelerator devices, thermonuclear explosive devices, fission reactors, and fusion reactors. A description of each concept system was developed. In the course of study it was concluded that the use of accelerators and thermonuclear explosive devices was not feasible. Therefore, only brief descriptions are given for these two concept systems.

9.1.2.1 Accelerator Devices

The concept of this system, depicted in Figure 9.1, has a partitioning stage and a target fabrication stage between reprocessing and transmutation using the accelerator. The bulk high-level waste from a reprocessing plant, either in aqueous or solid forms, could not be effectively transmuted using accelerator devices. The waste partitioning facility shown in the figure is to separate the bulk high-level waste by partitioning the waste into separate chemical element streams or a mixture thereof. The feasibility study of transmutation of only the cesium and strontium fission product components of the waste by the neutrons from a spallation accelerator has indicated that one accelerator would be required for every three or four fission reactors of the same thermal power. If other components of the high-level waste were also competing for these neutrons, the number of required accelerators per

fission reactor would make the process no longer feasible. The concept would fail both the criteria of energy balance and waste balance. Therefore, the waste would have to be processed (i.e., partitioned) to chemically separate the waste into fractions with different characteristics.

After irradiation by the accelerator neutrons, the target material would contain some fraction of the original radioactive waste since it would not be feasible to irradiate until 100 percent of the waste isotopes had undergone transmutation. In addition, the target material would contain highly radioactive short half-life daughter isotopes and radioactive elements due to transmutation in any structural material which contains the sample. These residual radioactive materials would be transported to the retrievable waste storage facility for decay and ultimate disposal using other means.

FIGURE 9.1. Concept For Transmutation By Accelerator Devices

9.1.2.2 Fission and Thermonuclear Explosive Devices

The overall pictorial representation of this concept is shown in Figure 9.2. A partitioning step to segregate the various toxic specimens is shown since it would probably be required in this concept. This explosive device concept does not appear feasible in part because of the large number of explosions estimated to be required to achieve the required total transmutation rate. If the desired constituents of the high-level waste were not separate from the rest, the material present in the target which was not a candidate for transmutation would compete for the available neutrons. Thus more explosions would be required and the concept would be less feasible than it appears to be when only selected constituents of the waste are considered as targets. Selected materials would be shipped to a tar-

get fabrication facility and, hence, to the disposal site. The targets would be lowered into a drilled hole along with the thermonuclear explosive device, the hole sealed, and the device set off. The neutrons created in the explosion would transmute some constituents of the waste. If it were possible to use the solidified encapsulated waste from the reprocessing step directly as the target, the waste canisters received from the reprocessing facilities would be sent directly to the transmutation site, eliminating the partitioning and fabrication stages.

9.1.2.3 Fission Reactors

The Present Fuel Cycle. The transmutation of actinide waste in fission reactors has been shown to be a feasible concept. Review of the present fuel cycle shown in Figure 9.3 helps to put this actinide transmutation concept in perspective. Uranium is

FIGURE 9.2. Concept For Transmutation By Fission And Thermonuclear Explosives

FIGURE 9.3. Present Nuclear Fuel Cycle

received at the fabrication facility from an enrichment facility and fabricated into fresh UO_2 fuel which is shipped to the reactor. After irradiation in the reactor to expected exposures of $\sim 27,500$ MWD/MT^(a) and $\sim 33,000$ MWD/MT for boiling and pressurized water reactors (BWRs and PWRs), the spent fuel is sent to reprocessing where U and Pu are recovered. Present reprocessing recovery efficiencies for U and Pu are around 99.5 percent. The radioactive waste to be sent to a federal repository,⁽¹⁾ consists of the residual U and Pu, all other actinide elements, and the fission products. The uranium recovered from commercial light water reactors (LWRs) is usu-

ally stored at the reprocessing facility. After a cooling period it may be sent to enrichment facilities to be used as feed material. It can also be sent directly to a fabrication facility to be incorporated into fresh fuel. The recovered uranium from fast breeder reactors (FBRs) will be recycled for blanket material to produce plutonium. The recovered plutonium is also stored at the reprocessing facility. After storage it may be sent to a fabrication facility to be used in making fresh $\text{UO}_2\text{-PuO}_2$ fuel.

A key item in the present nuclear fuel cycle is the economics of recovering uranium and utilizing the plutonium. If the economics dictate

a. Megawatt days per metric ton of fuel.

reprocessing the uranium because of its residual enrichment value, then the plutonium may either be stored for fast breeders or recycled in thermal reactors. If economics indicate that the uranium is not worth recovering and that the plutonium can be saved for fast breeder reactors, then this fuel may not be reprocessed until the time the plutonium is needed in fast breeder reactors. In this event, the spent fuel would probably be stored in water basins for the time period between discharge from the reactor and the advent of the breeder market. Another alternative is to reprocess LWR fuel to recover plutonium irrespective of the worth of the uranium for recycle. In any event, fissionable material has inherent value in that it can produce energy. Assignment of this economic

worth will be the crucial factor in determining when to reprocess LWR fuel. These economic arguments have not taken into account the costs of waste management. The costs of managing waste may have some impact on the timing of reprocessing and choice of isotopes to recycle.

Transmutation Cycles. This concept considers recycling constituents of high-level waste back to the reactor for subsequent transmutation to less toxic species. The concept is depicted in Figure 9.4. Specific constituents in spent fuel coming from the reactor are recovered during reprocessing and partitioning and are sent to a fuel fabrication facility for incorporation into rods for subsequent insertion in the reactor.

FIGURE 9.4. Concept For Transmutation by Fission Reactors

The technical feasibility for this concept is assessed by considering the constituents of the waste as being three categories of radionuclides. These categories are based upon the length of time the waste must be securely stored such that the remaining quantities of radionuclides represent no significant threat to the health and safety of the general public in the event of uncontrolled releases. The categories are:

1. 1000 years or greater
2. 100 to 1000 years
3. Less than 100 years

In high-level waste the actinide elements and perhaps the fission product I-129 fall in the first category. The fission products Sr-90 and Cs-137 are the primary constituents of the second category. Most of the other fission products fall in the third category. The reason certain fission products occur in the high-level waste in the amounts they do is that the probability for their undergoing transmutation in fission reactors is very small (i.e., small nuclear cross sections). As a result, they do transmute in-situ. The radionuclides in Category 3 are basically of this type and therefore are not viable candidates for transmutation in fission reactors. Some of the fission product radionuclides in Categories 1 and 2 are likewise of this type.

A potential scheme for transmutation appears to be chemically reprocessing the fuel, recovering all of the uranium and plutonium (i.e., eliminate process losses), segregating those species of high-level waste

which represent significant intermediate and long-term toxicity from those which rapidly decay to stable isotopes, and treating the significantly toxic species via transmutation.

As discussed in Section 7 of this volume, current technology for chemical reprocessing of nuclear fuel does not, however, provide product or waste streams according to these categories. Therefore, technological improvement is needed to separate actinide elements, and certain of the fission products (e.g., Sr-90 and Cs-137) from the balance of the waste stream. This partitioning is needed to recover these materials for transmutation in fission reactors. If partitioning and transmutation can be achieved, then the toxicity of these wastes may be effectively reduced to levels commensurate with those of Category 3 materials. Category 3 materials could then be managed by means such as storing in a retrievable storage facility.

9.1.2.4 Fusion Reactors

Fusion reactors, commonly referred to as Controlled Thermonuclear Reactors (CTRs), potentially can provide a copious supply of neutrons which could be used for effective transmutation of nuclear waste. The mean energy of neutrons produced in fission reactors is about 2 MeV, whereas the mean energy of neutrons produced in deuterium-tritium fusion reactors is about 14 MeV. The higher energy neutrons in a fusion reactor represent a better source for neutron-induced transmutation since high-energy neutron reactions can be used

directly or the neutrons thermalized to provide an intense source of low-energy neutrons. A schematic of this concept is presented in Figure 9.5. As in the case of fission reactor transmutation, it is likely that the waste stream from the reprocessing plant would have to be partitioned to obtain separate product streams for the actinide elements, strontium and cesium, and perhaps others in order to be managed in a CTR transmutation device.

The factors which must be considered in the cycle of processing, fabrication, and irradiation in the blanket of a fusion reactor are similar to those of transmutation in a fission reactor. The irradiation time in the fusion reactor is limited by two factors. One is the mechanical integrity of the canning material due to the intense neutron radiation field. A second factor is the decreased efficiency of the transmutation process with irradiation time

because of the decreasing concentration of target nuclei and the competition of daughter nuclei for the available neutrons. After some irradiation period, which has not yet been defined, the waste target must be discharged from the reactor. The discharge material will probably contain a significant fraction of the original target material. In addition, it will contain some short half-life daughter isotopes and radioactive container material. It appears likely that the toxicity of the original target material will not be reduced sufficiently in one irradiation step that it could be sent to storage. More likely, the material would be returned to and reprocessed in the partitioning facility. After one cycle of irradiation, the partitioning facility would probably have effluent streams of nonradioactive compounds and of radioactive short-lived compounds. The latter would be sent to storage and, subsequently,

FIGURE 9.5. Concept For Transmutation By Fusion (CTR) Reactors

to the federal repository. Since a sustained controlled thermonuclear reaction has not yet been achieved, use of this particular concept for waste management requires a breakthrough in this technology, in contrast to the fission reactor concept which requires only technological refinement.

9.2 TECHNICAL FEASIBILITY

Evaluation of technical feasibility was made for each transmutation concept system. The results of the evaluation are summarized in Table 9.1. For accelerator systems, only the spallation type accelerator showed real promise for feasibility. A spallation accelerator with a neutron-producing target could meet the

feasibility criteria under certain conditions for the transmutation of actinides to fission product. However, this process would be very costly at best and is judged to be much less attractive than the use of fission reactors or of fusion reactors. Certain long-lived fission products might possibly be transmuted with the neutrons from a spallation accelerator. This concept would, however, require chemical separation, treat only a small fraction of the fission products, and require a large capital investment in a large number of accelerators. Tentative calculations indicate the more likely feasibility of transmutation of Category 2 fission products when

TABLE 9.1 Summary of Transmutation Device Feasibility

Device	Technically Feasible for Transmutation			
	Fission Products			Actinides
	Category(a) 3	Category(a) 2	Category(a) 1	Category(a) 1
Accelerators				
• Electron Accelerator	No	No	No	No
• Proton Accelerator	No	No	No	No
• Spallation Accelerator	No	Possibly	Possibly	Possibly
Thermonuclear Explosives	No	No	Possibly ^(b)	Yes
Fission Reactors	No	No	No	Yes
Fusion Reactors	No	Possibly	Yes	Yes

- a) Category 1: Storage required for >1000 years
 Category 2: Storage required for 100 - 1000 years
 Category 3: Storage required for <100 years
 b) For specific isotopes

they are used directly as the spallation target. This possibility appears to be expensive and will require improvements in accelerator technology beyond that which already exists. Underground explosion of thermonuclear devices is estimated to be feasible for transmutation of actinides and marginally so for some separated isotopes of fission product. It is not clear that the concept is feasible from a seismic standpoint since large numbers of 100-kiloton (kt) explosions would be required annually. The transmutation of actinides in fission reactors is technically feasible, could reduce the actinide inventory to be managed by large factors, and would incur only a modest cost penalty. Fission product transmutation in thermal fission reactors does not meet the criteria. However, transmutation of certain long-lived fission products in fast reactors might be possible if the fission products can be separated. It is technically feasible to transmute moderate and long-lived fission products and actinides by the neutrons of fusion reactors if their development is successful. Supporting details on feasibility of each concept system are provided in Appendices 9.A through 9.D of this volume.

9.2.1 Accelerator Devices

Basically four schemes were evaluated:

- Direct bombardment by charged particles from accelerators having energies of tens of MeV

- Acceleration of the beta decay process by Coulomb excitation
- Use of two processes of photon transmutation: electron bremsstrahlung and stimulated gamma emission
- Use of a high energy (≥ 1 BeV) proton accelerator to produce by spallation an intense source of neutrons for transmuting radionuclides.

The last of these, namely the BeV proton-induced spallation device is the only concept showing any promise. The others are ruled out on a technical basis since they do not meet the criterion of energy balance. Each of these is discussed briefly below. Additional details are provided in Appendix 9.A of this volume.

9.2.1.1 Charged Particle - Nuclear Reactions

The penetration of medium-mass nuclei by protons requires that the protons have energies of tens of MeV. Nuclei with higher atomic numbers require even more energy. To achieve one nuclide-eliminating reaction with a charged particle would require an energy input of ~ 5000 MeV (i.e., 5000 MeV/reaction). The energy released in the power reactor where the waste was created is ~ 200 MeV/fission or about 1000 MeV per FP of radioactive waste. Clearly more energy is expended in transmuting the waste than was acquired in creating it. For waste transmutation by beams of charged particles from accelerators to become more attractive requires increases in both the probability of a reaction and the number of nuclei transmuted per reaction.

9.2.1.2 Beta Decay Acceleration By Coulomb Excitation

Beta decay from certain excited states of some nuclides proceeds more rapidly than beta decay from the ground state or, in some instances, more rapidly from the ground state than from certain metastable states. If this is the case for a radioactive fission product, it may be possible to reduce the effective lifetime of a nuclide by inducing a transition to a more rapidly decaying state. The fission product isotope Kr-85 is representative of these circumstances and therefore was studied.

Based upon an assumed energy balance, which reflects an optimistic amount of energy spent on transmuting Kr-85 nuclei, the reaction cross section needed to accomplish this was derived as a function of the energy of the particle (proton) causing the excitation. Comparison of the reaction cross section required for transmutation to occur with estimates of the physical values, shows differences of about three orders of magnitude (10^3), the physical values being smaller. Thus, the energy balance criterion is not met and this scheme is not technically feasible.

9.2.1.3 Photon Transmutation Processes

Two different photon processes were considered for radionuclide transmutation. These were photodisintegration of nuclei by electron bremsstrahlung, and stimulated gamma emission.

Electron Bremsstrahlung. Bombarding targets with electron beams in

the tens to low hundreds of MeV range produces a "shower" of photons from bremsstrahlung, from annihilation of electron-positron pairs, and from other processes. Some of the photons in the "giant" resonance energy region will undergo nuclear interactions such as (γ, n) , (γ, p) , $(\gamma, 2n)$ or (γ, np) . With nuclear waste material as targets, these nuclear reactions might lead to stable or less toxic nuclei. The feasibility criteria for such a system are the reaction yield, which consists of the number of reactions per incident electron, and the energy supplied per reaction. Assuming electrons accelerated to 34 MeV, the yield of photons was found to be too small and the energy required per transmutation reaction was found to be roughly two orders of magnitude larger than that gained (i.e., 200 MeV/fission in the reactor). Thus the energy balance criterion was not met, and this scheme is concluded to be not technically feasible.

Stimulated Gamma Emission. The stimulated emission of light was discovered in 1960. This process is known as the Laser (Light Amplification by Stimulated Emission of Radiation). In this process excited energy states of atoms or molecules decay very rapidly when placed in an intense field of electromagnetic radiation of the proper wavelength. One mode of the decay of excited energy states of nuclei is by gamma ray emission. Since gamma rays are electromagnetic radiation, as is light, the possibility of stimulated gamma ray emission, a Graser, has been discussed since the discovery of the

Laser. In the Graser concept, a matrix material contains the waste isotope(s) and other radioactive material of higher intensity but shorter half-life. Hence the probability exists that emission by a given nucleus increases with increasing intensity and exceeds the natural decay rate of the waste isotope. The two major problems which tend to discourage hopes for this concept are:

- According to the best available theory, stimulated emission is possible only for neutron boson emission. For practical purposes, this means photon emission and no stimulated emission is possible for alpha or beta decay of nuclei.
- The low attenuation material medium needed for stimulated gamma decay may be unattainable.

The concept of stimulated gamma emission as a transmutation method depends on the existence of nuclear states which could be reached by gamma excitation and which decay to a stable or short-lived isotope more rapidly than natural decay of the fission product. No such states have been identified. In addition, if such states were found, then stimulated emission requires reducing the attenuation of gammas by several orders of magnitude in order for it to be a technically feasible scheme.

9.2.1.4 Spallation Accelerators

High energy (≥ 1 BeV = 1,000 MeV), large current proton accelerators have been proposed⁽²⁻⁵⁾ which would provide the most intense, continuously operating source of neutrons

yet attained. The Intense Neutron Generator (ING) which was studied by the Atomic Energy of Canada, Ltd., group⁽²⁾ was the most serious investigation of a large spallation accelerator. The high energy neutrons produced through the spallation process in a Pb-Bi target were to be moderated in a surrounding D₂O medium to provide a thermal neutron flux of at least 10^{16} n/cm²-sec. Thermal flux values of this magnitude are required for the transmutation of low neutron cross section fission product (FP) isotopes. Several groups⁽³⁻⁵⁾ have studied the possible use of spallation accelerators for the transmutation of fission products over the years. Studies have also been made of the use of spallation accelerators to transmute fertile to fissile material, a concept which is closely related in technical feasibility to the FP transmutation concept.

The energy invested in transmuting one fission product nucleus was estimated to be between 23 and 110 MeV. Since these values are less than the 200 MeV acquired in creating the fission products, the energy balance criterion may be met. Candidate fission products for transmutation in a spallation accelerator are Cs-137, Sr-90, and Tc-99. The energy needed to transmute these isotopes is less than the energy invested in their creation.

The inventory of fission products such as Sr-90, Tc-99 and Cs-137 can be reduced significantly only by very high neutron flux levels, on the order of 10^{16} to 10^{17} n/cm²-sec. On the basis of a proton beam power of

65 MW (the ING proposal),⁽²⁾ it is estimated that roughly two spallation accelerators are needed to handle the inventory of one LWR.

The rates of transmutation of Sr-90, Tc-99, and Cs-137 were calculated and compared to the natural decay rates. The results are shown in Table 9.2. As shown, the rate is highly dependent on the thermal flux level attained with the device. It is very effective in accelerating the decay of Tc-99 because of the magnitude of the Tc-99 cross section (~ 22.6 barns). Clearly, thermal flux levels greater than 10^{16} n/cm²-sec are needed to make the concept attractive for transmutation of Sr-90 and Cs-137. It is assumed that daughter products may have to be removed from the system frequently to keep them from competing for the neutrons. Materials problems in the transmutation target, including its clad, and other parts of the system can be anticipated to be severe.

In examining the waste balance, it is found that a spallation accelerator may create more waste than it can transmute. The spallation protons with energy of 1 BeV on the lead target creates more short-lived radioactive product nuclei. The approximately 20 neutrons produced per incident BeV proton will come from on the order of 5 to 10 parent nuclei, with almost all of the daughters being left in radioactive precursor states. Almost all of the radioactive waste created, however, has half-lives much shorter than the fission products which would be transmuted. This would not be true for a uranium target.

Another conceptual use of the spallation accelerator is to use the radioactive fission product waste as the proton target of the accelerator. In this concept the fission product nuclei are transmuted directly by spallation and further by nuclear processes induced by the secondary

TABLE 9.2. Transmutation Rates using Thermal Neutrons from a Spallation Accelerator

Isotope Transmuted	Time (yrs) To Eliminate 99% of the Material		
	Natural Decay	Transmutation Using Accelerator	
		10^{15} n/cm ² -sec	10^{16} n/cm ² -sec
Sr-90	191	83	14
Tc-99	1.40×10^6	6.6	0.66
Cs-137	199	170	80

neutrons. A study team of the Japanese Atomic Industrial Forum has speculated that eighty-five Cs-137 nuclei could be transmuted per incident proton.⁽⁵⁾ The Los Alamos Scientific Laboratory group which has reviewed this study suggests that 200 FP nuclei might be transmuted per incident proton (Appendix 9.E of this volume).

9.2.2 Fission and Thermonuclear Explosive Devices

Because thermonuclear explosive devices are known to produce large yields of neutrons, the use of these neutrons for the transmutation of radioactive waste has been proposed.⁽⁶⁻⁸⁾ A preliminary evaluation by PNL indicated that this concept was not technically feasible. The evaluation was by Los Alamos Scientific Laboratory (LASL) personnel who strongly disagreed with the PNL Study findings and concluded that the concept may be very attractive. The details of the LASL review are contained in Appendix 9.E of this volume. Subsequently, PNL reevaluated the concept based on the LASL review. The details of the PNL evaluation are given in Appendix B of this section.

The concept which was evaluated consisted of the explosion in a hole (1.5km deep) of a fission-actuated thermonuclear device with an explosive yield of 100 kilotons. The explosive yield was taken from the LASL review as the largest which could be seismically decoupled from adjacent explosions since a large number would be required on an annual basis.

The main conclusions of the PNL evaluation of the feasibility of the concept are:

- The concept can be considered only for radioactive waste with half-lives much greater than the 12.3 year tritium because of significant production of tritium residue in the device. Hence, the transmutation of short-lived radioactive waste is not technically feasible.
- The estimated cost of transmutation of the long-lived fission product isotopes, TC-99 plus I-129 plus CS-135 in elemental form, is almost twice the cost of the electricity produced in their creation. Hence, the transmutation of long-lived fission product elements or structural material waste is not technically feasible.
- The technical feasibility of the transmutation of separated isotopes of long-lived fission products cannot be obtained from the PNL evaluation but would require more rigorous calculation of the neutronic behavior of the concept.
- The estimated cost of transmutation of the actinide waste of Np, Am and Cm is estimated to be less than 20 percent of the electrical cost of their creation. The concept is, thus, technically feasible for actinide transmutation. This concept is, however, judged to be much less attractive than the concept of actinide recycle in fission reactors both on a cost basis and also on the large annual number of thermonuclear explosives required for the transmutation.

9.2.3 Fission Reactors

Since fission reactors produce neutrons, it has been suggested^(3,9,10) that radioactive waste might be transmuted in fission reactors. Early studies^(3,9) proposed burning fission products in high-flux reactors. More recent studies⁽¹⁰⁾ conducted at Oak Ridge National Laboratory (ORNL) have considered transmuting fission products and actinides in fission reactors. Claiborne⁽¹⁰⁾ did an extensive investigation of transmutation of actinides in a prototypical commercial pressurized water power reactor (PWR) and showed that the toxicity could be reduced by one to two orders of magnitude by recycling actinides in PWR UO_2 fuel. The ORNL studies concluded that transmutation of fission products in fission reactors was not technically feasible, but that transmutation of actinides was feasible. Independent analyses made at PNL confirm both of these conclusions.

Kubo⁽¹¹⁾ and Kubo and Rose⁽¹²⁾ at Massachusetts Institute of Technology have studied Claiborne's results and similarly have concluded that actinide recycle in fission reactors is not only technically feasible but an attractive waste management concept. Kubo and Rose further conclude that actinide recycle in fast breeder reactors would be even more attractive. This conclusion, however, is largely based on intuition and is, as yet, unsupported by any definitive calculations. Their extension to Claiborne's work has been primarily devoted to analysis of the further

reduction in potential toxicity which would accrue from improved separations efficiencies for actinides higher than plutonium.

The technical feasibility for transmuting fission products and actinides is summarized below. Supporting details are given in Appendix 9.C of this volume.

9.2.3.1 Fission Product Transmutation

Transmutation of fission products in fission reactors is not technically feasible because the waste inventory and transmutation rate criteria cannot be met. The cross sections for the fission products and the thermal neutron flux levels attained in fission reactors are both too low. Indeed, if the cross sections were appreciable, the fission products would not exist in the quantities they do since they would burn out in-situ. Table 9.3 was extracted from Reference 10 to illustrate why fission product transmutation is not technically feasible. As shown in the table, the neutron cross sections are small, thus requiring a high neutron flux. The data given in the bottom of Table 9.3 show that flux levels of the order of 10^{16} to 10^{17} n/cm²-sec are required to achieve a reasonable gain in transmutation rate relative to natural decay. In summary, it is concluded that transmutation of fission products in thermal fission reactors is not feasible since none of the technical feasibility requirements appear to be met.

TABLE 9.3. Properties of Several Important Fission Product Nuclides and Time Required for 99.9 Percent Reduction of Their Inventory by Decay and Neutron Transmutation⁽¹⁰⁾

Nuclide	Sr-90	Cs-137	Kr-85	H-3	I-129
Half-life, years	28.9	30.2	10.74	12.33	1.6×10^7
Burnout cross section, barns ^(a)	1.2	0.17	1.8	nil	35
Curies/metric ton in spent fuel ^(b)	77,600	108,000	11,400	708	0.0367
Relative hazard in spent fuel ^(c)					
m ³ air at RCG/metric ton	2.6×10^{15}	2.1×10^{14}	3.8×10^{10}	3.5×10^9	1.8×10^9
m ³ water at RCG/metric ton	2.6×10^{11}	5.4×10^9	--	2.3	6.1×10^5
Time required for 99.9% decay and burnout, years ^(d)					
Decay only	288	302	107	123	1.6×10^8
$\phi = 10^{14}$ n/cm ² ·sec ^(e)	249	295	106	123	63
$\phi = 10^{15}$ n/cm ² ·sec ^(e)	112	245	98	123	6.3
$\phi = 10^{16}$ n/cm ² ·sec ^(e)	17	91	57	123	0.63
$\phi = 10^{17}$ n/cm ² ·sec ^(e)	1.8	12	11	123	0.06

- a. Effective thermal cross section in typical spectrum of a PWR having average thermal flux of 2.91×10^{13} n/cm²·sec.
b. Per metric ton of uranium charged to a PWR having average specific power of 30 MW/metric ton and burnup of 33,000 MWd/metric ton.
c. Volume of air and water potentially contaminated to RCG (10 CFR 20) by the content of a metric ton of spent fuel.
d. Indicated times are doubled and tripled for reduction of inventory by factors of 10^6 and 10^9 , respectively.
e. Average thermal flux assuming spectrum typical of that in a PWR.

9.2.3.2 Actinide Transmutation
Claiborne⁽¹⁰⁾ has conducted the most extensive study to date of actinide transmutation in LWRs. The result of Claiborne's study are used extensively here to delineate technical feasibility of actinide transmutation in fission reactors. Some preliminary calculations were made at PNL of actinide recycle using Claiborne's strategy and examining some other strategies. These three strategies are depicted in Figure 9.6. The results of the PNL calculations confirm that actinide recycle in thermal fis-

sion reactors is technically feasible, since it meets all of the selection criteria.

Review of Claiborne's Work.

Claiborne's strategy, shown as strategy 1 in Figure 9.6, was to store the U and Pu recovered during chemical processing and recycle the other actinides in new PWR UO₂ fuel assemblies. Thus each fuel rod in every PWR fuel assembly contained these actinides. His analysis covered several chemical processing extraction efficiencies. He showed that the actinide inventory in accumulated

1. Reference Strategy (Proposed by Claiborne^[10])

2. Alternative Uranium Strategy

Same as 1 except recycle actinides in 10% of new fuel rods, 90% of new fuel normal UO_2 fuel

3. Plutonium Recycle Strategy

FIGURE 9.6. Actinide Recycle Strategies Studied

waste is substantially reduced when actinides are recycled.

His results are cast in terms of potential relative hazard,^(a) defined as the amount (volume) of water (or air) required to dilute each nuclide of a mixture to its Radiation Concentration Guide value (RCG). The cumulative hazards of actinide waste for the short and long terms are displayed in Figures 9.7 and 9.8, respectively. The top curve on these figures is the potential hazard of the actinide waste accumulated if the actinides from a 1000 MWe PWR, operating at 80 percent capacity for 60

years, are not recycled in the reactor (i.e., other disposal schemes are used 10 years after reprocessing). The bottom curve is for the recovery of 99.5 percent of all actinides during chemical processing, with the U and Pu stored, and all other actinides recycled. The difference between the two curves represents the reduction of the potential relative hazard of actinide waste. Claiborne also shows that the reduction is even more substantial (factors of 200) if recovery of 99.9 percent of U and Pu can be achieved in chemical processing. The postulated reductions in

a. We prefer to use the term toxicity index rather than hazard index because its connotation is clearer. However, since we are taking information directly from Claiborne's report,⁽¹⁰⁾ we use his notation here.

FIGURE 9.7. Short-Term Cumulative Hazard of Actinide Waste from 60-Year Operation of a Typical PWR (10)

cumulative hazard may not be meaningful on an absolute basis. What they do reflect is the reduction in quantity of actinide waste which must be managed or otherwise disposed of. There is an implicit assumption that if this quantity is reduced, the future hazard is reduced proportionately.

PNL Studies. Survey calculations were made at PNL using the ALTHAEA code⁽¹³⁾ for the three strategies shown in Figure 9.6. The first strategy which was calculated was that assumed by Claiborne in order to have a verification of his results.

For the first strategy of recycling in every UO_2 rod, the calculated amount of each actinide element during recycle was consistently larger than the values obtained by Claiborne.

FIGURE 9.8. Long-Term Cumulative Hazard of Actinide Waste from 60-Year Operation of a Typical PWR (10)

Of the numerous possibilities existing to cause differences in calculated values, the most logical being cross section differences and that the calculations made by Claiborne may not have included neutron spatial self shielding effects, since he used the ORIGEN code⁽¹⁴⁾ which does not have this provision. Comparison of neutron multiplication values shows that the values obtained by Claiborne are consistently lower but with the same trends as those obtained in the PNL study. This tends to support the contention of differences due to spatial self shielding effects. The differences in concentrations indicate that the hazard reductions in transmutation of actinides presented above from Claiborne's study might be overestimated. A penalty of about 4 percent in the enrichment of U-235 is encountered for equilibrium cycles when actinides are recycled in every UO_2 fuel rod.

A second set of calculations was made assuming that the actinides would be recycled in every tenth UO_2 rod rather than in every rod. The actinide element inventories for this case along with those of recycling in every UO_2 rod are presented in Table 9.4. Comparison of the differences in values between cycles for each strategy shows that the actinide inventory is not reduced as much by recycling the actinides when concentrated in a few rods as compared to recycling in every UO_2 rod. The enrichment penalty for this strategy ranges from ~3 percent in U-235 content at second recycle to ~5 percent at equilibrium. The incentive for re-

cycling actinides in fewer rods is to reduce the economic penalty which would occur if remote fabrication of UO_2 fuel containing actinides is required. In this event the additional enrichment penalty would be a small fraction of the savings derived from reduced fabrication costs.

The third strategy calculated was the storage of the recovered U from chemical processing and the recycle of the rest of the actinides in LWR plutonium recycle fuel ($\text{UO}_2\text{-PuO}_2$). The plutonium recycle scheme employed was typical of current fuel management schemes for recycling self-generation plutonium. The inventory of the actinides Np, Am, and Cm fall in between the values obtained for the UO_2 fuel strategies, thus making it a technically feasible concept. However, the plutonium generated contains large quantities of Pu-238 which would dictate expensive fabrication in a remote facility. The fissile content also becomes so low that the fuel would eventually have to be driven by other fuel in the reactor.

In summary, actinide transmutation in fission reactors appears to be technically feasible and is attractive in that significant reductions in the toxicity index of actinides can be achieved. The cycle could be accomplished in UO_2 rods, either every rod or some fraction thereof, or in plutonium recycle rods. This study indicates that using existing chemical separation and recovery efficiencies, the recycle of actinides in light water power reactors can achieve a decrease in the short-term toxicity index of about a factor of 10 and

TABLE 9.4. Comparison of Actinide Inventories for Two Recycle Strategies Using UO_2 Fuel in A PWR.

Recycle No.	Strategy →	Actinide Inventory (gms/MT of Heavy Metal in Reactor)					
		Np		Am		Cm	
		1 ^(a)	2 ^(b)	1	2	1	2
0		521	521	144	144	32.0	32.0
1		784	812	176	176	92.0	92.5
2		921	994	183	187	135	141
3		993	1118	184	192	161	177
4		1031	1193	184	194	176	202
5		1052	1253	184	195	184	222
6		1063	1300	184	195	189	233
7		1068	1334	184	196	191	239
8		1071	1360	184	196	192	242
9		1073	1380	184	196	193	243

a. Recycle actinides in every UO_2 rod as shown in Figure 9.7.

b. Recycle actinides in one-tenth of the UO_2 rods.

about a factor of 50 decrease in long-term hazard index. These reduction factors may be significantly improved by achieving higher separation efficiencies and better optimization of the reactor irradiations. As Kubo⁽¹¹⁾ points out, a greater reduction in the toxicity indices may also be possible by recycling actinides in FBRs. There are, however, no calculations presently available to support the latter speculation. Lastly, as discussed in peer group review meetings (see Appendix 9.E of this volume), a special purpose reactor designed specifically to destroy actinides (i.e., convert them

to fission products) has merit and should be evaluated. Such a facility might be constructed and operated by the Federal government.

9.2.4 Fusion Reactors

The unique features of a fusion reactor, or Controlled Thermonuclear Reactor (CTR), as a waste transmutation device are the high energy of the neutrons available and the high flux anticipated.^(15,20) The most troublesome fission product nuclei to be considered for transmutation are those which have relatively small neutron reaction cross sections. Due to

the high energy, $(n,2n)$ reactions can contribute significantly to the process, while the high flux and high source strength makes waste transmutation at reasonable rates appear possible a priori.

Studies were made of waste transmutation in blanket regions of a CTR. Included were: 1) the use of fast neutron flux for transmutation of Sr-90 and Cs-137, 2) the use of thermal neutron flux for transmutation of the fission products Sr-90, Cs-137, Kr-85, I-129, and actinides, and 3) transmutation of large amounts of Cs-137 in a moderating blanket.

A summary of the principal findings of these studies is given below. Supporting technical details are given in Appendix 9.D of this volume.

CTR power plants can, in principle, transmute all of the Sr-90 and Cs-137 created by the electrical economy, even if the economy assumed were all nuclear power. The daughter products produced in the transmutation process appear to approach well defined equilibriums after a year or so of operation. They should, therefore, cause few perturbations on the characteristics of operating CTR power plants. In addition, the fission product nuclei can contribute to additional neutron production in the CTR blanket.

Both fission products and actinides can be effectively transmuted with thermal neutrons using dilute target samples. For a representative value of a fusion plasma power den-

sity, calculations indicate that the toxicity half-life^(a) is reduced by about 5 orders of magnitude for the actinides and by 8 orders of magnitude for I-129. For the other fission products the toxicity half-life is reduced roughly in the range between 5 and 100.

Quantities of Cs-137 could be transmuted under the projected CTR blanket loading conditions^(19,20). The reductions in Cs-137 toxicity are, however, projected to be at most a factor of about three.

The irradiation of actinides in the blanket of a CTR would greatly reduce the cumulative toxicity index due both to the high energy of the D-T fusion neutrons and to the intense neutron sources expected for fusion reactors. Calculations made for irradiations of small samples of actinides in CTR blankets indicated reductions of toxicity indices by factors of 1,000 to 10,000 for expected neutron source levels. Definitive calculations have not yet been made for more massive loadings of actinides but this concept is clearly capable of reducing the toxicities by a factor of at least 10 to 100 below that of actinide recycle in LWRs.

In summary, the technical feasibility of transmutation of actinides and selected fission products seems clearly established. The limitations which will be imposed by factors such as radiation damage and heat transfer

a. Defined as the time required for the toxicity index for a particular isotope and its daughters to decay to one-half of its original value.

will require definition when the attainment of a viable CTR appears to be closer than it is at present.

9.2.5 Candidate Transmutation Concept System

Since it is technically feasible to transmute actinides in fission reactors and CTRs, and certain fission products in CTRs, these two reactor technologies combine to form a potentially viable long-term strategy for waste management. This strategy is described below.

9.2.5.1 System Description

The layout of the system is given in Figure 9.9. In the near term (year 2000 or 2010), the actinides obtained from the partitioning process would be recycled in fission reactors and the fission products

placed in a retrievable storage facility. In the long term, if the development of present concepts of CTRs is successful, the fission products would be retrieved from storage and recycled along with the actinides in the CTR.

A pictorial representation of the projected nuclear power economy is given in Figure 9.10 to illustrate the impact of this strategy. The installed capacity of fission reactors up to year 2000 is taken from the AEC projection given in Reference 21. The actinide inventory projected for this growth rate (shown in Volume 1 of this report) is also shown on Figure 9.10. It is not unlikely that fission reactors will follow the bell shaped curve, since new technologies will be developed which will compete

FIGURE 9.9. Transmutation Waste Management Strategy

FIGURE 9.10. Projected Nuclear Power Economy in U.S.A.

more favorably in the power marketplace. If actinide recycle is undertaken in fission reactors, the actinide inventory should tend to follow the fission reactor growth curve. The fission reactors being built in the latter period of this curve will have increasingly larger amounts of actinides recycled through them. However, if the CTR becomes a viable power source, then actinide recycle can be transferred to these plants, eventually transmuting all of the actinides into fission products. Then the most toxic of the fission products can themselves be converted in the CTR to stable or less toxic species.

9.2.5.2 Requirements for the Concept System

The general system flow diagram, shown in Figure 9.11, starts with bulk high-level liquid waste from the reprocessing plant. The waste may be chemically conditioned and stored for a 5-year period to provide for cooling before being chemically segregated into actinide and fission product streams. The actinides, long-lived fission products, and short-lived fission products are each solidified and transported to the fabrication site. The transportation is expected to be done in conventional types of shielded and cooled casks via common carrier (truck or rail).

FIGURE 9.11. Overall System Requirements for Managing High-Level Radioactive Wastes by Transmutation Concept

Since transmutation of most of the short-lived fission products does not appear feasible, the option to use other waste management concepts such as retrievable storage is expected to be exercised.

The actinides are fabricated, either into fuel or target rods for fission reactors and/or fusion (CTR) reactors. The fission products would be fabricated into CTR target rods in shielded process lines by remote fabrication.

The fabricated rods would be shipped to the reactor site using conventional shielded shipping casks.

Upon receipt at the reactor site, they are inserted into the reactor. The insertion into the reactor will be done with machines currently in use for refueling reactors.

The actinides and fission products in the fuel and/or target rods are transmuted during reactor operation. Any residue, or heel, would be returned to reprocessing and the cycle repeated.

9.3 ESTIMATED RESEARCH AND DEVELOPMENT REQUIREMENTS^(a)

Research and Development needs are primarily directed toward determining

a. Members of the peer group and others who reviewed the initial draft of this report had numerous comments and suggestions on this section of the report. As a result, portions of this section presented in the draft have subsequently been revised to incorporate these suggestions.

if viable strategies for application of transmutation exist. This is the objective of the Research and Development study described below. Partitioning is an integral part of any transmutation cycle. Thus, the viability of partitioning must be established early in any study to develop transmutation schemes. The near-term potential of actinide transmutation using fission reactors dictates that Research and Development emphasis be placed in this area. However, advanced technologies, such as CTRs, hold future promise, and a sustained low-level Research and Development effort is defined to evaluate the application of these technologies for waste transmutation.

The Research and Development needs are estimated to require 20 years and to cost about \$133 million. The needs are outlined in two tasks. Task 1 includes the Research and Development needs for developing the fission reactor transmutation concept and is estimated to require between 10 and 15 years and cost \$130 million to complete. Task 2 includes the Research and Development needed to monitor advanced strategies and make preliminary evaluations of their applicability as transmutation devices. This task is estimated to cover 20 years and cost \$3 million.

A fundamental assumption underlying the estimates presented here is that transmutation will be accomplished in commercially owned facilities. It should be noted that the possibility of transmutation in government-owned special purpose actinide burning reactors merits consideration. In fact, it was the

consensus of the peer review group that this should be studied (Appendix 9.E of this volume). However, estimates of the needs and costs required for researching and developing this waste management strategy were not developed for this study.

9.3.1 Task 1 - Actinide Recycle in Fission Reactors

The Research and Development study discussed below is directed toward the development of a viable strategy for application of transmutation in fission reactors. Research and Development expenditures for actinide recycle in fission reactors are estimated to be: \$3 to 5 million for partitioning; \$50 million for terrestrial disposal of the residual waste; and \$75 million for actinide recycle engineering. The latter investment of \$75 million assumes that government provides \$25 million and industry funds \$50 million. The \$25-million investment by government would be to develop base technology; whereas, investment by industry would implement the technology to a viable commercial operation. The needs relative to terrestrial disposal of the waste fraction not transmuted are outlined in Volume 2 of this report and will not be discussed here. The needs for the balance of the actinide recycle Research and Development study are outlined below.

The Research and Development program for actinide recycle includes two phases. The first phase covers three to four years to evaluate the efficacy of actinide recycle in various types of central power stations currently operating (LWRs and HTGRs)

and being developed (LMFBRs and GCFBRs). The efficiency of actinide transmutation in each of these reactor types would be evaluated, the limits of technology for partitioning actinides would be determined, and the impact (in terms of risk) of implementing actinide recycle in these fuel cycles would be estimated. To assess the desirability of actinide transmutation requires evaluation of the effect of recycling actinides on all waste streams in the fuel cycle (e.g., in the process steps of fabrication and reprocessing/partitioning) as well as the reduction of high-level waste via burnup in the reactor. If this first phase indicates the need, the program will proceed to the second phase. The second phase includes the Research and Development needed to implement the concept on a commercial scale. Technical bases and criteria needed to guide design would be developed. Experimental pilot programs, and a full-scale demonstration program would most likely be required to establish the viability of the concept. Process engineering studies associated with fabrication, reprocessing, and partitioning will be needed to establish the technology and to develop guidelines and specifications which assure safety during all fuel cycle operations. A cooperative effort between governmental laboratories and private industries is required to meet the objectives of this program.

The two phases of the Research and Development program in Task 1 and the activities within these phases are displayed in Figure 9.12 as a func-

tion of program years. Each phase and the Research and Development activities within each phase are discussed below.

9.3.1.1 Phase 1 - Evaluate Efficiency of Actinide Transmutation in Fission Reactors

The objective of Phase 1 efforts is to confirm the success of transmutation of actinides in fission reactors and provide quantitative data for a basis to evaluate if the need is compelling relative to other waste management alternatives. The optimum nuclear fuel cycle for transmutation will be identified. The efforts are broken into three major activities which are shown in Figure 9.12.

The first activity is to evaluate actinide transmutation, considering each of the various reactor types currently operational or expected to be commercially viable as central power stations between now and the early 1990s to determine the efficiency of each in reducing actinide inventory of high-level waste.

The reduction of toxicity in actinide recycle is directly related to the efficiency of partitioning actinides from the high-level waste stream. Therefore, Research and Development efforts are needed to determine the level of partitioning that is technically achievable, and these efforts are listed as the second major activity in this phase of the program.

The third activity is to evaluate the impact of recycling actinides in

Milestones:

1. Identify optimum type of reactor for transmutation.
2. Evaluate total risk of implementation.
3. Phase 1 completed. Go or no go decision on transmutation relative to other waste management alternatives.
4. Initial decision on demonstration experiment based upon early data from capsule irradiations.
5. Final Decision on proceeding with demonstration experiment.
6. Demonstration completed.
7. Phase 2 completed.

FIGURE 9.12. Estimated Research and Development Program for Actinide Transmutation in Fission Reactors

these reactors by assessing the incremental risks at the affected process stages in these nuclear fuel cycles. This information is needed to assure that advantages accruing to waste management objectives via destruction of actinides in the reactor are not negated by increases of actinides in processing waste streams such as in fabrication and partitioning.

The data developed in these three activities will be used to determine the viability of transmutation for each of these nuclear fuel cycles in order to identify the most likely fission transmutation cycle. This information can then be used in comparison with similar data on alternative disposal methods (e.g., geologic, seabed, extraterrestrial) to decide

whether transmutation is compelling in management of high-level waste.

It is estimated that Phase 1 of the program would be completed in about three to four years at a cost of around \$5 to \$7 million. Perhaps about one-half of this investment will be needed for Task 2 on partitioning since fission reactor transmutation requires a partitioning step. Details of Research and Development for partitioning are given in Section 7 of this volume. Industry is expected to be actively involved in this evaluation, and industries' involvement in this phase aids in making the decision as to whether transmutation is viable and compelling in management of high-level waste.

9.3.1.2 Phase 2 - Development and Demonstration of Actinide Recycle in Fission Reactors

Phase 2 of the program would be initiated only if the results of Phase 1 prove positive and it is decided there is need for transmutation in management of high-level waste. The objective of this phase of the program is to demonstrate the viability of the concept of recycling actinides in the nuclear fuel cycle(s) identified as most likely in Phase 1. The time required for accomplishing this objective is estimated at somewhere between 10 and 15 years and the cost about \$75 million. The efforts in this phase represent a large-scale cooperative program involving government and industry. It assumes that government will fund the development of base technology and the transfer

of the technology to industrial capability and that industry will fund the development and application of the engineering capability for recycling actinides in fission reactors. As shown in Figure 9.12, Phase 2 is outlined as four major activities.

Technical bases are needed to insure that actinides can be adequately handled in all transmutation cycle process steps--fabrication, irradiation, reprocessing, partitioning, waste treatment, and transportation between these steps. Thus, the first major activity is to develop the broad technical base needed to implement actinide recycle in fission reactors. Included in this activity is evaluation and development of data on the chemical and physical constants for actinides. In addition, experimental data will be developed to provide bases for making accurate engineering predictions of mechanical, chemical, thermal, and nuclear behavior during all process operations in the transmutation fuel cycle and to assure that the basic theoretical knowledge is not limiting the development of actinide recycle engineering.

The second major activity in Phase 2 of the program is the development of criteria and specifications to design, manufacture, transport, reprocess and partition the actinide-bearing fuel/or target elements to be irradiated in the power reactor. Process engineering studies are required to establish these technologies as viable commercial processes and develop specifications which assure adequate safety margins in all steps of the nuclear fuel cycle.

In activity 3, shown in Figure 9.12, irradiation experiments will be conducted to measure the behavior of small samples bearing actinides in reactor environments. These data will permit early determination of the transmutation rates of actinides and provide data for assessment and normalization of design methods. Moreover, this information can be used in the first activity to aid in determining the adequacy of knowledge concerning physical constants used in design analysis.

The outcome of early measurements on irradiation of small samples bearing actinides will assist in deciding whether to proceed with the design of a large scale demonstration, listed as the last major activity of Phase 2. In the event that the outcome is favorable, then activity 4 will proceed to a full scale demonstration of the concept. This demonstration could be conducted in an AEC-owned facility or, if available, a utility-owned and operated reactor.

The program outlined in Phases 1 and 2 involves major expenditures of funds covering roughly 15 years of development. These efforts represent contributions which would be forthcoming from numerous governmental laboratories, universities, private companies, and public agencies. It is important that these efforts be coordinated to assure proper guidance and direction of the research program to meeting waste management objectives. Though program management has not been delineated as an activity, it should be recognized that it is needed.

9.3.2 Task 2 - Advanced Concept

Evaluations

Since transmutation using CTRs shows promise, as does possibly spallation accelerators and nuclear explosives, an effort, albeit low relative to tasks of other phases in the program, should be sustained to evaluate these concepts. For example, it would be worthwhile to undertake a modest effort to better ascertain the accelerator requirements for transmutation in order to determine the breakthrough necessary in acceleration technology for this alternative to represent a technically feasible transmutation concept. In addition, other advanced schemes are expected to be brought to the attention of the waste management program in the future for evaluation of their feasibility. For example, the development of laser technology may lead to processes applicable for waste transmutation.

It is estimated that the evaluation of advanced concepts would each cost up to \$3 million expended over about 20 years. If significant breakthroughs are realized in these advanced technologies and preliminary evaluation of transmutation shows merit, these efforts are expected to expand accordingly.

9.4 ESTIMATED TIME FOR REQUIREMENTS FOR OPERATION

It is estimated that between ten and fifteen years would be required for implementing operation of actinide recycle in fission reactors. If transmutation of actinides were undertaken in commercial LWRs, then operation could be implemented in about

ten years. Implementation of recycle in commercial fast reactors would require longer time periods and depend upon the advent of commercial operation of fast breeder reactors. If it appears advisable to recycle actinides in special purpose burner reactors, then sufficient time would be needed to design and construct such a facility. A time span of fifteen to twenty years is estimated to be required for this case.

Estimates of the time span needed in transmutation of actinides and fission products in CTRs depends on achieving a viable CTR. Current estimates of a commercially viable CTR is in the period between the years 1995 and 2000. We estimate that it would require at least 5 years beyond the commercial implementation date for operation of actinide recycle in CTRs.

Coupling the technological developments required for actinide transmutation in fission reactors and the projected time scale of the introduction of LWRs, HTGRs and LMFBRs in the production of electrical power suggests that the early 1990s represent a reasonable target for application of actinide recycle in a commercial facility. This would allow sufficient time for development and demonstration of partitioning. Application of actinide recycle in government-owned facilities could probably be accommodated in the same time frame. Thus, a date somewhere between 1990 and 1995 would seem to represent a conservative estimate irrespective of the mode of operation.

9.5 CAPITAL AND OPERATING COSTS

The transmutation of actinides in a commercial type light water power reactor (LWR) has been shown to be technically feasible. The potential reduction in long-term toxicity index of high-level waste is in the range of two orders of magnitude if actinides are recycled in a LWR. In HTGRs and FBRs, equivalent or more reduction in toxicity index is expected. In a CTR, transmutation of actinides and selected fission products (e.g., Sr-90 and Cs-137) appear technically feasible and more rapid reductions in toxicity appear attainable. However, the CTR faces a technological breakthrough in attaining a sustained thermonuclear reaction.

For purposes of estimating capital and operating costs, the reference case selected was actinide recycle in LWRs because these reactors are presently competitive with other electrical generation systems. The base case evaluated takes the Am, Cm, Bk, Cf and Np extracted from the high-level waste stream in the partitioning step and recycles them back through the fabrication and irradiation step. This path is parallel to the plutonium recycle path presently being utilized in LWRs. Wastes from the transmutation processing would be stored in a retrievable storage facility until the time that CTRs are developed. These costs are not included since their projection is difficult in terms of if and when CTRs become available.

9.5.1 System Characteristics for Cost Bases

Claiborne's analysis⁽¹⁰⁾ has been used as a base for much of the discussion of actinide reaction in LWRs. A cursory analysis of the costs associated with Claiborne's recycle strategy shows that it is economically unattractive. Once actinide recycle is initiated, all UO_2 fabrication would have to be remote. This remote handling would result in fabrication costs approximately five times higher than present UO_2 fabrication costs. Since fabrication costs represent a significant fraction of the total fuel cost, this recycle strategy is far from optimum.

However, obvious alternative strategies exist. If the actinides were recycled in a small fraction of the UO_2 rods (<10 percent), then at least 90 percent of the rods could be fabricated without a cost penalty. The remainder would have to be fabricated remotely. A fuel management strategy describing this alternative is as follows:

1. Partitioning will recover Th, U, Np, Pu, Am, Cm, Bk and Cf from the high-level waste stream from the reprocessing plant. One percent of the fission products will be present in this stream.
2. The actinides will be recycled in 10 percent of the rods.
3. The actinide rods will be remotely fabricated.
4. The actinide fuel elements in conjunction with the remaining UO_2 elements will be designed to give the same reactor endurance (i.e., the same goal exposure) as a UO_2 fuel assembly.

5. The fuel assemblies remain in the reactor for the same length of time as UO_2 only assemblies.

The cost bases for this alternative transmutation strategy are described in detail in the following paragraphs and will be used to obtain a preliminary cost penalty for the transmutation of actinide waste in a LWR.

Estimates of radiation levels and associated fabrication costs for plutonium recycle fuels⁽²²⁾ were used as a basis for determining if remote fabrication was a requirement. An estimate of the neutron emission rate to be expected from the actinide bearing UO_2 fuel was obtained from Claiborne's work.⁽¹⁰⁾ As can be seen from the values on Table 9.5, which is extracted from the referenced document, neutron dose rates of 10^{11} n/sec-MT are realized after only a few cycles. By comparison, the "high exposure" plutonium considered in BNWL-273⁽²²⁾ has a neutron rate of about 10^7 n/sec-MT. The increased neutron yield of four orders of magnitude would make the manufacture of fuel containing the actinides intolerable in a glovebox facility.

The transportation of these toxic materials can be minimized by having the actinide target manufacturing facility an integral part of the reprocessing plant. In that way the shipping containers, which are normally returned empty from the reprocessing plant to the reactor, can be utilized for shipment of the actinides to the reactor site. Kubo and Rose⁽¹²⁾ have come to a similar conclusion. Thus no significant cost

TABLE 9.5. Effect of Recycling on Hazardous Radionuclides in the Reactor and the Processing Plant(10)

Recycle Number	Air Required for Dilution to RCG, m ³ /metric ton of fuel		Neutron Yield After 150 d. Decay, n/sec metric ton of fuel		
	Reactor Discharge	150 d. Decay (No U or Pu)	Actinides (No U or Pu)	Cf Only	Cm Only
0	1.20×10^{17}	1.41×10^{16}	5.50×10^8 (a)	1.96×10^5	4.76×10^8
1	1.71×10^{17}	2.02×10^{16}	1.53×10^9	1.73×10^8	1.33×10^9
2	2.12×10^{17}	4.30×10^{16}	4.66×10^9	2.77×10^9	1.87×10^9
3	2.23×10^{17}	4.86×10^{16}	1.47×10^{10}	1.24×10^{10}	2.20×10^9
4	2.27×10^{17}	5.15×10^{16}	3.50×10^{10}	3.26×10^{10}	2.42×10^9
5	2.30×10^{17}	5.31×10^{16}	6.65×10^{10}	6.39×10^{10}	2.60×10^9
10	2.36×10^{17}	5.80×10^{16}	3.33×10^{11}	3.30×10^{11}	3.24×10^9
15	2.41×10^{17}	6.20×10^{16}	6.32×10^{11}	6.29×10^{11}	3.71×10^9
20	2.45×10^{17}	6.50×10^{16}	8.71×10^{11}	8.66×10^{11}	4.03×10^9
25	2.48×10^{17}	6.74×10^{16}	1.04×10^{12}	1.03×10^{12}	4.23×10^9
30	2.50×10^{17}	6.88×10^{16}	1.15×10^{12}	1.14×10^{12}	4.36×10^9
40	2.52×10^{17}	7.02×10^{16}	1.25×10^{12}	1.25×10^{12}	4.49×10^9
50	2.53×10^{17}	7.08×10^{16}	1.30×10^{12}	1.29×10^{12}	4.54×10^9
60	2.53×10^{17}	7.09×10^{16}	1.31×10^{12}	1.30×10^{12}	4.57×10^9

a. Based on 99.5 percent extraction of actinides.

is expected in considering transportation in this cycle. Likewise, existing facilities and machines (e.g., fuel handling) at reactors would be adequate for receiving and charging actinide-bearing UO_2 fuel into and out of the reactor.

The neutronic penalty incurred when actinides are recycled in fresh UO_2 fuel is discussed in Section 9.2.3 and Appendix 9.C. The neutronic penalty associated with the case where the actinides are present in every tenth rod is shown in Figure 9.C.5. The penalty is expressed in terms of the additional U-235 enrichment required to obtain the same reactivity endurance for the fuel rods containing actinides. The incremental en-

richment penalty after 10 recycles will be used to estimate the incremental cost penalty. Based on the calculations summarized in Figure 9.C.5, the enrichment in all the fuel rods must be increased from the base of 3.3 wt% to 3.47 wt%.

9.5.2 Estimated Costs

Recycling actinides in 10 percent of the UO_2 fuel was assumed as the reference case for estimating costs. The estimated annual incremental costs for transmutation of actinides in LWRs are summarized in Table 9.6. The annual fuel costs shown are for the processing and manufacture of actinide-bearing elements. The costs are given for each 1,000 MT of fuel.

TABLE 9.6. Estimated Annual Incremental Fuel Cycle Costs for Transmutation of Actinides in LWRs Assuming Remote Fabrication of 10 Percent of the Fuel

Component	Cost (millions of dollars/year)/1000 MT of Fuel
Partitioning	10
Fabrication	21
Enrichment	14
Total	45

The costs are approximately \$45 million per year per 1,000 MT fuel.

It was assumed that the partitioning step, whereby the actinides are separated from the fission products in the reprocessing plant, adds \$10/kg or \$10 million per year for the reference throughput.

The incremental cost of fabricating fuel elements containing the actinides is very difficult to estimate since little is known about the characteristics of the material and therefore, the fabrication process and plant design. Fuel fabrication costs of \$70/kg were estimated for LWR fuel by escalating the pellet costs reported by Burnham et al.⁽²²⁾ to obtain current costs.

Escalation rates of 5 percent per year for labor and 7 percent per year for building and equipment were used. These escalation factors are probably conservative since increased throughput can more than offset these costs.

For the purpose of this analysis it was assumed that the fuel would be remotely fabricated in a shielded

cell facility. The costs for such fabrication are not well known although some experimental facilities have been operated. It is estimated that an annual fabrication cost penalty would be \$140 - \$280/kg of fuel fabricated remotely. Since only 10 percent of the fuel would have to be fabricated this way, the penalty would be approximately \$21/kg of total fuel fabricated.

The incremental enrichments per recycle and the associated incremental costs are shown in Table 9.7. The total cost for base power is estimated to be around 6.5 mills/kWhe. Using the reference reprocessing plant throughput (5 MT/day) and assuming the reactors operate at 85 percent capacity, the worst case for enrichment penalty (0.050 mills/kWhe) translate to an incremental penalty of about \$14 million per year per 1,000 MT of fuel.

TABLE 9.7. Incremental Fuel Cycle Cost for Increased Enrichment Due to Recycle of Actinides

Recycle Number	Incremental Cost (mills/kWhe)	U-235 Enrichment (wt%)
0	0.0	3.30
1	0.022	3.375
2	0.032	3.402
3	0.0038	3.426
4	0.041	3.440
5	0.043	3.450
6	0.045	3.456
7	0.047	3.462
8	0.048	3.466
9	0.049	3.468

To put these costs in perspective, the incremental cost of \$45 million (Table 9.6) is propagated to an incremental electrical cost. Sixty-1,000 MWe PWRs require about 1,825 MT of fuel per year. These sixty PWRs operating at 85 percent capacity produce 447×10^6 megawatt hours of electricity per year. Assuming electricity sells for 23 mills/kWh (i.e., \$23/MWh), the total income would be \$10.3 billion per year. From Table 9.6 the additional fuel cost required is approximately \$80 million for these 60 reactors. Thus, the cost of electrical power would be increased by 0.8 percent per year. For comparison, it has been estimated⁽²³⁾ that the cost of power for the Tennessee Valley Authority may double to meet all existing and planned pollution limits.

9.5.3 Alternate Fission Schemes Which Might Reduce Cost

The case presented as a base case assumed that 10 percent of the UO_2 rods would contain all the actinides and would be remotely fabricated. In addition, it was assumed that these rods would be designed to be comparable with a standard fuel cycle design. These are design constraints which affect the overall cost penalty.

More detailed calculations may develop more optimum fueling sequences and lower costs. It may be possible to place all the actinides in special target rods which are removable at the end of cycle but can be used initially for power shaping. These rods could have higher actinide concentrations and remain in the re-

actor for longer periods of time. In this way the fabrication cost can be reduced and also amortized over a longer time period. Employing either one of these strategies could result in a significant decrease in the fabrication penalty which is presently the controlling cost.

If no Cf-252 and less than 0.1 percent of the fission products are present in the actinide waste stream, it may be possible to fabricate the actinide-bearing fuel assemblies in a glove box if the actinide concentration is not significantly higher than 0.5 wt% of the total fuel weight. This concentration limit may require the actinides to be spread over all the rods, resulting in an incremental cost of approximately \$17/kg of fuel instead of \$21/kg for the base case. The enrichment cost would be \$10/kg of fuel instead of \$14 in the base case, but the removal of Cf-252 and more fission products would require a cost of approximately \$20/kg instead of \$10 in the base case. The end result is a shifting of expenses but no real change in the total dollar figure (i.e., \$47/kg instead of \$45/kg for the base case).

Recycling actinides in plutonium fuels in both LWRs and FBRs may ultimately be the best solution from a cost standpoint. In addition, since HTGR fuel will be fabricated remotely, the cost penalty projected in LWR fuel fabrication would not be applicable if actinides were recycled in HTGRs.

One of the early studies of plutonium utilization⁽²⁴⁾ considered designing a fuel element which could

be used in both thermal and fast reactors without chemically reprocessing. In light of current technological problems of maintaining structural integrity with zirconium clad LWR fuel and the nuclear waste management considerations, perhaps this type of concept should be re-examined.

The uncertainties in future occupational exposures allowed during fabrication of both LWR and FBR fuel will influence the base costs used to calculate incremental penalties. If these facilities must go to more remote operation, then the optimum strategy utilized for actinide recycle may change. It is quite possible that these uncertainties may actually lower the overall cost penalty associated with the recycle of actinides in a LWR. As a result, the cost penalty associated with the transmutation of actinides may actually be overstated by the values presented in Table 9.6.

Consideration should also be given to evaluating the merit of having special-purpose reactors optimized for destroying actinides that are produced in commercial and government facilities. These reactors could be constructed and operated by the Federal government and the costs of this scheme added as a surcharge in the nuclear fuel cycle.

9.6 PUBLIC RESPONSE

The actinide radionuclides represent the greatest potential long-term risk from high-level waste. The potential risk to the public from these materials is thus present long after the benefits from nuclear power pro-

duction have been received. The potential long-term risk is therefore directly related to the inventory of actinides, and transmutation can reduce this inventory. Thus transmutation can result in lowering the potential risk to future generations. A pilot survey of the public response to alternative waste transmutation management concepts was made.

Of the seven elements of risk (see Section 3 of Volume 1), the ability to have protective reaction, retrievability, and detectability appear to be the most important factors. In the transmutation concept, the most toxic species of nuclear waste are kept in process. By keeping these materials in process, their location at any time is precisely known (i.e., can be easily detected and retrieved). Prior knowledge of accident probability and consequences allows the development of adequate protective reaction measures. The material will certainly be in a stable configuration (e.g., clad fuel rods), and the utilization operations are nearly identical to those in current use in power reactors. The factors of distance and population density should represent no more concern than the present ones for nuclear fuel cycle facilities.

9.7 POLICY CONSIDERATIONS

The transmutation concept is compatible with existing policies and programs. Present federal policy⁽¹⁾ (10 CFR 50, Appendix F) states that liquid "high-level radioactive waste" must be converted from aqueous to a dry solid 5 years after reprocessing

and the dry solid shipped to a federal repository no later than 10 years after reprocessing. High-level waste is defined in this statement of policy as "those aqueous waste resulting from the operation of the first-cycle solvent extraction system, or equivalent, and the concentrated waste from subsequent extraction cycles, or equivalent, in a facility for reprocessing irradiated fuels." For the purposes of transmutation, this definition would have to be modified to the extent that actinides other than uranium and plutonium would not necessarily be classified as high-level waste.

The actinides in waste from other processing steps are classified as low-level waste because they are in dilute concentrations. Ultimately these materials may enter the high-level waste stream. The definitions of by-product, source, and special nuclear materials given in Parts 30, 40, and 70 of Title 10, Code of Federal Regulations and the license requirements for owning and handling these materials do not appear to present legal barriers to the transmutation concept.

9.8 ENVIRONMENTAL CONSIDERATIONS

Of the many transmutation concepts studied, actinide recycle in thermal fission power reactors (LWRs) is the only concept that utilizes an existing operational device. Consequently, the analyses of incremental environmental impact have been limited to this concept.

Environmental analyses consider the impact on the land, water, air

and community. Actinide recycle in LWRs requires unique processing steps and some additional processing in some of the present fuel cycle steps. The additional processing is required because of the enrichment penalty associated with the LWR recycle of actinides. The following discussions identify and quantify the additional processing requirements for the standard fuel cycle components. The incremental environmental impacts of the additional processing steps are also discussed.

9.8.1 Incremental Increase in Fuel Cycle Processing

In the transmutation base case, the U-235 enrichment required to obtain the same fuel cycle length had to be increased to 3.47 wt% U-235 instead of 3.3 wt% U-235. This analysis is based on a requirement for 1,825 MT/year of fuel (supplying sixty 1,000 MW(e) LWRs) processed at this higher enrichment. The additional processing is shown in Table 9.8.

Thus the uranium mine, mill, fluoride conversion, and enrichment

TABLE 9.8. Additional Uranium Required to Supply 1,825 MT/Year of Actinide Recycle Fuel(a)

Additional Natural Uranium	755 MT/Year
Additional Units of Separative Work	910 MT/Year

a. Assuming diffusion tails composition of 0.30 wt% U-235

plants will experience higher throughput but no facility modification for actinide recycle. The remaining components of the fuel cycle, excluding transportation, do not experience any higher throughput although modifications may be required to handle actinide recycle material.

9.8.2 Incremental Impact from Increased Uranium Requirements

The environmental impact from fuel cycle components has been quantified by the AEC.⁽²⁵⁾ The following analysis is adopted from that study. Mining, milling, hexafluoride conversion and enrichment are considered. In every case, the impact is based on the requirement to produce 1,825 MT/year of fuel to recycle actinides in a LWR.

The incremental impact for all the additional processing is shown in Table 9.9. These numbers, although some appear large, represent an increase of about 8 percent in the processing requirements. Thus they do not significantly increase the present impact of the fuel cycle.

9.8.3 Incremental Impact of a Combined Reprocessing and Fuel Fabrication Plant for LWR Actinide Transmutation Fuel

To minimize the amount of actinide transport required to carry out the transmutation concept, the fuel fabrication plant has been located adjacent to the reprocessing plant. This represents a unique fuel cycle arrangement which does not currently exist and therefore has not been evaluated in any environmental statements. In addition, this preliminary

study of transmutation has not been the subject of a detailed pathway analysis; thus dose values reported in this section are very preliminary estimates which must be verified at a later date using methods described in Section 3 of Volume 1. The incremental change in dose received for processing transmutation fuel will be estimated by using a "Toxicity Index." As stated earlier in this report, it is recognized that this procedure gives limited insight on dose estimates.

Two environmental assessments, one for a typical reprocessing plant⁽²⁵⁾ and one for a plutonium recycle fabrication plant⁽²⁶⁾ are used. The effect of plant construction and operation on the land, water and local community is essentially the same as the effect of existing fuel cycle facilities. Because the additional facilities associated with fabrication and partitioning are easily sited in the exclusion radius around a reprocessing plant, the combined plant may actually result in better land utilization.

The actinides produce more heat than conventional fuel; thus this additional heat would have to be dissipated in the environment. The entire annual inventory of 1,825 MT of actinide recycle fuel would generate about 5 MW of thermal energy. Most of this heat would appear in process cooling water. In a properly designed system, there would be no significant environmental impact from that heat load. The process cooling water should contain no radionuclides. Present designs use an intermediate closed cooling loop with its own

TABLE 9.9. Summary of Incremental Environmental Considerations for Uranium Mining, Milling, Fluoride Conversion and Enrichment for Actinide Recycle in LWRs (Based on 1,825 MT/Year of Fabricated Transmutation Fuel for a LWR)

<u>Impact</u>	<u>Mining</u>	<u>Milling</u>	<u>Hexafluoride Conversion</u>	<u>Enrichment</u>
<u>Natural Resource Use</u>				
<u>Land (hectares)</u>				
Temporarily Committed	6.4	0.04	1.0	2.6
Undisturbed area	2.7	0.02	0.9	1.7
Disturbed area	1.2	0.04	0.08	0.7
Permanently committed	0.2	0.16	0.008	0.0
Overburden moved (MT x 10 ⁻⁶)	0.2			
<u>Water (M³ x 10⁻³)</u>				
Discharged to air	-	42.8	14.8	2712
Discharged to ground	81.2	-	-	-
Discharged to water bodies	-	-	156.8	325,500
<u>Fossil Fuel</u>				
Electrical Energy (MW-hr x 10 ⁻³)	0.4	0.47	2.3	2387
Equivalent coal (Mt x 10 ⁻³)	0.016	0.16	0.78	888
Natural Gas (M3 x 10 ⁻⁶)	-	0.33	0.93	
<u>Effluents</u>				
<u>Chemical (MT)</u>				
<u>Gases</u>				
SO _x	0.62	6.4	30.9	3363
NO _x	0.16	2.7	10.3	8875
Hydrocarbons	0.0016	0.23	0.66	87
CO	0.004	0.06	0.20	217
F ⁻			0.12	4
Particulates	0.16	2.7	10.3	8875
<u>Liquids</u>				
Tailings Solutions		41,200		
Ca ⁺⁺				43
Cl ⁻				65
Na ⁺				65
SO ₄ ⁼				43
Fe ⁻				3.0
NO ₃ ⁻				22
Solids		15,860	41.2	
<u>Radiological (Curies)</u>				
<u>Gases (including airborne particulates)</u>				
Rn-222		13.0		
Ra-226		0.004		
Th-230		0.004		
U natural		0.006	0.014	0.015
<u>Liquids</u>				
U & daughters		0.4	0.4	0.15
<u>Solids</u>				
U & daughters		208	0.4	-
<u>Thermal</u> (MW-hr x 10 ⁻³)		4	8	7595

cleanup system to transfer the process heat from the process equipment to the cooling water. Thus there are two isolation barriers separating the process equipment from the heat sink.

The emissions to the air could change because of the additional processing required. Table 9.10 summarizes the results obtained using data from References (25) and (26) for present facilities. These must be adjusted to a combined reprocessing-fabrication facility located at a remote reprocessing site. Table 9.11 summarizes the result for transmutation fuel reprocessed at the combined site. Preliminary estimates show that the releases from the fabrication part of the facility can be neglected in comparison to the actinide releases from the reprocessing plant.

TABLE 9.10. Uranium Fuel Reprocessing-Fabrication Plants at Individual Sites - Gaseous Radioactive Effluents from Processing 1,825 MT/Year

Reprocessing Plant Releases			
Radionuclide	Organ of Interest	Annual Release, Ci	Annual Dose, (a) mrem
Kr-85	Whole Body	18×10^6	0.4
	Skin		27.4
H-3	Whole Body	8×10^5	1.3
I-129	Thyroid	0.12	17.6
I-131	Thyroid	1.2	12.2
Other Fission Products	Whole Body	51	<0.2
Transuranics	Bone	0.21	0.31
Fabrication Plant Releases			
Plutonium	Bone	4.0×10^{-4}	0.60

a. Estimated maximum exposure at plant boundary using currently licensed plants as a basis.

TABLE 9.11. Transmutation Fuel Reprocessing-Fabrication Plant Gaseous Radioactive Effluents from Processing 1,825 MT/Year

Reprocessing Part of Facility			
Radionuclide	Organ of Interest	Annual Release, Ci	Annual Dose, (a) mrem
Kr-85	Whole Body	18×10^6	0.4
	Skin		27.4
H-3	Whole Body	8×10^5	1.3
I-129	Thyroid	0.12	17.6
I-131	Thyroid	1.2	12.2
Other Fission Products	Whole Body	51	<0.2
Transuranics	Bone	0.27	0.60
Fabrication Part of Facility			
Transuranics	Bone	5×10^{-5}	0.0001 (b)

a. Estimated maximum exposure at plant boundary using currently licensed plants as a basis.

b. Reduced dose due to increased distance to site boundary for the combined facilities.

The amount of krypton, tritium, iodine and volatile or semivolatile fission products emitted from the plant should not change significantly. The number of grams of transuranics released annually is expected to be the same for a transuranic plant as for a standard reprocessing plant. However, Claiborne⁽¹⁰⁾ estimates that toxicity index for airborne release has doubled for the composition of transuranics that have been recycled ten times. Thus, the exposure in Table 9.11 for transuranic processing was doubled.

A comparison of Table 9.10 with 9.11 shows that a preliminary estimate of the maximum dose received by an individual at the plant boundary for the current fuel cycle are not significantly changed by transmutation processing.

9.8.4 Environmental Impact from Waste Solidification Processing

Five years after reprocessing, the liquid waste must be solidified. Based on a summary of solidifier operations, decontamination^(a) up to 10^{10} are possible for nonvolatile materials.⁽²⁷⁾ Based on these values, the amount of transuranics emitted from the solidification process can be estimated. Table 9.12 summarizes the estimated emissions of radioactive waste for the LWR fuel and the transmutation fuel after 10 cycles.

The emissions from the waste solidification step for transmutation were estimated assuming only 0.5 percent of the actinides appeared in the partitioned waste stream. The full reduction to 0.5 percent of the LWR levels is not realized because the isotopes build up from successive recycles. This buildup was estimated by Claiborne⁽¹⁰⁾ and was used to adjust the LWR numbers to estimate the

actinide release from solidifying the waste from the transmutation process. The cumulative inventory of fission products is not significantly different because the fuel was taken to the same goal exposure. Thus the incremental risk from solidification is adequately described by considering only the actinides.

9.8.5 Environmental Impact from Transportation

The major environmental impact of actinide recycle on transportation is associated with material processing. Although more uranium enrichment is required, the transportation aspects of uranium shipments have negligible environmental impact. The recycle of actinides represents shipments additional to these, and their shipment results in an incremental impact. In addition, the reduced impact from transporting solidified waste with reduced actinide content is also important.

Actinide recycle will increase the amount of material which has to be shipped between fuel cycle facilities. The doses received along the routes could be significantly greater if no additional precautions were taken. One obvious way to minimize the transportation safety aspects of the problem is to have adjacent construction of reprocessing and transmutation fabrication plants. In this way no transportation of the transuranics would be necessary. The spent fuel casks are presently returned empty to the reactor. They could be used

TABLE 9.12. Radiological Discharges from Waste Solidification (5 Years After Reactor Discharge) (1,825 MT/Year of Fuel)

Element	Waste from LWR Fuel		Waste from Transmutation Fuel	
	Ci/yr	g/yr	Ci/yr	g/yr
Neptunium	5.5×10^{-4}	8.2×10^{-3}	1.8×10^{-6}	6.6×10^{-5}
Uranium	3.3×10^{-7}	8.8×10^{-2}	3.3×10^{-7}	8.8×10^{-2}
Plutonium	9.3×10^{-3}	9.7×10^{-4}	9.3×10^{-3}	9.7×10^{-4}
Americium	3.1×10^{-3}	2.6×10^{-3}	3.3×10^{-6}	1.3×10^{-5}
Curium	3.8×10^{-2}	5.5×10^{-4}	2.0×10^{-6}	2×10^{-5}
Higher Isotopes	1.0×10^{-10}	--	2×10^{-7}	--

a. Decontamination factor is the ratio of material in the product feed to the material released to the environment.

to transport actinide recycle elements. This would imply a two-cycle holdout period for actinides, which is not cost prohibitive. On a semi-annual refueling schedule a 1-year turnaround of actinides would still be possible.

Using these fuel cycle logistics it is estimated that the normal transport of 1,825 MT/year of transmutation fuel elements to and from the reprocessor, a distance of 1,000 miles, would result in an annual whole body dose of 120 mrem/year to the population.⁽²⁸⁾ This figure is double normal operation because twice as many shipments are required. The dose can be no greater than double because the actinide recycle casks will have to be designed to the same external radiation limits as present casks.

The transmutation of actinides could reduce the impact of solid waste transportation in two ways. One reduction occurs because the volume of waste solidified may be less. The other reduction results from the reduction in radiation, particularly neutron, which can be realized by extracting the actinides. Since the actinides make up less than 10 percent of the waste, the reduction in waste flow will not be considered. A reduction in exposure to people along the route from the reprocessing plant to the waste storage site may be realized. At the present time, if the waste from the processing of 1,825 MT/year of LWR uranium fuel is shipped 500 miles to a repository 10 years after reactor discharge, the exposure to the general populace along the route would be 60 mrem/year.⁽²⁸⁾ The amount this could be

reduced by actinide recycle is not known, but it could be significant. In this analysis no reduction is considered.

9.9 SAFETY ASPECTS OF ACTINIDE RECYCLE IN LWRs

The safety aspects of actinide recycle must consider the increased accident potential from the additional uranium requirements and also the accidents from the resulting new processes. The potential for accidents in transportation steps must also be considered.

In evaluating the potential for accidents, the results of higher uranium requirements are not significant. First, the accident potentials from mining, milling, fluoride conversion, and enrichment are quite small. Secondly, the additional uranium requirement is only about 4 percent above the present demand. Thus the increase is relatively small. The potential hazards from the new processing steps are more critical. In particular, accidental releases in the reprocessing plant could be important, and they are discussed separately. The safety aspects of the rest of the transmutation processing steps are included in the following discussion.

9.9.1 Effect of Actinide Recycle in LWRs on Accidents in Fuel Processing Plants

The initial step in the reprocessing plant is fuel dissolution. This is followed by a separation of uranium and plutonium from the all fission products, americium, curium and in some cases neptunium. This entire

stream is called the high-level waste stream. The wastes are then concentrated and normally would be sent to liquid storage tanks. Additional treatment of the high-level waste stream is required to separate the other actinides. This may increase the risk of an accidental release in the reprocessing plant.

A major concern with the additional processing steps is the high level of radioactivity of the actinides. The alpha activity causes decomposition of solvents used in the chemical separation into gases which have explosive potential if they recombined. In addition, organic solvents which increase the probability of a fire in the equipment will probably be required.

The criticality accident at the head end of the reprocessing plant will probably be no more severe than the presently analyzed criticality event since the dose is from gases and semivolatile fission products. The actinides do not fall in that category. The next most frequently analyzed accident in reprocessing plant safety analysis reports is the explosion of a high-level waste concentration tank. Most of the dose from this accident comes from ruthenium. However, curium, if present in increased levels, could add to the hazard potential. The problem of a criticality in the actinide separation steps must also be analyzed. The severity of these potential accidents is not known at the present time.

9.9.2 The Effect of Actinide Recycle in LWRs on Accidents in Other Fuel Cycle Components

Following separation in the reprocessing plant, the actinides would normally be transported to the fabrication plant. If the fabrication plant were in the same building, the risk from any additional transportation step would be eliminated.

In the fabrication plant, the hazard potential associated with recycling both plutonium and the higher actinides would be expected to increase. The release of plutonium during normal operation of an 1,825 MT/year plutonium fabrication plant is 300 $\mu\text{g}/\text{year}$.⁽²⁶⁾ The release of actinides from a similar sized actinide recycle plant would be about 30 $\mu\text{g}/\text{year}$, but the potential hazard represented by this release should be about doubled, due to the nature of material released.⁽¹⁰⁾ This conclusion was obtained from the observation that the actinide inventory in an actinide recycle fuel fabrication plant will be one-tenth the actinide inventory of a similar sized plutonium fuel fabrication plant. It should be noted that it is assumed here that the actinide recycle plant is fabricating uranium fuel, not plutonium fuel.

A spent fuel cask may have to be used for transportation of the recycle assembly from the fabrication plant to the reactor. However, since there is no volatile material in an assembly, the hazards associated with

this transportation step are expected to be quite small.

In the reactor, the increased risk associated with higher actinide recycle is not significantly greater than the risk associated with normal reactor operation. There is no postulated accident that could produce significant quantities of particulates, and thus the introduction of nonvolatile actinides does not increase the risk.

Following irradiation, the fuel must be shipped back to the reprocessing plant. In this step there is no accident postulated which releases nonvolatile materials. Thus transporting the spent fuel should not be an increased risk consideration.

The accident potential for the waste solidification step is quite small. The hazards, although small, would be reduced as a result of transmutation.

This analysis has identified only two areas where the risk of transmutation could be significantly increased. These are at the reprocessing plant and during fabrication. All other fuel cycle components have risks which are either reduced or not significantly different from the risks already associated with those components.

At the present time, the consequences of accidents in reprocessing plants and plutonium fabrication plants are being investigated in a separate AEC study. When this investigation is complete, it should be possible to quantify the increased accident potential for transmutation processing. Until that time, the

risk will be evaluated using only the chronic release emissions.

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APPENDIX

SECTION 8: EXTRATERRESTRIAL DISPOSAL

APPENDIX 8.A

NASA Executive Summary

Feasibility of Space Disposal of Radioactive Nuclear Waste

**NASA TECHNICAL
MEMORANDUM**

NASA TM X-2911

NASA TM X-2911

**FEASIBILITY OF SPACE DISPOSAL
OF RADIOACTIVE NUCLEAR WASTE**

I - Executive Summary

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12. Sponsoring Agency Name and Address National Aeronautics and Space Administration Washington, D.C. 20546				14. Sponsoring Agency Code	
15. Supplementary Notes					
16. Abstract <p>This NASA study, performed at the request of the AEC, concludes that transporting radioactive waste (primarily long-lived isotopes) into space is feasible. Tentative solutions are presented for technical problems involving safe packaging. Launch systems (existing and planned), trajectories, potential hazards, and various destinations were evaluated. Solar system escape is possible and would have the advantage of ultimate removal of the radioactive waste from man's environment. Transportation costs would be low (comparable to less than a 5 percent increase in the cost of electricity) even though more than 100 Space Shuttle launches per year would be required by the year 2000.</p>					
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FOREWORD

An exploratory study to assess the feasibility of sending radioactive waste materials generated by the nuclear power industry into space for disposal was conducted by the National Aeronautics and Space Administration (NASA) and is summarized in two volumes: I - EXECUTIVE SUMMARY and II - TECHNICAL SUMMARY. The study was performed at the request of the Atomic Energy Commission (AEC) as part of a review of various storage and disposal concepts for nuclear waste management.

The study was performed by personnel from various NASA centers, NASA Headquarters, and the AEC. The various sections of the two volumes were written by members of the group and compiled by Robert E. Hyland of the NASA Lewis Research Center. The principal contributors and their respective areas of contribution are as follows:

Robert E. Hyland	Coordinator, package concept and reports
NASA Lewis Research Center	
Robert Thompson	Destinations, vehicles, and trajectories
NASA Lewis Research Center	
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John Vorreiter	Reentry shield
NASA Ames Research Center	
Nathan Koenig	Launch site and facilities
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FEASIBILITY OF SPACE DISPOSAL OF RADIOACTIVE NUCLEAR WASTE

I - EXECUTIVE SUMMARY

National Aeronautics and Space Administration

Lewis Research Center

SUMMARY

The concept of disposing of radioactive waste into space was studied and found to be feasible. Tentative solutions are presented for technical problems of safely packaging the separated long-lived actinide waste. Disposal of these wastes is the primary concern because they will remain radioactive for extremely long times. The package design includes shielding to achieve reasonably low external levels of radiation. The logistics and potential hazards of launching these packages into either high Earth orbits or solar orbits or to escape the solar system have been evaluated. These destinations have been found to be the most promising. Although the solar system escape requires greater energy, it appears to be the most desirable for ultimate disposal.

The total costs of a system for space disposal of radioactive waste are based on the rate of accumulation of fission products and uranium-free actinides in reprocessing plants serving the nuclear power industry and on the launch costs, the destinations, and the launch frequency. The number of waste packages to be launched per year depends on the degree of separation of the long-lived actinides. For example, a package containing about 200 kilograms of separated actinide wastes with about 0.1 percent residual fission products could be ejected out of the solar system for a cost of about \$150 000 per kilogram. Fifty to 100 Space Shuttle launches of such packages per year would be required in the 1990-1995 time period to handle the actinide waste. To this cost must be added the estimated cost of separating and encapsulating the actinide waste. Although the space transportation cost would be several billion dollars per year, the cost prorated over the nuclear electrical capacity is less than 0.1 cent per kilowatt-hour.

A packaging design concept has been evolved that appears on a qualitative basis to provide protection against the radioactive waste in accident environments. The concept, however, does need a follow-up experimental program and safety assessment to establish a system design.

INTRODUCTION

This report (part I) is a condensed summary of an exploratory study (part II) of the feasibility of radioactive waste disposal into space performed by the National Aeronautics and Space Administration (NASA) at the request of the Atomic Energy Commission (AEC). This study was conducted to provide a preliminary assessment of the safety of containment and of launch capability and estimates of transportation costs. It is to be factored in with other studies on potential means for long-term management of high-level radioactive wastes. Battelle Pacific Northwest Laboratories coordinated these studies under contract to the AEC.

RADIOACTIVE WASTE ACCUMULATION

The electric power industry in the United States is projected to have an installed nuclear capacity that may reach 1000 gigawatts electric by the year 2000. The yearly production rate of nuclear wastes that accompany the increasing nuclear capacity in the U.S. is presented in figure 1. The nuclear wastes consist of fission products and actinides (i.e. radioactive elements above actinium, such as neptunium, plutonium, and curium).

Figure 1. - Projected nuclear waste from U. S. powerplants.

Integration of these rates indicates that by the year 2000 about 9000 metric tons of fission products and 1200 metric tons of actinides will have been accumulated. This assumes that no transmutation of actinides has taken place by further in-pile irradiation. The actinide inventory can be reduced to 300 metric tons by separation of essentially all uranium isotopes. This residual actinide inventory is the waste that is considered in the study. Transmutation of actinides, assuming neutron flux levels in typical pressurized water reactors, could reduce this inventory to about one-third if in-pile transmutation were considered feasible. Many of the actinide isotopes have half-lives measured in tens and hundreds of thousands of years. Representative fission products and actinides are described in table 1. These materials represent a long-term hazard to man and must be either stored or disposed of in an acceptable manner. For some of the isotopes with long half-lives, this could mean several hundred thousand years for storage.

TABLE 1. - SOME RADIOACTIVE ISOTOPES WITH LONG DECAY TIMES

Waste	Isotope	Half-life, yr	Decay processes
Fission products	Tritium (^3H)	12.3	Beta (electron)
	Strontium-90	27.7	Beta (electron)
	Technetium-99	2×10^5	Beta (electron)
	Iodine-129	1.6×10^7	Beta (electron), gamma ray
	Cesium-137	30	Beta (electron), gamma ray
	Samarium-151	87	Beta (electron), gamma ray
Actinides	Plutonium-239	2.4×10^4	Alpha (He) particle, gamma ray ↓
	Neptunium-237	2.1×10^6	
	Americium-241	458	
	Americium-243	7.6×10^3	
	Curium-244	18	

SPACE DESTINATIONS

The potential space destinations considered were narrowed down to high Earth orbit, solar orbit, and solar system escape. They are illustrated in figure 2.

(a) High Earth orbit. Velocity increment from low earth orbit, ΔV , 4.11 km/sec; single shuttle launch to 370-km orbit; two burns to $\sim 90\,000$ -km circular orbit (above synchronous orbit); time between burns, ~ 20 hr.

(b) Solar orbit to 0.9 AU. Velocity increment, ΔV , 4.11 km/sec; single shuttle launch to 370-km orbit; two burns to circular solar orbit (0.9 or 1.1 AU); time between burns, ~ 6 months.

(c) Solar system escape. Velocity increment, ΔV , 8.75 km/sec; two shuttle launches to 370-km orbit (one shuttle carries payload and expendable tug, the other carries reusable tug); two burns at perigee; time between burns, ~ 8 hr.

Figure 2. - Potential space destinations.

HIGH EARTH ORBIT

Placing waste packages in high Earth orbits (about midway between synchronous orbit and the lunar orbit) requires a relatively low increment in velocity (4.1-km/sec change in velocity from parking orbit). Daily launch opportunities exist for such flights. Retrieval of waste packages from such orbits is reasonable. Until the long-term integrity of the waste package can be guaranteed, such orbits can be considered as interim storage destinations for only hundreds to thousands of years.

SOLAR ORBIT

Solar orbits (nearly circular at ~ 0.9 AU) can be achieved with a relatively low increment of velocity (4.1 km/sec) and also can take advantage of daily launch opportunities. Their disadvantage is that the circularization burn occurs approximately 1/2 year after injection into the transfer orbit, thereby reducing the reliability of a successful circularization. A malfunction at that time could lead to a possible Earth encounter. Since the long-term stability of such orbits is uncertain, they are not recommended for permanent disposal at this time.

SOLAR SYSTEM ESCAPE

Although direct escape from the solar system requires a high increment in velocity (8.75 km/sec), such disposal of radioactive waste from man's environment is permanent. Furthermore, the integrity of the package is required for a much shorter time period (years as compared with hundreds of centuries) since it will leave our solar system.

The solar system escape launch appears to be the most desirable and was found to be economically and technically reasonable.

OTHER DESTINATIONS CONSIDERED

Sending the waste packages directly into the Sun is not possible with present launch vehicles. Indirect flight could be accomplished with present vehicles by using the more advanced planet swing-by trajectories. However, this is not practical because of limited launch opportunities.

Lunar and planetary destinations were not considered because of the possibility of planet contamination and the very high increment in velocity required for soft landings.

SPACE TRANSPORTATION VEHICLES

The launch vehicles and space tugs considered were those that are available or are being planned and consist of expendable and reusable stages. They are shown in figure 3. The corresponding vehicle launch costs are shown in table 2. The Space Shuttle, in conjunction with either reusable or expendable space tugs, provides the lowest cost per kilogram of payload (total weight of waste package) delivered to the various destinations. Tables 3 and 4 summarize the costs for the various launch vehicles. Because the shuttle is a manned vehicle, its use considerably enhances the

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Figure 3. - Space transportation systems.

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TABLE 2. - SPACE TRANSPORTATION VEHICLE LAUNCH COST FOR RADIOACTIVE WASTE DISPOSAL MISSION

Launch vehicle	Launch cost, dollars
Titan III/Centaur	19.00×10 ⁶
Saturn V/Centaur	155.00
Space Shuttle:	10.50
Reusable tug	1.75
Expendable tug	5.80

TABLE 3. - LAUNCH VEHICLE PERFORMANCE AND COST SUMMARY FOR HIGH EARTH ORBITS AND SOLAR ORBITS.

[Velocity increment, ΔV , 4.11 km/sec.]

Launch vehicle	Payload, kg	Launch cost, dollars/kg
Titan III/Centaur	3 860	4920
Saturn V	32 660	4590
Saturn V/Centaur	35 290	4390
Space Shuttle:		
Reusable tug (current size)	4 170	2940
Reusable tug (optimum size)	4 670	2620
Centaur (current size)	6 490	2460
Centaur (optimum size)	8 480	1920

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TABLE 4. - LAUNCH VEHICLE PERFORMANCE AND COST
SUMMARY FOR DIRECT SOLAR ESCAPE MISSION.

[Velocity increment, ΔV , 8.75 km/sec.]

Launch vehicle	Payload, kg	Launch cost, dollars	Cost, dollars/kg
Saturn V/Centaur	7480	155×10^6	20 720
(2, 1, 1) Shuttle/tug configuration: ^a			
Without perigee propulsion	2270	28.75×10^6	12 660
With perigee propulsion	3270	28.75	8 790
(3, 1, 2) Shuttle/tug configuration: ^b			
Without perigee propulsion	3040	41.0×10^6	13 490
With perigee propulsion	4400	41.0	9 320

^a Two shuttle launches, one expendable tug, one reusable tug.

^b Three shuttle launches, one expendable tug, two reusable tugs.

reliability of the mission from launch to ignition of the tug engine following deployment.

For the waste package mounted on a space tug within the manned shuttle orbiter, a dose level of 1 rem per hour at 1 meter from the surface of the package has been assumed. This value is reasonable and can be further attenuated by distance and by intervening structure in order to reduce the dose to the crew. The waste package will be subcooled prior to launch. Upon reaching orbit its decay heat will raise the package temperature. This heat will be dissipated by radiation to space when the cargo bay doors are opened. Reflectors will be provided in the cargo bay to direct the heat out through the bay door opening.

A typical shuttle launch-to-landing sequence is shown in figure 4. The shuttle launch vehicle is assisted at lift-off by two solid-fueled rocket motors. These are separated and dropped for recovery while the orbiter continues, fueled by the expendable external fuel tank. This external tank is jettisoned and deorbited by a small retrorocket. The orbiter's payload (waste package plus tug) is deployed from the bay of the orbiter. The orbiter later returns and lands at the prescribed landing site. Depending on the destination, the space tug with the waste package either awaits a second tug (solar escape) or initiates its firing sequence to place the package on its desired trajectory.

A method of mounting the space tug with the waste package in, and deploying it from, the orbiter is shown in figure 5. If a malfunction should occur after deployment and before initiation of propulsion by the tug, the orbiter could retrieve either or both. If the malfunction were to occur in later stages of the mission, another tug, capable of retrieving the package, would be dispatched.

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Figure 4. - Space Shuttle launch-to-landing sequence.

(a) Mounted in cargo bay.

(b) Readied for deployment.

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Figure 5. - Space Shuttle orbiter with nuclear waste package and tug.

NUCLEAR WASTE PACKAGE DESIGN

The radioactive wastes from nuclear powerplants can be processed to separate them into two waste streams: fission products, and actinides with residual amounts of fission products. Fission products in various concentrations were assumed to remain in the actinide waste because the cost of complete separation would be too great.

A representative package design is shown in figure 6. The radioactive wastes are contained within a storage matrix which acts as a partial neutron and gamma shield as well as a heat-conducting medium. The actinide waste is in the form of small spheres approximately 3.5 millimeters in diameter. The spheres are coated with a refractory metal and an oxidation-resistant material for retention of radioactive waste at high temperatures. The matrix, containing approximately 10 percent actinides by volume, is enclosed in a sphere of stainless steel to protect it against impact and fragmentation. This sphere also contains layers of neutron and gamma shielding material. The impact protection sphere is enclosed within an aerodynamically stable reentry body designed to

Figure 6. - Representative nuclear waste package.

TABLE 5. - WEIGHT BREAKDOWN OF TYPICAL
NUCLEAR WASTE PACKAGE

[Solar system escape for actinides.]

Component	Weight, kg
Actinide waste	200
Matrix containing waste	625
Gamma shield	1190
Neutron shield	180
Impact sphere	640
Reentry body (heat shield)	410
Total	3245

survive reentry heating. This reentry body consists of two layers. The outer layer is a composite fiber of quartz woven into a mat with a silica binder that acts as a highly reflecting medium for steep-angle reentry protection. The inner layer is composed of 3D graphite to handle the convective heat load from shallow-angle reentries.

A biological dose constraint of 1 rem at 1 meter from the surface of the waste package was assumed for the configuration that was designed for solar escape. The package is thus weight optimized to contain about 1 kilogram of waste for every 30 kilograms of total package weight when actinide wastes contain 1 percent residual fission products. As the composition of fission products is reduced to 0.001 percent, the weight fraction of actinide waste increases to 1 kilogram in every 10 kilograms of package weight. These optimized weights are essentially independent of space destination. A representative package weight breakdown is presented in table 5. For some of the heavier payloads considered, the heat generated by the radioactive waste was a limiting factor in the design of the waste package. The waste package design concept presented in part II of this report would be applicable for disposal of other compositions of radioactive waste.

TYPICAL DISPOSAL MISSION

The sequence of events for a typical waste disposal mission to solar system escape is as follows:

- (1) Launch shuttle 1 to 370-kilometer parking orbit.
- (2) Deploy reusable tug to rendezvous position.
- (3) Launch shuttle 2 to 370-kilometer parking orbit.
- (4) Deploy expendable tug and waste package to rendezvous with reusable tug.
- (5) Maneuver tugs to dock in tandem configuration.
- (6) Reusable tug fires to required ΔV , separates, and returns to shuttle 2.
- (7) Expendable tug fires and injects waste package into solar system escape trajectory.

The major components involved in such a mission are shown in figure 7.

Component	Weight, kg
Nuclear waste package: Waste (actinides plus 0.1 percent fission products) Shielding (LiH, W, matrix) Impact sphere Reentry body (heat shield) Adapter	200 1 995 640 410 120
Space Shuttle: Orbiter (dry weight) Liquid propellant and tank Solid rockets	68 000 737 000 1 030 000
Reusable space tug: Propellant weight Burnout weight	23 900 2 900
Expendable space tug: Propellant weight Burnout weight	22 000 2 900

Figure 7. - Component weights for nuclear waste space disposal mission. Required for mission: one shuttle carrying reusable space tug, and another shuttle carrying expendable space tug and nuclear waste package.

LAUNCH FREQUENCY

The frequency of Space Shuttle launches required is an important factor in considering the space destinations, the costs, and the launch facility requirements. For each radioactive waste composition and each disposal package design, the number of required annual shuttle launches was determined through the year 2010 for the three space destinations described. The high Earth orbits and the circular solar orbits require approximately the same number of annual flights and are plotted together in figure 8. This figure is for the extreme case of disposing of all fission products that have been ground stored for 10 years.

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Figure 8. - Number of Space Shuttle launches required per year for disposal of all fission products into solar orbit or high Earth orbit. Prior 10-year Earth storage.

Figure 9. - Number of Space Shuttle launches required per year for disposal of only actinides into high Earth orbit or by solar system escape. Prior 10-year Earth storage.

After 1990, more than one launch per day would be required. This launch frequency is not considered practical at this time.

The launch frequencies required for space disposal of only the separated actinides are more reasonable, as shown in figure 9. Required launch rates vary from less than 10 to 350 per year through the year 2010 depending on the fission product composition of the actinides and the destination.

With the launch facilities that are available and that could be made available, as many as 140 launches per year are possible. The estimated cost for additional equipment and facilities to handle this many launches per year is \$230 million. This cost includes two new launch pads.

SPACE TRANSPORTATION COSTS

The launch costs for the shuttle and tugs, presented in table 2, are the overriding space transportation costs. These costs, coupled with the packaging cost (\$650/kg of actinides) and the expense of additional launch facilities (estimated at \$70 000/flight for 140 flights/yr), determines the costs for transportation of radioactive waste to the space destinations considered. (The cost of separating the fission products from the actinides is not included here.) The waste disposal per mission and the space transportation costs are presented in table 6. To present these costs in perspective, they may be put in terms of the additional power cost to the consumer (i.e., space transportation costs per kW-hr of electrical power generated in producing the nuclear waste). The space transportation cost for the disposal of all the fission products is 1 to 5 cents per kilowatt-hour. For disposing of only the separated actinides, the cost is 0.01 to 0.1 cent per kilowatt-hour. The cost depends on the space destination and on the composition of residual fission products contained within the actinides.

The results of an optimization study that balanced estimated fission product separation costs against waste package transportation costs are shown in figure 10 and point to a fission product composition of less than 1 percent as desirable. Compared with the present cost of electricity, the space disposal of the separated actinide wastes represents less than a 5 percent increase in power costs to the consumer.

Adding the estimated cost of separating fission products to the cost of transporting the waste to space yields the total cost. The optimum total cost, 0.1 cent per kilowatt-hour, occurs for an actinide waste containing about 0.1 percent fission products and for disposal beyond our solar system.

The total annual costs for transporting actinides containing 0.1 percent fission products after a 10-year temporary storage on Earth, as shown in figure 11, range from \$30 million to \$5 billion per year.

TABLE 6. - TRANSPORTATION COSTS FOR DISPOSAL OF RADIOACTIVE WASTE

Type of waste	Destination for disposal	Amount of waste disposed of per mission, ^a kg	Transportation cost, ^b dollars/kg
Fission products	Earth orbit or solar orbit	189	88 000
	Solar system escape	73	394 000
Actinides plus 1 percent fission products	Earth orbit or solar orbit	288	57 000
	Solar system escape	113	255 000
Actinides plus 0.1 percent fission products	Earth orbit or solar orbit	447	37 000
	Solar system escape	200	151 000
Actinides plus 0.001 percent fission products	Earth orbit or solar orbit	858	19 000
	Solar system escape	308	94 000

^a Mission launch system: for high Earth or solar orbit, Space Shuttle with Centaur (optimum size); for solar system escape, two Space Shuttles, one reusable tug, and one expendable tug.

^b Includes cost of packaging and additional launch facilities but not the separation cost.

Figure 10. - Optimization of costs for space disposal of actinide waste by solar system escape.

Figure 11. - Annual space transportation cost for space disposal of actinide waste.

SAFETY

The safety goal for nuclear waste disposal in space is to transport the radioactive waste to an acceptable destination in such a manner that potential radiation exposures and contamination are negligible.

The accident conditions considered and the responses of the design waste package are summarized in table 7. In all cases the response of the waste package to the proposed accidents indicates that the release of radioactive waste would be prevented by the various protection shells designed into the total waste package. The package response analysis was verified, where possible, by simulation experiments. However, much additional development and testing are required to confirm the design concept.

TABLE 7. - POSSIBLE ACCIDENTS AND PACKAGE RESPONSES

Type of accident	Accident condition	Package response
Blast overpressure	150 atm	No yielding to 175 atm
Fragmentation	Fragments up to 1070 m/sec	No penetration to 1360 m/sec
Fireball	2750° C, 20 sec	No melting
Residual fire	2400° C, 5 min	Outer stainless-steel layer near melting
Reentry heating	300 kW/cm ² , 3 to 4 sec	Sufficient thickness
Impact on earth, water, or concrete	300 m/sec	Some deformation, no release
Postimpact	Deep burial	Outer vessel rupture due to pressure after about 5 days
	Deformed - no burial	Integrity maintained

With an appropriate package design and launch operation, the overall risks are expected to be low. Since the mission hardware and launch parameters were of a preliminary nature only, a risk assessment on a quantitative basis could not be performed.

The key requirement for the overall safety of waste disposal missions is early recovery of the waste package in the event of an accident during any phase of the mission. For most accidents the early recovery could be handled satisfactorily. For some accidents, particularly an uncontrolled abort occurring in the later stages of a mission (i.e., after deployment and prior to the tug achieving the required ΔV), recovery from space may be difficult if not impossible.

CONCLUSIONS

GENERAL

The results of this exploratory study indicate that space disposal of the long-lived radioactive actinides from nuclear waste appears feasible from the viewpoint of both economy and safety. The transportation costs for ejecting the actinides out of the solar system, for example, would represent less than 5 percent increase in the consumer bill for electric power generated by nuclear powerplants. Such missions involve certain risks, however small, which would have to be balanced against the benefits to be derived from removing the long-lived radioactive waste from man's environment and thus relieving future generations of the responsibility of protecting themselves against our radioactive waste. Quantitative evaluation of the risks requires more study, development, and testing.

SPACE DESTINATIONS

Of the destinations considered, three look promising: high Earth orbits (above synchronous orbit altitude), nearly circular solar orbits inside the Earth's orbit, and solar system escape. Only the last destination provides a permanent disposal of the nuclear waste. It is therefore the most promising destination, even though the costliest. Sending the waste directly into the Sun is not within the capabilities of present vehicles. Sending it into the Sun with acceleration assists from planetary swing-by is not practical.

SPACE TRANSPORTATION VEHICLE

The currently planned Space Shuttle, in conjunction with space tugs, provides a substantially lower cost per kilogram of waste delivered to the space destinations than any of the current expendable launch vehicles. Because the shuttle is manned and has considerable maneuvering capability, the overall safety aspects of such a transportation system could be superior to those of expendable launch vehicle systems.

WASTE PACKAGE DESIGN CONCEPT

The nuclear waste package design allows sufficient radioactive waste per package for economic disposal and should prevent release of radioactive waste under the accident conditions reviewed. Further study could optimize the design to increase the waste content and to better define its limitations.

SAFETY

No quantitative risk assessment was possible because the mission hardware and the mission parameters are preliminary. Only a qualitative evaluation was performed. This evaluation indicated the design could prevent release of radioactive waste under conditions imposed in accident environments. With appropriate system design and launch operations, the risks involved are expected to be relatively low.

COSTS

The transportation costs for space disposal of radioactive actinides would represent an increase in the consumer's electric costs of approximately 5 percent. To this transportation cost must be added the cost for separating the actinide waste and the fission product waste. Preliminary data from a study conducted by Battelle Pacific Northwest Laboratories for the Atomic Energy Commission indicate that the separation costs will be of the same order or less than the costs of transportation out of the solar system. Both the space transportation cost and the launch frequency are feasible and practical for the disposal of separated actinide waste. However, the space disposal of all fission product waste is neither economically nor practically feasible at this time because the large quantities would require an excessive launch rate.

Lewis Research Center,
National Aeronautics and Space Administration,
Cleveland, Ohio, October 25, 1973,
770-18.

APPENDIX 8.B

Study of Extraterrestrial Disposal of Radioactive Wastes
Part II

**NASA TECHNICAL
MEMORANDUM**

NASA TM X-68147

NASA TM X-68147

**STUDY OF EXTRATERRESTRIAL DISPOSAL
OF RADIOACTIVE WASTES
PART II**

by R. E. Hyland, M. L. Wohl, R. L. Thompson,
and P. M. Finnegan
Lewis Research Center
Cleveland, Ohio
October 1972

STUDY OF EXTRATERRESTRIAL DISPOSAL
OF RADIOACTIVE WASTES

Part II

Preliminary Feasibility Screening Study of Extraterrestrial Disposal
of Radioactive Wastes in Concentrations, Matrix Materials,
and Containers Designed for Storage on Earth

by R. E. Hyland, M. L. Wohl, R. L. Thompson, and P. M. Finnegan

Lewis Research Center

1. INTRODUCTION

AEC has initiated a program aimed at providing near-term and long-term solutions to the problems associated with the handling and management of radioactive wastes. Battelle-Northwest has been requested to conduct studies and to make evaluations of all currently envisioned long-term waste management methods.

The objective of these efforts is to identify feasible long-term waste management systems and their components; identify the research and development necessary for their establishment; and estimate the schedule and costs associated with selected systems. In addition, these studies will be used as the basis for providing a discussion of alternatives in the statement of environmental impact required for authorization of a Federal waste repository project.

The concepts to be studied include:

- (1) The application of alternate geologic storage techniques.
- (2) An international off-shore repository.
- (3) Storage in the seabed.
- (4) Use of the permanent ice caps.

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- (5) A ten-mile deep hole.
- (6) A deep cavity generated by a nuclear device.
- (7) Extraterrestrial transport.
- (8) Transmutation.
- (9) Other methods as yet unidentified.

NASA has been requested by the AEC to study the feasibility of extraterrestrial transportation of radioactive wastes. More specifically, NASA has been requested to study the extraterrestrial transport of at least three types of radioactive waste materials:

- 1. Radioactive wastes in concentrations, matrix materials, and containers currently designed for storage on Earth.
- 2. Actinide wastes with 0.1 and 1 percent contamination by other radioactive wastes.
- 3. The third type to be defined later in the study.

The general approach in each of these studies will be similar. The studies will be divided into several phases. The first phase is a preliminary feasibility screening study. This phase will establish the maximum amount of the particular radioactive waste that could be transported to space per launch. It will also establish the minimum cost of disposing of this particular waste in space and estimate the number of launches required per year. The effect of integration of the package with a vehicle and accident conditions will not be treated in this phase. The waste disposal container will be designed considering primarily normal operation. The primary emphasis will be on heat transfer, radiation shielding, and criticality. If this preliminary feasibility screening study indicates that the cost is reasonable, then a phase II feasibility study will be conducted which will include integration with the launch vehicle and consideration of all the safety aspects of the study. If the phase I study indicates that this maximum waste payload and minimum cost system may not be feasible, then the phase II study will be postponed until other more promising space disposal systems are considered.

If the method appears feasible after the phase II feasibility study, then the third phase of the study would be conducted. This phase would identify the research and development necessary to demonstrate feasibility and estimate the schedule and costs associated with the development and operation of the system.

This report describes the results of the phase I preliminary feasibility screening study for the first class of radioactive wastes, that is, the radioactive wastes in concentrations, matrix materials, and containers currently designed for storage on Earth.

2. DESCRIPTION OF WASTE PRODUCTS, MATRIX

MATERIALS AND CONTAINERS

2.1 Description of Waste Products

The radioactive wastes in this report are the fission products that remain after processing as indicated in references 1 and 2 followed by a ten-year hold in temporary storage. This processing separates the fission products from the unfissioned fuel, structural materials, and the actinide class of radioactive wastes. However, the fission products after processing still contain small amounts of these materials. Their effects are considered negligible in this report.

Figure 2-1 shows how the activity (curies/gm) and the thermal power (watts/gm) of the fission products vary with time after discharge from the reactor. Both have decayed by about a factor of ten by the end of the ten-year hold in temporary storage. At that time about half the heat is generated by three isotopes, ^{90}Y , ^{137}Cs , and its daughter ^{137}Ba .

Table 2-1 shows the characteristics of fission products from light-water reactors (LWR) and from liquid metal fast-breeder reactors (LMFBR). For the purposes of this study the important characteristics are the amount of fission products per MWe day and the thermal power and activity per gram of fission products. LMFBR waste products have less thermal power and activity per gram because the isotopic distribu-

tion of the fission products produced by fast and thermal reactors are somewhat different. In addition, the amount of fission products produced per MWe day is less for the LMFBR because the expected efficiency of the plant is expected to be higher, 40 percent compared to 33 percent for the LWR. For reference, the LMFBR produces 2.64 gm/MWe day compared to 3.23 gm/MWe day for the LWR.

2.2 Description of Matrix Materials

Four types of solid-matrix materials designed for storage on Earth are described in Table 2-2 (ref. 3). The four types are spray melt, pot calcine, phosphate glass, and fluid-bed calcine. The spray melt was selected for this study. It was selected because its activity (curies per unit volume) is high, yielding a more compact disposal package. In addition (1) it has a tough structure compared to crumbly, brittle, and granular structure of the other materials permitting it to remain intact in case of impact, (2) it has a high maximum stable temperature, 1170 K compared to 1170, 770, and 870 K for the other materials, allowing it to operate at higher temperatures thus permitting larger diameters and higher payloads, and (3) it has a comparatively high-thermal conductivity, 1.8 watts/m(K) compared to 0.5 and 1.8 for the other materials, permitting high heat removal rates or large diameters without exceeding centerline temperatures.

Spray melt appears to be the best of the four candidate matrixes presently available but it has several drawbacks from the space disposal viewpoint. It is desirable to have a material with a higher thermal conductivity, higher maximum stable temperature, and higher values of curies per unit volume. Any future studies should consider other matrix materials when they become available.

2.3 Description of Fission Product Storage Containers

The containers designed for Earth storage of fission products in the above matrix materials are cylinders. The cylinders are made of a non-reactive material which would be stainless steel for pot calcine and either stainless steel or mild steel for the spray melt, phosphate glass, and fluid bed calcine. Diameters of 6 to 24 inches and lengths of 8 to 10 feet are being considered (ref. 3). The diameter for Earth storage is selected primarily to assure the maximum centerline temperature of the matrix material is below the maximum stable temperature. The centerline temperature will be higher for the larger diameters. The lengths are selected on the basis of requirements such as loading, handling, and maintenance.

In the design of a cylindrical container for space disposal the diameters will also be selected to assure that the temperature of the matrix material does not exceed the maximum stable temperature, which is 1170 K for spray melt. The diameter of the vessels will, however, be permitted to exceed 24 inches if this is advantageous. The length of the cylinder will be determined by the maximum payload of the launch vehicle or the space tug, whichever is limiting. Table 3-1 shows the maximum allowable payload weights for the candidate combinations of payload, tug, and launch vehicle. The information in this table will be discussed in the next section.

Figure 2-2 is a schematic drawing of the cylindrical containers configured for space disposal. The fission products are contained in the inner stainless steel cylinder. This cylinder is surrounded by a depleted uranium gamma shield. The cylinder with the gamma shield is then inserted in an outer stainless steel radioactivity containment vessel. Both the inner and outer vessels are welded and helium leak checked for tightness. For the purposes of this study all materials are assumed to be in intimate contact so that the temperature drop across the interfaces can be neglected. This is an optimistic assumption and the effect will have to be checked if the concept warrants a phase II feasibility study.

The uranium shield was selected because it gave the lowest shield weight. However, uranium changes phase (α to β) when its temperature goes above 930 K and the phase change causes the material to expand prohibitively. The maximum normal operating temperature is about 600 K. The feasibility of using depleted uranium for the gamma shield would be reconsidered in the phase II study because abnormal and emergency conditions may cause the shield temperature to exceed the phase change temperature.

3. PAYLOADS, COSTS, AND DESTINATIONS FOR CANDIDATE SPACE VEHICLES

This subject is discussed in detail in reference 4 "Space Transportation Considerations for Disposal of Radioactive Wastes" by J. Ramler, R. Thompson, and S. Stevenson. This section summarizes some of the pertinent information in this report and discusses the reasons for selecting the shuttle as the launch vehicle and Earth escape as the destination for this study.

The candidate destinations starting with the highest ΔV requirements and lowest payloads are: direct solar impact, direct solar escape, solar impact via Jupiter, solar escape via Jupiter, solar orbit, solar orbit via Venus, solar orbit via Mars, Earth escape and Earth orbit. The candidate expendable vehicles starting with the highest payload capability are: Saturn V/Centaur, Saturn V, and Titan III E/Centaur. There is one shuttle design but two types of shuttle launches. One is a single shuttle launch which carries both the payload and the tug. The tug transports the payload from low Earth orbit to its final destination. The other requires two or more shuttle launches. One carries the payload. The other shuttle or shuttles carry one tug each. The payload and tug (or tugs) rendezvous and are assembled in low Earth orbit. The tugs may be either expendable or reusable.

Table 3-1 shows the payloads and costs for the candidate destinations and candidate launch vehicle and tug combinations. This table shows that direct solar impact is not possible with today's vehicles. It also shows that the shuttle is the most economical launch system.

The shuttle was therefore selected as the launch vehicle. Earth escape¹ was selected as the destination for this phase. High Earth orbit permits carrying about the same payload but was not selected because this destination was considered to be too near the Earth. This was an arbitrary decision and will be re-evaluated with other promising destinations in later phases of the study when other aspects, in addition to payload, are considered in the selection of the destination.

The trajectories used to calculate the above payloads were not dog-legged. Dog-legged trajectories may be required to avoid potential impact on land after abort during ascent. Dog-legging the trajectory requires more fuel and, if the shuttle is fully loaded, the payload decreases. When the trajectories are dog-legged, the payload for the single shuttle launch may be reduced more than that for the dual launch as follows: In the single launch case, the shuttle is fully loaded and thus the final payload decreases for missions with a dog-leg. In the dual case the tug-carrying shuttle is fully loaded but the waste carrying shuttle is only partially loaded, 34 000 pounds, compared to a maximum allowable of 62 000 pounds. The payload is limited to 34 000 pounds because the maximum the tug can put into Earth escape orbit is 31 000 pounds (the 3000 pound difference is due to structures that remain in the shuttle). Thus, 28 000 pounds of additional fuel could be carried for the dog-leg maneuver without reducing the payload. The second shuttle with the tug may not have to be dog-legged since land impact of the tug is less hazardous. If the shuttle with tug is not dog-legged, the

¹Earth escape is a solar orbit obtained by one burn from Earth orbit. The important characteristic of this orbit is that it intersects the Earth's orbit and introduces the possibility of Earth impact. Solar orbit refers to orbits about the Sun which are either inside or outside the Earth orbit with negligible probability of impacting the Earth.

maximum payload for the dual launch mode will not be much lower than 31 000 pounds. This is the main reason for looking at the dual shuttle launch mode of operation. Without such a potential advantage the single shuttle would be preferred because it is a less complicated launch mode.

The cost comparison was made using existing expendable vehicle designs. If space disposal appears feasible, then the development of an expendable vehicle should also be considered. Cost savings may result due to mass production and potentially higher payload capacity.

4. ANALYSIS

The phase I analysis has two main parts. First, determination of the maximum amount of fission products that could be carried in the shuttle-orbiter-tug vehicle to earth escape. Second, calculation of the launch cost per pound, per curie, per MWe day, and per kw-hr electric to establish the effect of launch cost on the electric generating cost. The number of launches required in 1985 and 2000 are also estimated. If the effect on electric cost makes the system potentially not feasible, then the phase II feasibility study would be postponed until phase I studies on potentially more promising waste-matrix-container systems have been completed.

If the results of the phase I study indicate this waste-matrix-container combination to be potentially feasible, then a phase II feasibility study would be conducted which would include design for in-shuttle cooling and for off-normal, emergency and accident conditions and would consider shuttle safety environment, abort, re-entry, impact, and heating after impact. The following sections describe the design criteria, procedure, and assumptions for the phase I analysis.

4.1 Design Criteria

For this phase of the study three classes of criteria are considered: radiation dose levels, matrix material temperature limits, and shuttle

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payload limits. In the double shuttle launch where the payload and tug are placed in orbit on separate launches, the maximum payload is determined not by the shuttle but by the tug. The tug limit is based on what payload it can take to a destination or put on a trajectory.

Radiation dose levels. - Dose levels in three situations were considered:

Shuttle crew	2.5 mrem/hr in the crew compartment which corresponds to 10 rem/hr at 3 meters from the center of the container
Shuttle instrument dose for unmanned shuttle	10^7 rad to the nearest instrumentation which corresponds to 500 rem/hr at 3 meters from the center of the container
After accident public exposure	1 rem/hr at 3 meters from the center of the container

Temperature limits. - Calculated spray melt matrix material temperature shall not exceed 1100 K which is 70 K below the maximum stable temperature for spray melt.

Shuttle payload limits for Earth escape. -

Single shuttle launch	Payload	17 000 lbs
	Structure on payload	500
	Structure in shuttle	3 000
	Tug	<u>44 500</u>
	Total	65 000
Two shuttle launch		
Shuttle 1	Tug	59 000
	Structure in shuttle	<u>3 000</u>
	Total	62 000

Shuttle 2	Payload	30 000
	Structure on payload	1 000
	Structure in shuttle	<u>3 000</u>
	Total	34 000 lbs

4.2 Procedure and Assumptions for Waste Container Design

The steps for a phase I design of a container and shield which meets the normal operation dose, temperature, and payload criteria are listed below:

1. Select a set of container diameters in the range-proposed for Earth storage. Diameters of 6, 12, 18, 24, and 28 inches were selected.
2. Calculate the radiation and heat source for a selected activity concentration and container diameter.
3. Calculate gamma shield thickness for one of the dose criteria assuming uranium metal as the shield material.
4. Calculate the surface temperature of the container in orbit assuming an emissivity of 0.8.
5. Calculate the temperature in the center of the matrix material assuming no temperature drop across the material interfaces.
6. Calculate the weight per unit length of payload where payload is the fission product, matrix material, containment cylinder, and shield.
7. Calculate the weight of the end shields and containment vessel end caps and subtract from the allowable payload to get the weight of the center section of the payload.
8. Calculate the length of the cylinder which makes the payload weight equal the payload criteria.
9. Calculate the weight of fission products per launch.
10. Perform above calculations for three dose rate constraints, three activity concentrations and for single and double shuttle launches.

4.3 Procedures and Assumptions for Calculation of Space Transport Cost

The cost of space disposal of radioactive wastes can be divided into several categories.

- (1) Temporary storage on Earth.
- (2) Separation, concentration, and preparation of wastes in matrix materials.
- (3) Design and fabrication of the space disposal container system and assembly of wastes and matrix material into the container.
- (4) Shipment of wastes to the launch site.
- (5) Space transportation cost.

This report is concerned with only one of these costs - the space transportation cost. The space transportation costs begin when the payload is delivered to the launch site. The major space transportation costs end when the payload gets to its destination. The costs, however, may not go to zero at this time. There may be additional monitoring costs depending on the disposal destination.

The other (all of the above) costs will have to be determined before a complete economic analysis can be made. The purpose of the present analysis is to determine the relation of the space transportation cost to the cost of generating electricity. Specifically, the space transportation cost will be compared with the bus-bar cost of electricity which is assumed to be 8 mills/kw-hr.

The factors that affect the space transport cost to the electric consumer are:

- (1) Launch cost including shuttle and tug.
- (2) Destination and gross payload.
- (3) Ratio of radioactive waste to gross payload.
- (4) Interest on funds collected and set aside for space disposal at the end of Earth storage time.
- (5) Radioactive waste Earth storage time.

The steps for a phase I economic analysis are:

1. Determination of the gross payload for the candidate destination which is Earth escape in this study. Gross payload is defined as the weight of the waste container system delivered to the destination and includes all structures and auxiliary systems fixed to the container.
2. Determination of the net waste container payload by subtracting the weights of the structures and auxiliary systems fixed to the waste container from gross payload.
3. Determination of the amount of fission products that can be carried in a container whose weight including the shielding equals the net payload.
4. Determination of the launch cost including shuttle and tug.
5. Determination of the cost per curie of fission products transported to space.
6. Determination of the discounted space transportation cost per curie disposed, that is, determine the amount of money per curie that could have been put in a trust fund for space transportation. This assumes that the consumer was charged for space transportation when he used the electricity and that the money was put in a trust fund and compounded at current interest rates.
7. Determine the amount of electricity (MWe days) that was generated per curie disposed.
8. Determine the cost of space transportation of wastes in units of mills per kw-hr of electricity.
9. Compare the cost of space transportation from step 8 (mills/kw-hr) to the bus-bar generating cost of 8 mills per kw-hr.

5. RESULTS AND DISCUSSION

This section has four main parts. Part one establishes the net payload and launch cost as a function of the destination and launch vehicle. The next part describes the design of the waste payload package

and discusses the effect of the design parameter on the dimensions of the container, the container temperature and thermal power, and the amount of fission products per launch. The third part describes the launch costs in mills per kilowatt hour of electricity and discusses the effect of container design parameters, earth storage time, and interest rates on the mill/kw-hr cost. The fourth part estimates the required number of shuttle flights per year to 2010 AD and discusses the effect of the destination and the design parameters on the number of launches.

5.1 Gross Payload and Launch Cost

For this phase I feasibility screening study Earth escape was selected as the disposal destination and the shuttle was selected as the launch vehicle. Gross payload is defined as the waste container plus the structure attached to it. At this time single and dual shuttle operations appear to have equal feasibility and are both considered. In single shuttle operation both the waste payload and tug are carried in the same shuttle. In dual shuttle operation one shuttle carries the waste payload and the other carries the tug. The following table summarizes the payload and cost data for Earth escape.

Vehicle	Gross payload, wt. (lb)	Launch cost, \$	Cost per pound of gross payload, \$/lb
Single shuttle	17 500	11 M	628
Dual shuttle launch	31 000	21 M	677

Payload and Cost Data for Earth Escape

The costs per pound of gross payload are \$628 and \$677 and are essentially the same within the accuracy of this study. The selection of single shuttle or dual shuttle operation will be made at a later time and will depend on additional considerations, for example, ratios of waste

weight to gross payload weight (this tends to increase with increasing gross payload), effect of dog-legging the trajectory (this tends to reduce the single shuttle payload more), the complexity of the dual operation compared to single shuttle operation and safety considerations.

For reference, Table 3-1 shows the payloads and costs for the other candidate launch vehicles and destinations. The candidate destinations starting with the highest ΔV requirement (lowest payload) are: direct solar impact, direct solar escape, solar impact via Jupiter, solar escape via Jupiter, solar orbit, solar orbit via Venus, solar orbit via Mars, Earth escape, and Earth orbit. The candidate expendable vehicles starting with the highest payload capability are: Saturn V/Centaur, Saturn V, and Titan III E/Centaur.

When selecting the candidate destination and launch vehicle, other factors besides payload and launch cost must be considered. For example, the Jupiter, Venus, and Mars fly-by missions require less ΔV but more accurate instrumentation and control than the more direct missions. In addition, Jupiter, Venus, and Mars are in the proper positions for launch for only about one month every 12, 19, and 25 months, respectively. Thus the fly-by missions would require all the launches for the 12 to 25 month period be made in about one month. This would require several launches per day during about a 1/2 hour launch window. More detailed discussion of these aspects can be found in reference 4.

Earth escape, Earth orbit, and solar orbit result in the highest payloads per vehicle but each has drawbacks that must be investigated. In the case of Earth escape the possibility of re-encounter with the Earth at some future time must be made negligibly small for thousands of years due to the long-life of the waste materials. In the case of Earth orbit the possibility of interference with other space activities must be studied and made acceptable. Solar orbit reduces these problems but requires additional burns later in the mission as does Earth orbit. Solar system escape would eliminate these problems but would be costly. These types of considerations are discussed by Ramler, Thompson, and Stevenson in reference 4. More detailed analysis of this type, integrated

with safety and economic analysis is required before a destination can be firmly selected.

5.2 Waste Payload Design

The physical features of the waste, matrix materials, and containers are described in Section 2, Table 2-2, and figures 2-1 and 2-2. The design procedures and assumptions for the container and shield were described in sections 4.1 and 4.2. The results of the parametric analysis of these designs are presented in figures 5-1 through 5-8 and are discussed below.

There are three main categories of design criteria:

(1) Radiation dose rates of 1, 10, and 500 rem/hr at three meters from the container centerline.

(2) Centerline temperature less than 1170 K which is the maximum stable temperature for the spray melt matrix.

(3) Gross payload weight: 17 500 pounds for single shuttle and 31 000 pounds for dual shuttle operation.

Additional independent parameters are:

(1) Radioactivity concentration in the matrix to a maximum of 10 curies/cc.

(2) Earth storage time.

The dependent parameters are:

(1) Diameter of the waste plus matrix material.

(2) Diameter of the outer containment vessel.

(3) Length of the container.

(4) Payload thermal power.

(5) Containment system outer surface temperature.

(6) Amount of radioactive waste in the container.

Diameter of waste matrix. - This diameter was determined the requirement that the matrix centerline temperature not exceed the maximum stable temperature of the spray melt matrix material which is 1170 K. Figure 5-1(a) and (b) show the design point diameter for the

matrix material to be about 28 inches for an activity concentration of 10 curies/cc and a radiation dose of 10 rem/hr and 500 rem/hr. The matrix diameter is the same for both dose rates due to two compensating effects. First, the lower dose rate requires a thicker shield and, for the same matrix material diameter, the temperature drop through the thicker shield is higher. Second, the thicker shield makes the outer container diameter larger and this reduces the surface heat flux and the surface temperature. These two effects essentially cancel each other and the 28 inch matrix material diameter satisfies the maximum centerline temperature requirement independent of dose in the range considered, which was 1 rem/hr to 500 rem/hr.

Diameter of outer containment vessel. - This diameter was determined by the gamma shield thickness for the three dose rates considered and a 1 inch thick impact shield as outer shell. The shield material was depleted uranium and its normal operating temperature was about 600 K. The uranium shield thickness, for the design dose rates of 1, 10, and 500 rem/hr were 4.5, 3.5, and 2 inches, respectively. These thicknesses were determined by the comparison method, using data from reference 5. For all doses the matrix material diameter was 28 inches and the inner containment vessel thickness was 1/2 inch. Figure 5-2 shows the outer diameter as a function of dose rate. The outer diameter is 40, 38, and 35 inches for dose rates of 1, 10, and 500 rem/hr.

Length of container. - Figure 5-3 shows the length² of the container as a function of the dose rate and for single and dual shuttle payloads of 17 000 and 30 000, respectively. The lengths for a 17 000 pound payload and for dose rates of 1, 10, and 500 rem/hr are 2.59, 3.33, and 5.21 feet, respectively. The length for a 30 000 pound payload and for dose rates of 1, 10, and 500, rem/hrs are 5.09, 6.35, and 9.63 feet, respectively.

Thermal power. - Since the matrix material diameter is a constant and independent of the dose rate, the thermal power generated per foot

²The length includes the active matrix plus the end shielding and container thicknesses.

of cylinder is also a constant of 6 kw/ft. The length of the container is determined by the allowable payload and the radiation dose as discussed in the previous section. Figure 5-4 shows the thermal power as a function of dose rate for single and double shuttle launches. The thermal power for a single shuttle payload of 17 000 pounds and dose rates of 1, 10, and 500 rem/hr are 9.6, 15.1, and 27.8 kilowatts, respectively. The thermal powers for a dual shuttle payload of 30 000 pounds and for the same dose rates are 24.6, 33.2, and 54.3 kilowatts.

Containment vessel outer surface temperature. - The power per foot of capsule is a constant as discussed in previous sections. The surface temperature is then a function of the dose rate which defines the container diameter and radiating area per foot. Figure 5-5 shows the surface temperature as a function of dose rate. The surface temperature for dose rates of 1, 10, and 500 rem/hr, 3 meters from the container centerline was 480, 500, and 525 K, respectively.

Amount of radioactive waste in the container. - The weight and amount of radioactivity per foot of capsule is the same for all capsules because the activity concentration is constant at 10 curies/cc and the waste in matrix diameter is constant at 28 inches. The amount of fission products per container is a function of the maximum allowable payload and the dose criteria. Figures 5-6 show the amount of fission products in the container in curies and the packaging weight ratio for single and dual shuttle launches and for radiation dose rates from 1 to 500 rem/hr at 3 meters from the container centerline. The number of Megacuries for dose rates of 1, 10, and 500 rem/hr at 3 meters are 1.91, 3.02, and 5.56 for single shuttle payloads of 17 000 pounds. The number of Megacuries for a dual shuttle payload of 30 000 pounds for the same dose rates are 4.90, 6.62, and 10.9.

5.3 Space Transportation Cost

The factors that affect the space transportation cost and the procedures and assumptions for calculating that cost are described in

Section 4.3. The purpose of the analysis is to estimate the space transportation cost to the electric power consumer and to compare this cost to the electric cost, which is 8 mills per kw-hr at the bus-bar and 24 mills per kw-hr average to the residential consumer.

The effect on the transportation cost of each of the main parameters in the cost analysis will be determined. The parameters, the baseline values for the parameters, and range of variation of the parameters are listed in table 5-1. The effect of a parameter will be determined by varying the parameter, keeping the other parameters fixed at the baseline value. The results are presented in table 5-2 and the effect of the following parameters on the space transportation cost are discussed below:

- (1) Destination
- (2) Dose rate
- (3) Earth storage time
- (4) Space disposal fund interest rate
- (5) Activity concentration

Effect of destination on cost. - The gross payload and the cost per pound of gross payload are presented in table 3-1 for the candidate destinations and the required vehicles. Gross payload is defined as the waste container and the structure attached permanently to it. The ratio of radioactive waste to gross payload depends on the character of the waste and the design of the container. The cost per pound of gross payload for the candidate destinations is listed below:

Earth escape, \$/lb of gross payload	628
High Earth orbit	628
Solar orbit via Mars or Venus	794
Solar orbit	800
Solar escape via Jupiter	3500
Solar impact via Jupiter	4700
Direct solar escape	4420
Direct solar impact	Payload is zero with existing vehicles

The effect of the destination on space transportation cost in terms of mills per kW hr electric is presented in table 5-2 for several destinations. The cost for Earth escape, solar orbit, and solar escape is 4, 5, and 28 mills/kw-hr.

Effect of dose rate. - The effect of varying the dose rate from 1 rem/hr to 500 rem/hr was determined for the Earth escape destination and is shown in figure 5-7. The cost per pound of waste delivered for the 10 rem/hr dose is 65 percent of the 1 rem/hr cost. The cost for the 500 rem/hr dose is 33 percent of the 1 rem/hr cost.

Effect of Earth storage time. - The effect of Earth storage times of 10, 20, and 40 years was determined and is shown in figure 5-8. The cost to the electric customer goes down as the storage time increases primarily due to the increased interest accumulated on the disposal fund. The cost of storing the material is small compared to the interest on the fund and is neglected in this analysis. The activity of the waste decreases with time which also tends to reduce the cost of disposal (less shielding, more waste payload). However, unless the nonradioactive decayed materials are removed from the waste to keep the curies per cc near the original level, the shield weight savings will be small. Time affects the cost in another and more significant way. Interest on the funds set aside for waste disposal increase much faster than the fission products decay. The time for 10 year old fission products to decay by half is about 30 years. The money doubling time is about 10 years at a seven percent interest rate. The effect on cost due to fission product decay during storage is neglected. Only the effect of interest on money in the disposal fund is considered. The cost per pound of waste delivered for the 20 year storage case was about half the 10 year cost and the 40 year storage time was about 1/8 of the 10 year cost. At a 7 percent interest rate the charge to the customer is reduced by half for each ten year storage time. Therefore, a storage time can be found for each destination which will make the initial charge to the electric consumer acceptable.

Effect of space disposal interest rate. - The interest on the space disposal fund can affect either the space transportation cost or the required storage time. The effect of interest rates of 5, 7, and 10 percent were determined and are shown in figure 5-8. The cost at a 7 percent interest rate for materials stored ten years is about 1.3 times that at 10 percent and about 80 percent of that at 5 percent. To get the same transportation cost at 5 and 10 percent as for 7 percent with a storage time of 10 years requires a storage time of about 14 years at 5 percent and 7 years at 10 percent.

Effect of activity concentration. - Increasing the concentration of the radioactive waste has a strong effect on the transportation cost but the amount the concentration can be increased is limited. Doubling the concentration would reduce the cost by about 45 percent. In general, the gain is not this great because the diameter must be reduced to keep the centerline temperature below the maximum stable temperature. The base case in this study had a concentration of 10 curies per cc and the full density fission products have a density about 26 curies/cc after 10 year storage. Thus going to full density fission products would reduce the costs by possibly more than half. The costs could be further reduced by removing the gamma emitters and/or the high thermal energy emitters and launching the harmful wastes that remain. The cost of launching separated radioactive wastes will be considered in later reports.

5.4 Number of Shuttle Launches per Year

The production rate of ten-year old fission products as a function of years from 1970 to 2000 is shown in figure 5-9. The amount of fission products that can be carried per launch is a function of

- (1) Destination
- (2) Dose rate
- (3) Fission product concentration

Figure 5-10 shows the number of launches per year to 2010 AD for Earth escape, single or double launch mode, Earth storage for ten years, and three dose levels. In 1985 the required number of launches for dose levels of 1, 10, and 500 rem/hr at 3 meters from the package center are 300, 210, and 115, respectively. The effect of the other parameters on the number of launches per year is shown in table 5-2. The payload to Earth orbit is about equal to the payload to Earth escape and the number of launches is also similar. The payload to solar escape is about 10 percent of the payload to Earth escape and the number of launches would be increased accordingly. Increasing the Earth storage time to 40 years decreases the total activity by 50 percent. The number of launches is decreased accordingly if the fission product concentration is assumed to be maintained at 10 curies/cc thus keeping the curies per launch constant (i.e., more grams of fission products for the same dose level with longer storage time). An additional decrease in launch frequency could be obtained by removing the decayed isotopes after a long storage time.

It appears that launching of all fission products at an early time period results in a higher cost and high launch frequency. Both can be avoided by holding for a longer time followed by separation.

6. CONCLUSIONS

For this report all of the fission products (i.e., no separation) were considered for space disposal after being stored in Earth-storage facilities for 10 years. The fission products were assumed to be mixed in a solidified matrix material and contained in cylinders. These cylinders were sized based on the temperature limits on the matrix material and shielded to reduce the radiation dose rate to levels ranging from 1 to 500 rem/hr at 3 meters from the center of the package. In this report the impact of accidents on safety was not considered, and thus the conclusions obtained pertain only to the package as designed for normal operations. This implies minimum cost and maximum quantity of fission

products per payload. The payloads were based on results of a previous study (ref. 4) in which the shuttle was selected as the lowest-cost vehicle. The destinations chosen for the report for comparison were Earth escape, Solar orbit, and Solar escape. The following conclusions were obtained from the results presented.

1. Matrix material such as spray melt can be used without exceeding temperature limits on matrix, but materials with higher thermal conductivity would be more desirable. Diameters of 28 inches or less were acceptable but not optimum based on fission products per package or cost.

2. The cost in terms of mills per kw hr electric, of space disposal of fission products (after 10 year temporary storage) in matrix materials and containers currently designed for Earth disposal and shielded (1 rem/hr) is 4, 5, and 28 mills per kw hr for Earth escape, solar orbit, and solar escape, respectively. This compares to 8 mills per kw hr bus-bar cost and about 24 mills per kw hr average consumer cost.

3. A major factor effecting cost was the Earth storage time. Assuming 7 percent interest on the funds set aside for space disposal, the cost to the electric consumer of space disposal is reduced by a factor of 2 for each 10 years of storage time. If the fission products are stored for 40 years prior to launch then the cost to the electric consumer are 0.5, 0.6, and 3.5 for Earth escape, solar orbit, and solar escape, respectively. There is, therefore, for each destination, a storage time that will make the initial charge to the electric consumer acceptable. Based on a normal operating condition design for solar escape, a storage time of more than sixty years is required to make the space disposal charge less than 10 percent of the bus-bar electric cost.

4. Large changes in dose rate are required to significantly affect the cost. Increasing the dose at 3 meters from the center of the package from 1 to 500 rem/hr results in factor of 3 reduction in cost.

5. The number of shuttle launches would exceed a launch per day within 5 years after the program was initiated if the material was launched as prepared for Earth storage and held for 10 years without further processing.

Inasmuch as fission products will decay with time, both the space transportation cost and number of launches can be reduced considerably by increasing hold time. Large reductions in launch costs might possibly be achieved if the fission products are separated and only, say, the actinides are launched into space. The actinides, in particular, present a special hazard if they are permanently stored on the Earth because they have such very long half-lives. The extent to which this principal hazard can be reduced at low launch cost warrants further study.

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TABLE 2-1. - FISSION PRODUCT YIELDS FROM TYPICAL POWER REACTORS

	LWR	LMFBR
Power density per ton of fuel loading	30 Mwt/metric ton	58 Mwt/metric ton
Thermal energy yield per ton of fuel loading	33 000 Mwt-days/metric ton	33 000 Mwt-days/metric ton
Efficiency	33 percent	40 percent
Electric energy yield per ton of fuel loading	10 900 Mwe-days/metric ton	13 200 Mwe-days/metric ton
Fission product mass per ton of fuel loading	3.51×10^4 gm/metric ton	3.49×10^4 gm/metric ton
Fission product mass per Mwe-day	3.23 gm/Mwe-day	2.64 gm/Mwe-day
Fission product activity after 10 year hold	28.5 curies/Mwe-day	21.2 curies/Mwe-day
Radioactivity per gram after 10 year hold	8.83 curies/gm	8.03 curies/gm
Gamma ray	3.0 curies/gm-0.7 Mev E_{avg}	
Beta ray	5.8 curies/gm-0.4 Mev E_{avg}	
Fission product thermal power/gm after 10 year hold	0.0286 watts/gm	0.023 watts/gm

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TABLE 2-2. - CHARACTERISTICS OF WASTE IN MATRIX MATERIALS
DESIGNED FOR STORAGE ON EARTH

	Pot calcine	Spray melt	Phosphate Gl.	Fluid-bed calcine
Form hardness	Cake, crumbly soft	Single, tough mold hard	Single, brittle mold very hard	Amorphous-granular moderate
Leachability in water, gm/cm ² day	1.0 to 10 ⁻¹	10 ⁻³ to 10 ⁻⁶	10 ⁻⁴ to 10 ⁻⁷	1.0 to 10 ⁻¹¹
Fission-prod oxides in mixture, mole %	Up to 80	Up to 30	Up to 25	Up to 50
Thermal condition, watt/M(K)	0.25 to 0.5	0.8 to 1.8	0.7 to 1.8	0.2 to 0.5
Density, gm/cc	1.1 to 1.5	2.7 to 3.5	2.7 to 3.0	1.0 to 1.7
Maximum stable temper- ature, K	~ 1170	~ 1170	~ 770	~ 870
Container material	Stainless steel	Mild or stainless steel	Mild or stainless steel	Mild or stainless steel
Maximum curies/cc	~ 10	~ 9	~ 7	~ 8

8.B.25

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TABLE 3-1. - CANDIDATE LAUNCH VEHICLES, DESTINATIONS, COSTS, COSTS PER POUND OF PAYLOAD

Candidate launch vehicle and tug combinations	Launch cost ^a	Destinations									
		Earth orbit or Earth escape		Direct solar escape		Solar impact via Jupiter		Solar escape via Jupiter		Solar orbit	
		Payload, wt (lb)	Cost per pound, \$/lb	Payload, wt (lb)	Cost per pound, \$/lb	Payload, wt (lb)	Cost per pound, \$/lb	Payload, wt (lb)	Cost per pound, \$/lb	Payload, wt (lb)	Cost per pound, \$/lb
Shuttle (1) and reusable tug (1)	11 M	17 500	628	0	0	0	0	0	0	10 400	1058
Shuttles (2) and reusable tug (1)	21 M	31 000	677	0	0	0	0	0	0	15 500	1355
Shuttles (2) and reusable tug (2)	22 M	37 300	590	0	^b 0	0	0	0	0	27 700	794
Shuttle (1) and expendable tug (1)	25 M	24 000	1041	2 700	9 259	5 300	4 717	7 000	3571	19 400	1288
Saturn V/Centaur	200 M	108 000	1852	16 400	12 195	25 700	7 782	32 000	6250	84 200	2375
Saturn V	200 M	10 000	2000	0	0	12 400	16 130	20 100	9950	78 000	2564

^aCost does not include guidance for tug.

^bShuttle (2), expendable tug (1), and reusable tug (1) 8140 lb, cost/lb = \$4420.

8.B.26

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TABLE 5-1. - RANGE OF PARAMETERS FOR SPACE
TRANSPORTATION COST ANALYSIS

Parameter	Baseline value	Range
Destination and gross payload		Solar orbit (10 400 lb)
Single shuttle	Earth escape (17 500 lb)	Solar escape (2700 lb)
Dual shuttle	Earth escape (31 000 lb)	Solar orbit (15 500 lb)
		Solar escape (2700 lb)
Dose rate at 3 meters from container center	1 rem/hr	10 and 500 rem/hr
Launch cost		
Single shuttle	11 million dollars	No range
Dual shuttle	21 million dollars	
Earth storage time	10 years	20 and 40 years
Interest on space disposal fund	7 percent	5 and 10 percent
Activity concentration	10 curies/cc	20 and 40 curies/cc

8.B.27

TABLE 5-2. - RESULTS OF SPACE TRANSPORTATION COST ANALYSIS FOR SINGLE SHUTTLE

Parameters for single shuttle launch mode	Cost			Amount of fission products per launch		Number of shuttle launches	
	Mills per kilowatt hour	Percent increase over base rate		Pounds	Megacuries	1985	1995
		Bus-bar at 8 mills	Consumer at 24 mills				
Baseline parameter case, Earth escape 1 rem/hr, 10 yr, 7%, 10 curies/cc	4	50	17	380	1.52	285	1520
Destination							
Solar orbit	5	62	21	304	1.21	342	1825
Solar escape	28	350	116	58	.23	1860	9900
Dose rate at 3 meters from container center							
10 rem/hr	2.6	32	11	600	2.45	180	960
500 rem/hr	1.3	16	5.5	1120	4.57	97	520
Earth storage time							
20 years	2.1	26	9	380	1.52	----	----
40 years	.53	6.6	2.2	380	1.52	----	----
Interest on space disposal fund							
5 percent	4.9	61	20	380	1.52	285	1520
10 percent	3.1	38	13	380	1.52	285	1520
Activity concentration 20 curies/cc	2.1	26	9	730	2.96	150	790

FIG. 2-1 FISSION PRODUCT ACTIVITY AND
THERMAL POWER AS A FUNCTION
OF TIME AFTER PROCESSING
SPENT FUEL ELEMENTS

8.B.30

Figure 2-2 Typical Radioactive Container and Shielding for Space Disposal.

Fig. 5-1a Temperature at Center of Cylindrical Containers
Containing Fission Product Waste Material Stored for
10 Years . Shielded with depleted uranium for 10rem/hr
at 3 meters from the center of the cylinder.

Fig. 5-1b Temperature at Center of Cylindrical Containers
Containing Fission Product Waste Material Stored for
10 Years. Shielded with depleted uranium for 500rem/hr
at 3 meters from the center of the cylinder.
($k = 1.7$ watts/meter $^{\circ}\text{K}$)

FIGURE 5-4: THERMAL POWER VS. DOSE RATE AT 3 METERS FROM CENTERLINE FOR SINGLE AND DUAL SHUTTLE LAUNCH PAIRDALE OF 11,000 AND 30,000 POUNDS.

Fig. 5-6b PACKAGING RATIO FOR ALL FISSION PRODUCTS - PHASE I
ASSUMING 10-YR EARTH STORAGE

Fig. 5-7 Effect of Dose Rate on Space Disposal Cost
 for the Baseline Fission Product Case.

Fig. 8-8 Effect of Earth Storage Time and Interest Rate on Space Heater Product Disposal Cost for the Baseline Case.

9. A.32

This value is not considered as an absolute bound but rather a practical bound. If this performance could not be attained the accelerator probably would not be built. Thus the energy required is estimated to be

$$\frac{23 \text{ MeV}}{\text{FP}} < \frac{E}{N} < 110 \text{ MeV/FP} \quad (\text{A37})$$

This required electrical energy must be less than the electrical energy produced in a fission power reactor per candidate transmutation fission product. This energy balance is strictly true only as long as electrical power is being obtained from fission reactors. The electrical energy produced per FP can be obtained from the production rates given on the Chart of the Nuclides. These values are estimated for a 1000 MW(e)-yr, slightly enriched uranium, 25,000 MWD/T fuel exposure with 85% duty cycle. The values given for candidate FP

Cs-137 41.6 Kg

Sr-90 18 Kg

Tc-99 27.5 Kg

correspond to the following values of electrical energy produced per FP nucleus:

Cs-137 900 MeV/FP

Sr-90 1370 MeV/FP

Cs-137+Sr-90 550 MeV/FP

Tc-99 985 MeV/FP

Cs-137+Sr-90+Tc-99 350 MeV/FP

APPENDIX 8.C

An Evaluation of Some Special Techniques
for Nuclear Waste Disposal in Space

**NASA TECHNICAL
MEMORANDUM**

NASA TM X- 62,272

NASA TM X-62,272

**AN EVALUATION OF SOME SPECIAL TECHNIQUES FOR
NUCLEAR WASTE DISPOSAL IN SPACE**

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ABSTRACT

This note presents a preliminary examination of several special ways for space disposal of nuclear waste material which utilize the radioactive heat in the waste to assist in the propulsion for deep space trajectories. These include use of the wastes (or an extract of the ^{90}Sr or ^{137}Cs compounds contained in the waste) in a thermoelectric generator (RTG) which operates an electric propulsion device and a radioisotope - thermal thruster which uses hydrogen or ammonia as the propellant. These propulsive devices are compared to the space tug and the space tug/solar electric propulsion (SEP) combination for disposal of waste on a solar system escape trajectory. Such comparisons indicate that the waste-RTG approach has considerable potential (disposing of perhaps four times as much waste) provided the combined specific mass of the waste container - RTG system does not exceed approximately 150 kg/kw_e .

Although this study stresses the solar system escape destination, several exploratory numerical calculations have been made for high Earth orbit and Earth escape destinations. These show that some care must be exercised in selecting an Earth escape path in order to avoid future near encounters with the Earth or Venus. In general, it is believed that useful calculations are possible using numerical integration which could help in an orbit or trajectory selection process.

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AN EVALUATION OF SOME SPECIAL TECHNIQUES FOR
NUCLEAR WASTE DISPOSAL IN SPACE

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Introduction

If the projected future United States power demands are to be partly satisfied by stationary nuclear power plants, then there will be an associated increase in the amount of nuclear waste material which results from the reprocessing of the spent reactor fuel elements to recover the unused fuel. While such reprocessing is an inherent part of the economical operation of such nuclear power stations, it can lead to surprising amounts of radioactive residue. Reference 1, for example, contains some projections which lead to an annual output of over 5×10^5 kg of waste by the year 2000. This, of course, depends on the electrical power demands continuing to increase as they have in the past and also that no new power producing methods (such as nuclear fusion reactors) emerge to meet the increasing demand.

As a result of such nuclear waste possibilities, the Atomic Energy Commission has asked NASA to study the feasibility of disposing of the waste products in space. Reference 1 and its associated documents constitute a direct response to the AEC request. This memorandum has been stimulated by that effort but is not an official part of the response to the AEC.

The purpose of this paper, then, is to consider several concepts by which the energy still contained in the nuclear waste material could be utilized to augment or complete the space disposal process of such waste. As indicated in figure 1 (taken from ref. 1), it can be seen that the energy output per unit mass (shown on the ordinate in figure 1) is about 300 w/kg if the waste is obtained at one year from the time of reprocessing.

8.C.4

This is a specific energy value equivalent to that of ^{238}Pu . However the nature of the waste is such that (see ref. 2) it is much less dense (in its solid form) and much more difficult to shield. The radiation is due to the high percentage of short lifetime radioactive elements in the waste. As indicated in figure 1, the energy output eventually becomes an exponential function of time. This represents a transition to activity from a few dominant, longer half life elements (i.e. ^{139}Cs and ^{90}Sr). This is shown more clearly in figure 2 (taken from ref. 3).

Thus, it would appear that there is at least an interesting amount of energy in the waste if it can be obtained early enough (i.e. the specific energy output is similar to that of isotopes usually used in space applications). Another possibility is the separation of special high heat output elements out of the waste. This will be considered as an alternate but more expensive way of utilizing the heat in the waste.

Two propulsive techniques will be considered which use the waste heat. One is the direct conversion of the heat into electricity by a thermoelectric generator (RTG) and the other is heating of some working fluid such as liquid hydrogen or ammonia and subsequently expelling the fluid at high velocity to produce thrust. This is similar to the "poodle" thruster concept described in reference 4, and will be referred to here as an isotope thermal device. Another somewhat related concept--solar electric propulsion (SEP) plus waste (RTG)--will also be considered but not evaluated in quantitative terms. These various concepts are illustrated in figure 3.

Waste Material Form

Considering the energy output only, it would follow that early acquisition and containment of the solid waste would be desirable. This would utilize the high energy output of the short half-life elements before they decay to less active states. However, it is recognized that the existing processing facilities may restrict the acquisition time to one or two years after reprocessing of the fuel elements. Thus, one form of

8.C.5

the waste products which will be considered here will be the solid state one and two years after reprocessing.

Of the various solid forms which are currently being considered (see ref. 2) their density varies between 1.33 and 2.8 gm/cm³. In reference 1, the spray melt solidification process was selected as a desirable form for packaging and heat conductivity purposes. The properties of this type of solid waste are listed in table 1.

TABLE 1. SOME PROPERTIES OF SPRAY MELT SOLID WASTE MATERIAL
(from refs. 1 and 2)

Heat output (one year)	300 watts/kg
Heat output (two years)	150 watts/kg
Density	3.0 gm/cm ³
Thermal conductivity	1.8 watts/cm ³ /°K
Maximum (center line) temperature	1170°K

These properties will be used here as those of typical solidified but otherwise unprocessed waste material.

The second waste form which will be considered here is the separation of ¹³⁷Cs and/or ⁹⁰Sr from the main waste stream. This could be done at about five years or more after initial reprocessing operation. At the present time, facilities for separating these elements out of the waste stream do exist (see ref. 5); thus the cost of increasing output or purity may not be excessive. However, the additional cost of performing the separation must still be included.

The purpose of separating out these isotopes is that they have a high specific energy output combined with a rather long half life. Some properties of these isotopes in their common (usually oxide) forms are given in table 2.

8.C.6

TABLE 2. SOME PROPERTIES OF ^{137}Cs and ^{90}Sr ISOTOPE COMPOUNDS
(from ref. 6)

Isotope	Shield Density* cm of Uranium	Density gm/cc	watts/gm	Half-life years
^{90}Sr (SrO)	2.3	4.7	0.453	27.7
^{137}Cs (CsF)	5.5	3.586	0.134	30

*The shield densities shown in this table are computed for a spherical 1 kw_t source and are for 10 rem/hr at 1 meter from the center of the source.

^{90}Sr , for example, has an energy output similar to ^{238}Pu which is often used as a heat source for flight RTG power supplies, but has a much more severe shielding problem and other safety disadvantages. However, both isotopes are relatively good heat sources if separated from the rest of the solid waste. Also, they could be bothersome to store on Earth because of their relatively long half lives.

Another waste form which also results from special processing is the separation of the actinide compounds from the waste material. These isotopes have very long half lives which implies extensive ground storage time if they are left in the waste. However, the heat output is very low and as such, does not constitute an interesting heat source for propulsive or power generating purposes. Rather it probably represents the most compact form the waste can take without utilizing some form of nuclear transmutation. However, the actinides are only the most troublesome part of the waste material and facilities for the storage or use of the rest must be provided.

Because the actinides represent only a fraction of the total waste, they are probably best considered along with more conventional space disposal techniques such as the shuttle/tug or shuttle/centaur. Use of such launch vehicle systems is being examined by the NASA, Lewis Research Center (see ref. 1) and will not be considered here.

Disposal Destinations

A number of destinations for the proposed waste containers are currently being studied. They include high Earth orbit, Earth escape, 0.99 AU and 1.1 AU circular orbits about the sun, solar system escape and solar impact (see figure 4). A comparison of the propulsive energy requirement of these destinations can be found in reference 1.

The nearby destinations such as high Earth orbit or Earth escape are of interest because the propulsive energy expenditure required to achieve such orbits is low and would probably be most attractive as a destination for chemical rocket systems such as the space tug or the centaur. However, they may also be of interest for the waste heated fluid concept as such devices may have low specific impulses, depending on the working fluid used (e.g., ammonia). However, while such destinations may be easy to reach, they create another problem regarding whether or not the waste is actually disposed of in such a case. This is particularly true of the Earth escape case where there is some chance, however small, that the waste container may someday return. Several exploratory numerical integrations were carried out in order to illustrate some of the problems that can arise in certain cases. Specifically, the cases so far studied are Earth escape and high Earth orbit.

All numerical integrations have been performed on a CDC 7600 computer using a version of the LeRC N-Body program (see ref. 7). The CDC 7600 has a 60 bit word length which allows single precision 14 digit arithmetic. Thus, very accurate numerical integrations are possible without the usual need for double precision arithmetic or accumulation.

Considering first the high Earth orbit case, it was first determined that the important perturbations were those due to the moon, sun and the Earth's oblateness. Inclusion of Jupiter and several other planets had little noticeable effect after several years of integration. The predominant changes in the orbit's elements were precession of the

line of nodes and the argument of pericenter. The orbit chosen for study was circular at 50,000 nautical miles altitude and inclined at 28.5° to the equatorial plane. These calculations for the Earth orbit case were very time consuming, requiring about 25 seconds of computer time per year of orbit time.

The Earth escape case was less expensive, using less than 50 seconds of computer time for 500 years of interplanetary flight. The orbit under investigation was an Earth escape trajectory with a perigee altitude of 100 nautical miles and an eccentricity of 1.1.

The main results for the Earth escape case were that some care must be taken to keep the orbit as much inside the Earth's orbit as possible. That is, Earth departure conditions should be such that the trajectory enters heliocentric space at aphelion. Otherwise it was found the trajectory would re-enter the Earth's sphere of influence several times within a 100 year span. On one other occasion a trajectory passed through the sphere of Venus at 273 years during a 514 year integration even when special care is taken to inject at aphelion. This indicates that care must also be exercised in selecting the trajectory perihelion. In the Earth escape cases, it was found necessary to include all the planets out to Jupiter. More planets could not be included because of present limitations of the program.

In all cases the planets and the moon were included with fixed orbit elements chosen from some particular epoch. This is a serious omission only in the case of the moon, which precesses around the Earth at the rate of about $18^\circ/\text{year}$. However it has become clear that useful calculations can be made which can very likely be of value in orbit selection and simulation.

Unlike the low energy cases, there are at least two other destinations which probably constitute true disposal. They are solar impact and solar system escape. Of these two, solar system escape is perhaps

the most preferable because of the generally lower energy requirements. One objection to solar system escape is that it may become someone else's problem in due time. However, the time to reach the nearest star is enormous and could leave the package no more harmful than a meteorite. (A detailed treatment of the probable hazards associated with these destinations can be found in ref. 10.)

Thus, it would appear that the most preferable destination is solar escape with solar impact a second choice. Of the other destinations, high Earth orbit or solar orbit are perhaps the least likely to return to Earth. However, the preliminary numerical calculations which have been completed for the Earth orbit case have indicated that the orbit will precess (not unlike the moon) due to solar, lunar and Earth oblateness perturbations. Thus, it may be difficult to track the waste containers for the hundreds or perhaps thousands of years which may be required by safety considerations.

Consequently, this section will consider only solar escape and impact as likely destinations for the propulsion systems under consideration herein. The other destinations will be given ample consideration (in ref. 1) and need not be considered here in any further detail.

It has been shown in reference 1, direct solar impact requires a velocity relative to the Earth of 30 km/sec. This stops the package relative to the sun and it falls straight down on a radial line to impact. Very few propulsion systems presently under consideration (with the possible exception of the laser ignited fusion device described in reference 8) could accomplish such a mission. For example, the waste-RTG and solar electric propulsion systems can simulate such a mission only by a slow spiral into the sun. Unfortunately, the effective velocity change for such a maneuver is the difference between the circular orbit speeds at the different radii. Thus, to reach the surface of the sun (a radius of 0.698×10^6 km) would require:

$$\Delta V = V_{C,\odot} - V_{C,\oplus} \approx 436. - 30 = 406 \text{ km/sec} \quad (1)$$

where V_C is the orbital speed at the indicated distance, which is probably beyond the capabilities of any ion thruster system.

A more optimal approach, even for chemical rocket systems, would be to first proceed outward to some high aphelion and then nullify the velocity at aphelion and drop into the sun. This, as well as the direct method, is illustrated in figure 4. The limit in this process is to first essentially escape the solar system (i.e., very high aphelion) and then apply a very small correction and return to solar impact. Unfortunately, the time involved in such a maneuver is excessive and some compromise must be made between the time and ΔV involved in the maneuver. Figure 5 illustrates the interchange between time of flight and ΔV .

Thus, it is clear that low energy (i.e. low ΔV) solar impact missions and solar escape are closely related and considering one is equivalent to considering the limiting case of the other. For this reason, only solar escape will be considered in the following sections.

Radioisotope Waste RTG System

As noted previously, it is best to use the waste early. Suppose, for example, that we obtain the waste at one year; then, from figure 1:

$$P/m = 0.3 \text{ kw/kg}$$

$$\alpha_{\min} = \frac{1}{P/m \eta_C} = \frac{1}{.3 \times .05} = 67 \text{ kg/kw} \quad (2)$$

where

$\alpha_{\min}$ = minimum specific mass of the power supply

η_C = efficiency of thermoelectric converter

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As indicated, this assumes that the RTG converter efficiency is 5 percent, which is typical of present day technology.

However, since the value of P/m falls off so rapidly in figure 1, some average value should be chosen. To do this, it will be assumed that the electro-static thruster system can operate no longer than 20,000 hr. (Again, this is typical of current estimates from test and flight data. A general description of electrostatic thruster developments and operations can be found in reference 11.) Thus, the value of P/m taken from figure 1 should be between one and three years. This gives an average $\alpha_{\min}$ of about 134 kg/kw.

Assuming the thruster efficiency to be of the form:

$$\eta_{th} = \frac{B}{1 + \left(\frac{D}{C}\right)^2} = \frac{0.842}{1 + \left(\frac{16}{C}\right)^2} \quad (3)$$

where

C = ion exhaust velocity, km/sec

B = propellant utilization efficiency

D = ionization loss factor, km/sec,

and that the propulsion time (t_p) is limited to 20,000 hr, a value of C can be found which gives the highest initial acceleration. This is given by the relation:

$$C_{opt} = \sqrt{\frac{2000 B t_p}{\alpha_{\min}} + (D \times 1000)^2}$$

$$= 34,200 \text{ m/sec}$$

The payload ratio, for optimum C , can be shown to be:

$$\mu_L = 1 - \frac{a_o \alpha_{\min} C_{opt}}{B} \quad (4)$$

where a_0 is the initial thrust/mass ratio.

Since no payload is to be carried in addition to the waste-RTG package $\mu_L = 0$ and,

$$a_0 = \frac{B}{\alpha_{\min} C_{\text{opt}}} = 1.84 \times 10^{-4} \text{ m/sec}^2 \quad (5)$$

Therefore, the propellant fraction, μ_p is

$$\mu_p = \frac{a_0 t_p}{C_{\text{opt}}} = 0.388 \quad (6)$$

Thus the ΔV capability of the system is

$$\Delta V = -C_{\text{opt}} \ln (1 - \mu_p) = 16,800 \text{ m/sec} \quad (7)$$

Using the same criterion noted before, (see equation 1) it follows that the package will spiral out into the asteroid belt before it runs out of propellant. (This includes Earth escape which requires an additional ΔV of about 8 km/sec.)

The above example illustrates that some additional velocity may be required at Earth departure in order to escape the sun's gravity field. To investigate this possibility, some computer calculations were made to determine the value of a_0 required to reach solar escape starting from various values of velocity relative to Earth (supplied by some chemical rocket stage such as the Centaur). These are shown in figure 6. This has been done with a limit on t_p of 20,000 hr. and a fixed value of $C = 30,000 \text{ m/sec}$ (this is the lowest practical value based on current thruster technology work. Lower values of C develop difficulties in accelerator grid spacing required).

Given the data shown in figure 6, it is then possible to determine what values of a_0 and $V_{\infty,1}$ are required to escape the solar system for

any chosen value of $\alpha_{\min}$. These are determined from the following equation:

$$\mu_L = 0 = 1 - \mu_p - \frac{a_0 C}{2 n_{th}} \alpha_{\min} \quad (8)$$

where μ_p and n_{th} can be determined from previous relations (equations (3) and (6)).

Assuming the use of the shuttle/centaur, we have the following relation between $V_{\infty,1}$ and m_0 (at Earth escape):

TABLE 3. SHUTTLE/CENTAUR EARTH DEPARTURE MASS CAPABILITY

$V_{\infty,1}$ (km/sec)	m_0 (kg)
0	10,400
3	8,500
6	4,900
9	2,050

For this launch system and using an $\alpha_{\min}$ of 134 as before, it is found that $V_{\infty,1} \approx 3.0$ gives $\mu_L = 0$ and leads to an ejected system mass (final mass) of 4,850 kg (10,700 lb). This is considerably better than the direct solar escape payload of 1,230 kg. given in reference 1 for the shuttle/tug (expended). Continuing this process for other values of α other than $\alpha_{\min}$ leads to the results shown in figure 7 where the mass sent to solar escape is shown as a function of the propulsion system specific mass. As indicated in this figure, all cases above $\alpha \approx 60$ kg/kWe will require some assistance (from a Centaur, tug or some other chemical rocket stage) during Earth escape. These results indicate that the best case of $\alpha = \alpha_{\min}$ is interesting and that more work should probably be done to better define α .

Solar-Electric Propulsion

Another set of calculations has been made for the case of a 20 kw solar electric propulsion (SEP) system as the ejection stage. These were performed for the same shuttle/centaur departure mode but used an α of 30 kg/kw (typical for SEP stages) and a typical solar cell profile of power as a function of distance from the sun. In this case, it was found that $V_{\infty,1} = 6.0$ was required (with $I_{sp} = 3000$ sec. as before) which gave an injected mass (excluding the SEP system) of 1320 kg at solar escape. As indicated in figure 7, this is essentially the same as the shuttle/tug system. Thus it would appear that the SEP approach would not have any great advantage over a simple expended tug. Also, the combined cost of both the SEP stage and the Centaur would probably exceed or equal that of a single tug.

Waste Thermal Thruster

Another device examined was one in which the heat in the waste is transferred into some fluid which is ejected to produce thrust. A preliminary set of calculations for such a device was made assuming that liquid hydrogen could be heated to a maximum temperature of $\approx 2000^\circ\text{F}$. (This is near the maximum centerline temperature of most solid waste forms given previously.) Assuming complete expansion into a vacuum, an exhaust velocity of about 7 km/sec ($I_{sp} \approx 700$ sec.) is theoretically possible. However, the amount of waste material (or any isotope) needed to produce a sizable thrust is critical. For example, to achieve a thrust/mass of .10 requires:

$$\frac{P}{\dot{m}_0} \approx \frac{F}{\dot{m}_0} \frac{C}{2} = \frac{.10 \times 7000}{2} = 350 \frac{\text{watts}}{\text{kg}}$$

From figure 1, it is clear that this is about as much heat output as can be expected from any radioisotope heat source.

In order to escape the solar system with an initial acceleration of .10 m/sec², more ΔV than the 8.5 km/sec required with very high values of a_0

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must be supplied. This is due to the energy expended in lifting the unused propellant through a gravity field. Such "gravity loss" factors can be found in such documents as reference 9. Specifically, it is found from reference 9 that ΔV must be increased by 1.75 to overcome the "gravity losses" associated with an initial acceleration of 0.10 m/sec^2 .

At this point it is appropriate to try and size a stage which will escape the solar system. Assuming that the liquid H_2 can be contained in tank with a mass of about 10 percent of the contained propellant, it can be shown that the mass ratio for the maneuver (excluding tanks) is:

$$\frac{m_f}{m_o} = (1 + \sigma)e^{-\Delta V/C} - \sigma = .0314 \quad (9)$$

where σ = tankage factor = 0.10.

If the transfer began in low Earth orbit with $m_o = 29600 \text{ kg}$, then the propulsion system could be no more massive than

$$m_{\text{eng}} = m_o \left(\frac{m_f}{m_o} \right) = 0.0314 \times 29600 = 927 \text{ kg}$$

Therefore, the thrust, F , can be no more than:

$$F = \frac{2P}{C} = \frac{2 \times 927 \times 300}{7000} = 79 \text{ N}$$

Thus,

$$a_o = \frac{79}{29600} = 2.68 \times 10^{-3} \text{ m/sec}^2$$

At this low a value of a_o , it would require a ΔV of about 8 km/sec just to escape the Earth (see equation 1). This indicates that the system can't escape from low Earth orbit without some high thrust assistance.

For the case of high thrust assist, new data similar to figure 6 have been generated. Using the data and table 3, it has been found that $V_{\infty,1} = 6$ gives the highest mass of ejected waste isotope. This mass is shown in figure 8 where the values for shuttle/tug and SEP (20 kWe) are also shown for comparison.

In figure 8 it can be seen that α has no effect on the injected mass over the range shown. This results from the higher values of a_0 (to the right in figure 8) that result when the Centaur is used for Earth escape. Thus, α has very little effect on ΔV (through changes in a_0) until very high values of α are assumed.

It is therefore concluded from figure 8 that there is insufficient heat energy in the waste (or some of its components such as ^{90}Sr) to be of much interest as a thermal thrust producing device. However, all of this has assumed only one shuttle launch. If more launches are used, the system may be made to compete with a single shuttle/tug. However, considering the optimistic assumptions about the propulsion system size, it would not seem likely that the device could have much economic advantage.

Combined Systems

It may also be possible that some combination of the systems so far discussed could be a better choice. This is perhaps most true of an SEP waste-RTG system. As the power from the solar cells drop off, the RTG power would remain to give a more uniform power distribution throughout the flight. However, the same solar cell cost argument stated before still applies here. Thus, this would appear to be another system worthy of further investigation along with the pure waste-RTG devices.

Other Heat Sources

As noted earlier, the possibility of using some extracted compound of ^{137}Cs or ^{90}Sr should also be considered. From tables 1 and 2 it is

clear that ^{90}Sr would very likely be a much better heat source than the waste or ^{137}Cs . The 450 watts/kg output plus the long half life would combine to give a value of $\alpha_{\min} = 44 \text{ kg/kw}$. This clearly gives superior performance (shown in figure 7) to the shuttle/tug combination. However, the cost of separating the ^{90}Sr compounds from the waste must be included. Also, it must be recognized that the compounds may be mixed and of a type giving lower energy output than the SrO value given in the table. Most important, however, are containment, shielding, and other safety considerations which will probably increase α much above $\alpha_{\min}$. This is a detailed design problem which is beyond such a preliminary survey. Our purpose here is to try to narrow the alternatives for more intensive future study.

Summary

This investigation, although very preliminary in nature, has indicated that there could be some useful ways in which the heat in the nuclear waste can be used to augment space disposal of the waste. In particular, it appears that an RTG system operating on waste or ^{90}Sr compounds separated from the original waste could be used together with an electric propulsion system to reach solar system escape. In each case the results are very attractive in the extreme case of no-containment weight estimates. This does not mean the scheme will be ultimately useful, but does indicate that further consideration could be worthwhile.

Other approaches, such as SEP only and a nuclear-thermal thruster using the waste heat and liquid hydrogen, do not appear attractive, even with the aid of optimistic assumptions.

Although this study stresses the solar systems escape destination, several exploratory numerical calculations have been made for the high Earth orbit and Earth escape destinations. These show that some care must be exercised in selecting an Earth escape path in order to avoid future near encounters with the Earth and Venus. In general, it is believed that useful calculations are possible using numerical integration which could help in an orbit selection process.

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SYMBOL TABLE

a	acceleration, m/sec ²
B	constant in thruster efficiency expression
C	exhaust velocity, km/sec
D	constant in thruster efficiency expression, km/sec
F	thrust, N
I_{sp}	specific impulse, kg-sec/kg
m	mass kg
P	power, watts
t	time, sec
V	velocity, km/sec
α	propulsion system specific mass, kw/kg
ΔV	velocity increment, km/sec
η	efficiency
μ	mass ratio
σ	tank mass/propellant mass

Subscripts

c	converter
eng	engine
f	final
L	payload
min	minimum
opt	optimum
p	propellant or propulsion
th	thruster

- o initial ($t = 0$)
- 1 Earth departure
- ∞ indicating conditions on the asymptote of a hyperbolic orbit
- $\odot$ Sun
- $\oplus$ Earth

Figure 1. - Fission product thermal power as a function of time after reprocessing of spent fuel elements. (From NASA TMX-68147)

Figure 2. - Heat production in high-level wastes from spent fuel processing. (After five years, Strontium and Cesium account for most of the heat production. Note that removal of these isotopes before several years of aging would have little effect on heat production of the remaining mixture.)

Figure 3. - Isotope waste heat propulsion concepts.

Figure 4. - Nuclear waste disposal space trajectories.

Figure 5. - Propulsive velocity increment and flight time for solar impact missions; $H_p = 556$ km.

Figure 6. - Initial acceleration required for solar system escape mission; $t_p = 20,000$ hr; $I_{sp} = 3000$.

$$\eta_c = 0.05$$

$$\eta_{th} = \frac{0.842}{1 + (16/C)^2}$$

$$\alpha(\text{SEP}) = 30 \text{ Kg/KW}_e$$

Figure 7. - Mass delivered to solar system escape by various propulsion system concepts; shuttle payload = 65,000 lb. at 100 n.mi.; $t_p = 20,000$ hr.

Figure 8. - Mass delivered to solar system escape; comparison of some alternative systems; $I_{sp} = 700$ (isotope-thermal); $\alpha(\text{SEP}) = 30 \text{ kg/kw}$.

APPENDIX 8.D

Feasibility of Using an Orbiting Accelerator
to Eject Radioactive Waste Products into Space

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GULF RADIATION TECHNOLOGY

Gulf-RT-C12457

FEASIBILITY OF USING AN ORBITING ACCELERATOR TO EJECT RADIOACTIVE WASTE PRODUCTS INTO SPACE

Interim Summary Report

by

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Prepared for

**Battelle Pacific Northwest Laboratories
Richland, Washington**

under

**Consultant Agreement BCA-741
Gulf Rad Tech Project 0512**

December 27, 1972

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FOREWORD

This report summarizes progress under Consulting Agreement BCA-741. Approximately \$4000 of the original \$10,000 allocation has been expended to date and, as agreed in the letter to Dr. A. Platt, May 30, 1972, it is at this stage that the program is to be evaluated.

1. INTRODUCTION

The purpose of this consulting agreement is to evaluate the use of an accelerator to dispose of particulate radioactive waste materials from earth orbit. The method consists of placing into orbit a sealed vessel containing the waste products, using the space shuttle as the launch vehicle. The waste would be contained in a solid matrix for obvious safety reasons. Once in orbit, the waste would then be converted to the form of small charged particles. These particles would then be electrostatically accelerated to the escape velocity.

The use of relatively large particles as opposed to atomic species is to prevent trapping of the radioactive waste material in the earth's magnetic field.

2. ENERGY CONSIDERATIONS

The main advantage in the use of an acceleration scheme, as opposed to firing the waste out of the solar system with a rocket engine, is the saving in energy costs. The solar escape velocity from earth orbit is 42 km/sec and orbital velocity between 100 and 300 km altitude is about 8 km/sec.

Thus, escaping the solar system requires an additional impulse of about five times orbital velocity, or 25 times the energy. Also, in using the accelerator method, one has only to accelerate the waste, not the container as well. In the discussions to follow, it is shown that the accelerator energy requirements are relatively modest in contrast to that expected using conventional rocket technology to achieve the required

velocity increment. Consequently, the main advantage of the accelerator approach compared to the use of chemical rockets is that removal of the radioactive waste from the solar system becomes economically (i.e., energetically) feasible.

3. EJECTION SCHEMES

It would appear appropriate at this stage to discuss in some detail all the possible ejection schemes utilizing an accelerator. The apparent advantages and disadvantages of each will be discussed.

3.1 EJECTION FROM THE SURFACE OF THE EARTH

For obvious reasons involving atmospheric drag, particle ablation, and charge transfer, it is not possible to use an accelerator to eject waste particles from the earth's surface. This suggests that the first step be to put the waste product plus container into orbit. Use of the space shuttle has been proposed.

3.2 EJECTION FROM EARTH ORBIT

3.2.1 Ejection into the Sun

In this ejection scheme, the particles are given enough velocity to leave earth orbit and fall into the sun. For an object initially at rest on the earth's surface, the velocity required is about 30 km/sec. Taking advantage of the velocity in earth orbit, this would be reduced to about 22 km/sec. There are several reasons, however, why this method is not desirable.

- A. The requirements on velocity and attitude control are complex. This is illustrated by the use of Figure 1. In this figure, the accelerator is considered to be in an orbit coplanar with the ecliptic and the earth is arbitrarily chosen to be moving in a counterclockwise direction about the sun while the satellite's motion is clockwise about the earth. It is apparent that any attempt to eject material into the sun from a position in orbit to the left of plane A-C will involve having to eject the material towards the earth, an unsafe practice. Thus one is restricted to working only in one part of the orbit. Furthermore, even in the acceptable portion of the orbit, the ejection velocity is dependent on the position and on the ejection direction thus requiring a programmed control on the particle velocity. As an example, for the specific case where the ejection direction is opposite to that of the earth's velocity,

Figure 1. Diagram of earth sun system with orbiting earth satellite

the ejection velocity required to "hit" the sun at different selected positions in orbit are:

Point A	22 km/sec
Point B	30 km/sec
Point C	38 km/sec

The orbital velocity is taken advantage of at only one point.

To design a system which would eject material only at one point in order to save energy costs would, of course, be impractical from the point of view of material processing. Also, a problem to consider in such a scheme for particle ejection is the added attitude-control problem which would result from the fact that the ejection of particles would cause minute, but still an accumulative, change in the orbital velocity of the accelerator. One would have to compensate for such an effect by using a small thruster.

- B. A velocity spread in ejected particles will result from the fact that it will not be possible to charge all of the particles identically. This will result in some of the particles going into orbit about the sun rather than striking it.
- C. The particles which approach the sun will be evaporated before striking it and may eventually find themselves back near the earth's orbit due to the solar wind. In theory, this should not be a problem since the dilution factor is great. However, one must consider here the fact that often we are dealing with an incredulous public when it comes to ecological matters.

3.2.2 Ejection from the Solar System

The speed required to escape the solar system from earth's position is approximately 42 km/sec. Two methods of escape are possible. They are:

- A. Ejection into the ecliptic plane. Referring to Figure 1 once more, this involves ejecting the particles from a position in earth orbit into the ecliptic plane. Since the expenditure of energy is important to us, we would not want to eject particles such that they traveled

within the earth's orbit about the sun. Thus, we would only want to eject particles in a direction parallel to the earth's velocity vector. Again, the velocity required is dependent on the position in orbit.

Assuming, as before, an ejection direction opposite to the earth's velocity, the particle velocities required at different points in earth orbit are determined as follows.

Point A

Ignoring the presence of the earth, the escape speed from the sun from earth position is 42 km/sec, but at point A the velocity of the earth is opposite to that of the motion of the satellite, thus, the velocity required to escape is 72 km/sec. In addition, if the local gravitational field of the earth, corresponding to a parabolic speed of 11.2 km/sec, is considered, then the velocity required at the earth's surface to reach an infinite distance from the solar system is $\left[(72)^2 + (11.2)^2\right]^{1/2} = 72.9$ km/sec. Furthermore, the 8 km/sec satellite velocity is subtracted to yield 64.9 km/sec.

Point C

In this case, the earth's velocity is assisting and the velocity required to escape, ignoring the presence of the earth, is 12 km/sec. The required velocity is then calculated in the following manner.

$$\begin{aligned}\Delta V &= \left[(12)^2 + (11.2)^2\right]^{1/2} - 8 \\ &= 8.4 \text{ km/sec} .\end{aligned}$$

Points B and D

Here, the ejection direction is perpendicular to the earth's velocity, and the required velocity is given by

$$\begin{aligned} & \left[(42)^2 - (30)^2 \right]^{1/2} \\ & = 29.4 \text{ km/sec.} \end{aligned}$$

Once again, if earth escape and the satellite velocity are taken into account, the required velocity is

$$\begin{aligned} & \left[(29.4)^2 + (11.2)^2 \right]^{1/2} - 8 \\ & = 23.5 \text{ km/sec.} \end{aligned}$$

The apparent advantages and disadvantages of the above ejection method are:

1. As with ejection into the sun, velocity control becomes an important consideration.
2. For cost reasons, it would be desirable to be able to continuously process the waste material. Unlike ejection into the sun, this scheme allows us to do that provided we are willing to pay the penalty of reaching maximum velocities 64.9 km/sec.
3. In this ejection scheme, a velocity distribution effect can be taken care of by merely assuring that the lowest charged particles get ejected at greater than the required velocity. All others will then be sure to escape. This was not so for the case of ejection into the sun. In that case, the particles would go into orbit about the sun provided solar escape speed had not been reached.
4. Similar to the scheme involving ejection into the sun, there is a problem due to drag effects and a small thruster is required.
5. One should also consider, despite the astronomical dilution factors involved, the public's reaction to the "contamination" of their solar system (planets, asteroids, etc.).

- B. Ejection out of the ecliptic plane. In this method, the particles are ejected from earth orbit into a direction out of the ecliptic plane. The most consuming configuration energy-wise would be to have the particles ejected normal to this plane. This would involve ejecting the particles in a direction counteracting the earth's velocity with a speed of 57 km/sec or 45 km/sec depending on the direction of the orbital velocity. The angles to the ecliptic plane corresponding to these velocities are 48° and 61° , respectively. Thus, the maximum velocity required to eject particles normal to the ecliptic is 57 km/sec.

The above configuration represents a relatively large expenditure of energy and represents a limiting case. The contamination level of the solar system could equally be made low by ejecting the particles at approximately 45° to the ecliptic. As an example, ejecting the particles normal to the ecliptic plane at a velocity of 42 km/sec would find them traveling at escape speed at an angle of 48° or 61° to the ecliptic plane depending on the direction of the satellite velocity. This configuration would represent a reasonable expenditure of energy for the escape of particles from the solar system without any significant contamination of the solar system itself. The calculations to follow will be based on the use of such a configuration.

There appear to be several advantages to the out of plane method.

1. The required velocity can be maintained at a constant value; i.e., one only has to be sure that the lowest charged particle receives a velocity greater than 42 km/sec in the configuration discussed above.

2. The attitude of the vehicle is always the same. This simplifies attitude-control problems.
3. For an orbit which is in the ecliptic such as drawn in Figure 1, the accelerator can eject particles in either the "up" or "down" direction. This immediately suggests the design of an accelerator with two opposing ejection ports to compensate for momentum differences.
4. The operation of the device can be continuous.
5. The impact on the public would probably be much less than for any of the other schemes.

3.3 EJECTION EITHER INTO THE SUN OR OUT OF THE SOLAR SYSTEM FROM THE MOON

Such a scheme would have all of the disadvantages described heretofore plus others:

- A. There would be an added expenditure of fuel (i.e., energy) which we have indicated becomes significant if the entire waste can has to be accelerated.
- B. The desire to keep the waste away from the earth would involve the use of more sophisticated attitude-control systems and built-in safety devices.
- C. The entire system would be complicated by the remoteness and the increased complexity of the telemetry involved.
- D. In the solar escape mode of operation, ejection out of the ecliptic would be complicated by the fact that the moon's orbit is significantly inclined to the ecliptic plane and variable with the seasons.

4. DILUTION CONSIDERATIONS

When dealing with a public that is fast developing an acute awareness of ecology, the extraterrestrial ejection of radioactive waste is sure to be a sensitive matter. The following calculation indicates the order of magnitude of the contamination levels expected near heavenly bodies. The assumptions are:

- A. The particles are ejected from the solar system out of the ecliptic plane.
- B. The accelerator orbit is coplanar with the ecliptic plane.
- C. The combined satellite-orbital motion and earth-orbital motion define a complex ejection source which can be considered a symmetric source at far distances.
- D. The first heavenly bodies (stars) are encountered at 4 light years (9.5×10^{17} cm) distance.
- E. The ejected beam is monoenergetic (42 km/sec) and has an angular divergence of 5° (0.087 radian) leaving the accelerator.
- F. The process rate is 100 tons per year (3 g/sec) from 100 power plants of 1000-MW capacity each.

The calculation is performed as follows. The area, A, swept out by the accelerator beam at a point 4 light years distant is

$$A = 2\pi d^2 (0.087) = 5 \times 10^{34} \text{ cm}^2.$$

The mass flow, m, is 3 g/sec. Thus, at steady state, the flux at 4 light years distance is 6×10^{-35} g/cm²-sec.

The speed of the particles at 4 light years distance from the sun will be the escape speed from that point, i.e., the speed at that point required to get the particles to infinity. The formulation is

$$V_{\text{escape}} = \left[\frac{2\gamma M_{\text{sun}}}{d} \right]^{1/2},$$

where γ is the gravitation constant, M_{sun} is the mass of the sun, and d is the distance from the sun.

Substituting the appropriate values for these quantities yields a speed of 1.7×10^4 cm/sec.

Therefore, the density at the 4-light-year distance is 1.2×10^{-31} g/cm³.

Furthermore, if we assume a radioactivity per unit mass of spent fuel at 9 Ci/cm³,⁽¹⁾ this would mean that a star of the same area as our sun at the 4-light-year distance would intercept about 1 μ Ci per day over its

Figure 2. Plot of acceleration voltage versus particle radius. Iron,
 $E = 2 \times 10^{10} \text{ V/m}$

entire surface. In actual fact, however, the material will have undergone decay before reaching this position in space. This can be seen by calculating the transit time.

For simplicity, we will assume an average velocity out to the 4-light-year point of 8.7×10^5 cm/sec. At such a speed it will take about 10^5 years. Most of the radioactive waste has a half-life much shorter than this value. It is expected, therefore, that the radioactivity level will be many orders of magnitude less.

5. ACCELERATOR DESIGN REQUIREMENTS

5.1 CHARGED PARTICLE CONSIDERATIONS

Assuming a spherical particle, the velocity-voltage relationship is given by

$$v = \left(6V E \epsilon_0 / r \rho \right)^{1/2}, \quad (1)$$

where v is the particle velocity (m/sec), V is the applied voltage. E is the electrostatic field strength at the surface of the sphere. ϵ is the permittivity of free space (8.85×10^{-12} F/M). r is the particle radius (m), and ρ is the particle density (kg/m).

Equation 1 indicates that for a spherical particle of given radius and material, and for a fixed accelerating potential V , the velocity is dependent on the square root of the field strength of the particle surface. Thus, it behooves one to try to obtain the highest possible practical field strengths. Theoretically, the maximum field strength at the surface of a negatively charged particle is determined by the electron field emission limit, which is about 10^9 volts/meter. If the same particle is charged positively, however, the maximum surface field intensity is determined by the forces binding the atoms to their lattice size. For an iron particle, for example, the maximum positive surface field strength is about 2×10^{10} volts/meter.⁽²⁾ Assuming an ion particle with this maximum surface field strength being accelerated to escape speed of 42 km/sec we can calculate the acceleration voltage required for particles of different sizes. Figure 2 is such a plot. Thus theoretically,

a 1- μ m-diameter iron particle would have to be accelerated through 6.5×10^6 volts to reach the solar escape speed. The maximum charge on the particle is given by

$$q = 4\pi r^2 \epsilon_0 E \text{ coulombs,} \quad (2)$$

and for a 1- μ m-diameter iron particle, the value of q is 5.6×10^{-13} coulombs, or 3.5×10^6 electronic charges.

The above calculations have been performed employing theoretically ideal values. The practicality of achieving these values in an accelerator will now be considered. One thing is clear: One should charge positively to take advantage of the higher field strengths attainable.

In an experiment involving the simulation of meteoric impact, Fruchtenicht⁽³⁾ was able to achieve E values of 2.5×10^9 volts/meter for iron particles by having the particles simply make contact with a highly charged surface. Furthermore, 1- μ m-diameter iron spheres possessing this E value were accelerated to velocities approaching 8×10^5 cm/sec using an acceleration voltage of 1.75×10^6 volts. This is the velocity predicted using Equation 1. Thus, Equation 1 can be used with some confidence to predict the acceleration voltage required for the waste fuel ejection problem. We make the following assumptions.

- A. The waste matrix material is assumed to be spray melt⁽¹⁾ with an average density of 3 g/cm³.
- B. The value of 2.5×10^9 volts/meter, as experimentally verified by Fruchtenicht for iron, is assumed to be identical for spray melt.
- C. The method chosen for ejection of the particles is solar escape from earth orbit, thus requiring particle velocities equivalent to 42 km/sec.

Employing the above quantities, one obtains the plot shown in Figure 3. Also included is the theoretical limit for the same material, assuming the possibility of a maximum theoretical E value equivalent to that for iron, i.e., 2×10^{10} volts/meter. To obtain more accurate values, one will have to determine the experimental E value for spray melt. In any event, the feasibility of the method is indicated.

Figure 3. Plot of acceleration voltage versus particle radius, spray melt, $E = 2 \times 10^{10} \text{ V/m}$ and $2.5 \times 10^9 \text{ V/m}$

It is obvious that the lowest possible acceleration voltage is desirable from the standpoint of expense. It behooves one, therefore, to seek:

- A. Methods of increasing the value of the electrostatic field E at the surface of the particle.
- B. Methods for producing the smallest-sized particles possible.
- C. A matrix material which is of the lowest possible density commensurate with all of the other requirements.⁽¹⁾

Some of the methods which may be used to increase the surface field strength, E , are:

- A. Assuming that the waste product matrix material has too low a surface field strength by itself, it may be possible to coat the particles with a material which has an acceptable value of E .
- B. Significant increases in the charge-to-mass ratio of liquid particles has been observed when small solid particles are suspended in the liquid.⁽⁴⁾ Indeed, the charge-to-mass ratio of a 20% suspension of carbon in glycerin increases the charge-to-mass ratio over that for pure glycerine by a factor of 10. This immediately suggests that we consider employing a similar technique in the waste fuel application. First, a liquid has many advantages from the viewpoint of handling and making particles in orbit. Second, the fuel matrix seeded with the radioactive waste, once made molten in orbit, may already possess a high enough charge-to-mass ratio. Experimental studies are indicated in this area.
- C. The ion implantation of positive charges into the particles has also been suggested.⁽⁵⁾ Mechanical handling would seem to be more difficult than either of the above methods; nevertheless, the method should be given some thought.

6. SMALL PARTICLE PRODUCTION

Many methods are used to produce small particles ($<100 \mu\text{m}$), and a classification is difficult. A brief description of each of the methods, appropriate to both solid and liquid phases, is presented below.

6.1 MECHANICAL CRUSHING AND GRINDING METHODS

Under this heading are included impact machines, roller mills, and attrition mills.

6.1.1 Impact Machines

As the name implies, the primary force behind size reduction in these units is impact. It may be impact created between the particles themselves, but more often it is impact between the particles and adjacent parts of the mill. Other comminution forces such as attrition, compression, and shear may also contribute, but to a minor extent.

Depending on the choice of machine, capacities can range from 25 to 3×10^5 lbs/hr and power requirements from 1/2 to 1000 hp. The resulting particle diameters can be as small as 5 μ m at 95 to 99% yields.

6.1.2 Roller Mills

The primary crushing or grinding action in roller mills is a compression mechanism. Impact forces rarely come into play, but attrition is important especially in the finer sizes. All roller mills operate by compressing the material between two surfaces, with at least one of them rotating.

Again, depending on the choice of machine, particle diameters as small as 40 μ m with 80 to 99% yields are possible. Capacities range from 400 to 6×10^5 lbs/hr and power needs from 1/2 to 600 hp.

6.1.3 Attrition Mills

Grinding by attrition is generally most applicable when the product has to be fine. The attrition can be either between the material and surfaces of the mill or between the particles themselves. Wear of mill parts is quite high where the attrition is between the mill and the material, so self-attrition is most desirable.

The capacity range of these units is between 3 and 1,000 lbs/hr, with power requirements between 5 and 200 hp, depending on the unit chosen. Particle diameters which result are in the 1- to 20- μ m range.

For more detail regarding the methods of size reduction discussed above, the reader is referred to Reference 6. In addition, new techniques

in the field of size reduction employing mechanical crushing and grinding are continually being developed.⁽⁷⁾

6.2 CRYSTALLIZATION METHOD

This process is used considerably in the chemical process industries, and normally involves three steps: Reactants are mixed (usually in a liquid system), a reaction occurs, and crystals form. These are then removed from the mother liquid, dried, and milled if necessary. Many variations are based on this method, examples of which can be found in Reference 8. Particles as small as 0.01 μm in diameter have been obtained.

6.3 SPRAY-DRYING METHOD

In this method, the material to be made into small particles is put into solution or a colloidal suspension and sprayed through a nozzle into a hot air stream. The dried particles are usually hollow spheres whose size depends on the droplet size from the atomizer and on the concentration of the hydrosol. Typical particle sizes are in the range of 20 μm and up, although Stern et al.⁽⁹⁾ describe a spray-drying method for generating aerosols of the order of 0.04 μm . The method has also been investigated in a fluidized calcination process for the conversion of liquid radioactive wastes from the reprocessing of nuclear fuels to solid form.⁽¹⁰⁾

6.4 SPRAY-FREEZING METHOD

Introducing small particles by spray-freezing involves spraying a solution or colloidal suspension of the material of interest onto a cold liquid film.⁽¹¹⁾ The droplets from the spray are frozen and suspended in the liquid. A slush-like mixture results. The liquid is then evaporated and the porous mass which remains is then vacuum-sublimed, and fine-particle powder is obtained. A high yield of particles less than 1 μm in size results.

6.5 GAS PHASE INTERACTION METHODS

6.5.1 Gas Phase Chemical Reactions

This method is usually employed when carbon blacks, as well as some pigments, are required. Such reactions include processes in which a

gaseous material decomposes into a lower-molecular-weight species that, by condensation, grows to a usable particle size. For example, carbon black results from the simultaneous condensation and dehydrogenation of hydrocarbon products in flames. Also titanium tetrachloride vapor, reacting with water vapor, is one source of titanium dioxide pigment. These materials can be collected on cold surfaces or in bag collectors after formation, with minimal treatment.

6.5.2 Nucleation and Condensation in Underexpanded Gas Flows

It has been known for some time that small clusters of atoms result from the rapid expansion of a vapor in a supersonic nozzle. Such a phenomenon has practical significance in the operation of turbojet⁽¹²⁻¹⁴⁾ and rocket^(15,16) engines and has prompted a great deal of research activity, both experimental and theoretical.⁽¹⁷⁻²³⁾

In the area of nozzle beam generation,⁽²⁴⁾ a technique used in many research experiments, condensation studies are particularly basic in nature. Beams containing small-mole fractions of argon, nitrogen, oxygen, and nitric oxide dimers have been obtained in this manner.⁽²⁵⁻²⁷⁾ Higher-order clusters (>30 atoms) have also been observed in argon.⁽²⁶⁾ Some theoretical considerations have been put forth by Hagena.⁽²⁸⁾

For our purposes, the yield of particles for the amount of vapor which has to be processed is low.

6.6 HIGH-ENERGY DISCHARGE METHODS

6.6.1 Arcs

Since in a high-intensity arc the major fraction of the arc input energy is dissipated at the anode, it is possible to incorporate material to be subdivided into the anode, either by itself or with a small amount of conductive material added. The material then issues as part of the anode tail-flame at temperatures approaching 7,000°K.⁽²⁹⁾ The resultant high rate of cooling causes condensation to occur, with the production of very fine particles. By varying the rate of cooling of the condensing vapors, particle size can be controlled. Particles from 0.005 to 0.2 μm have been produced by this technique. A number of variations employing

the basic concept are described in the literature. One such technique is discussed in Reference 30.

6.6.2 Plasma Torches

In many instances, the plasma torch is not used as a method for producing small particles; rather, it is used for the spheroidization of a material available in powder form.⁽³¹⁾ In this method of operation, materials, often refractory, which are close to the desired shape are put in a high-temperature plasma. As the particles melt, surface tension produces spheres of the same volume as the original particles. The molten droplets are then usually sprayed into water, where they harden. In other applications, spheres are made from wire⁽³²⁾ or metal rod.⁽³³⁾ Particles less than 20 μm in diameter can be made.

6.6.3 Exploding Wires

In this method, a wire of the metal required in powder form is exploded instantaneously by discharging high current through it.⁽³⁴⁾ Particles of the metal or its oxide, in the case of explosion in the atmosphere, are formed in the 0.02- μm -diameter range. The method is a useful one from the point of view of many research needs, but the noncontinuous nature of its operation makes it an unlikely choice for any kind of production application.

6.7 THERMAL DECOMPOSITION

Decomposition methods have been used to prepare metal oxide particles by spraying solution droplets of the metal salt into an oxidizing flame, which changes the salt into the oxide. Particle size is controlled by the size and salt concentration of the droplets and by the temperature and velocity of the flame. Studies on the thermal oxidation of plutonium have indicated formation of an oxide aerosol in the size range 0.1 to 10 μm .⁽³⁵⁾

6.8 NUCLEATION METHOD

In this method, the vapor from a suitable source is mixed with the vapor of the material for which it is desired to form the aerosol. Atoms of the former substance serve as nucleation centers and the aerosol results.

The mixture is then diluted with air to minimize coagulation. The LaMer aerosol generator employs this principle.^(36,37)

6.9 CONVENTIONAL LIQUID ATOMIZERS

Under this heading are included all methods in which tiny droplets result when a film or sheet of liquid encounters an appreciable velocity difference between itself and a new environment. The shearing forces produced cause the film to collapse into filaments and droplets that travel away from the sheet's main body.

The velocity difference can be produced by several means:

- A. Centrifugal spray nozzles. In this method, pressure energy is converted into a high-velocity swirling film, and a liquid mist results. Extensive studies have been conducted on this method.⁽³⁸⁾
- B. Rotating disc atomizers. There are a number of variations on this method.^(39,40) The principle is to emit the liquid film from the edge of a spinning disc or bowl and thus create the condition of a large velocity difference.

The droplets formed using the above methods are typically in the 100- μ m size range. In addition, energy utilization efficiency is rarely more than a few percent, and uniform droplet size cannot be attained without an auxiliary method for removing the over- and undersize. Theoretical papers are available on the general topic of drop formation from rapidly moving liquid sheets.⁽⁴¹⁾

6.10 TWO-FLUID OR PNEUMATIC NOZZLE ATOMIZERS

In this type of device, a slow-moving liquid stream is injected into a gas stream that is moving at near-sonic velocity. Examples of studies conducted using this method are found in References 42 and 43. The two-fluid method can produce droplets typically in the 100- μ m size range, but the power consumed per unit mass of material atomized is quite high.

6.11 ULTRASONIC ATOMIZER

A high-frequency (about 20 kHz) transducer provides ultrasonic vibrations that are focused at the surface of the liquid from below. A liquid spout (called an ultrasonic fountain) then arises, and a fog is emitted

from its base. Droplets are formed with diameters under 5 μm . More details can be found in Reference 44.

6.12 UNSTABLE LIQUID JET

Under the action of surface tension forces, a free column of liquid will spontaneously disintegrate into droplets. All the droplets will be of about the same size, with a diameter of about twice the jet diameter. It is assumed that the surrounding gas in such a case does not influence the disintegration. The droplets formed are not exactly of the same size and have not exactly the same speed; thus, they collide and coalesce. This coalescence can be considerably delayed by making the size, spacing, and speed of the droplets more uniform through velocity modulation of the jet.

The theoretical basis for the method is the work of Raleigh.⁽⁴⁵⁾ According to his analysis, a cylindrical jet of inviscid liquid is unstable with respect to an axially symmetric perturbation which has a wavelength λ greater than π times the unperturbed jet diameter. Indeed, the optimum wavelength to diameter ratio (λ/D) for droplet formation is 4.5. In such a case, one droplet per wavelength results. The size and frequency with which the droplets are produced depend upon the flow rate of the liquid through the capillary, the capillary diameter, its resonant frequency, and the amplitude of the oscillation of the capillary tip. The method is most suitable for producing beams of droplets of near uniform size at a relatively high production rate.

Various schemes have been employed to obtain instabilities in a liquid jet. Typically, some means of electromagnetic disturbance^(46,47) have been employed as well as piezoelectric transducers.^(48,49) Strom⁽⁵⁰⁾ has used a membrane vibrated by means of electrostrictive elements. In addition, modulation via the pressure of the liquid inside the capillary has been employed by a number of workers.^(51,52) A variation on this method is presently being used at Gulf Energy & Environmental Systems Company (GEES) to fabricate fuel particles for use in the high-temperature gas-cooled reactors.

6.13 ELECTROSTATIC ATOMIZER

Such a device produces droplets when an electric stress produced by the applied electric field exceeds the liquid's surface tension force. The electric stresses draw the liquid to a filament and disperse it into charged droplets.

By itself, a column of liquid issuing from a capillary will disintegrate into droplets under the action of surface tension forces. For a vertical capillary, for example, the droplet at the exit of the capillary will grow in size until its weight overcomes the restraining forces of surface tension along the edge of the capillary tip. At this point, the drop falls from the capillary, and the process repeats itself. When an electric potential is applied between the capillary and a ground plate centered beneath the capillary, the liquid becomes charged and strong downward forces on the droplet result. The effect of the applied voltage will be to reduce the size of the falling drops and to increase their frequency of formation. The size will continue to decrease with increasing voltage until the diameter of the falling drops is roughly twice the diameter of the capillary. At this point, the meniscus at the capillary tip will become electrolydynamically unstable, and harmonic electrical spraying of liquid drops from the meniscus will commence. The transition from the dripping mode to the spraying mode will be marked by a sharp decrease in the size of meniscus and the diameter of the emitted drops, and a sharp increase in the frequency of droplet emission. References 53 to 56 are descriptions of work based on the use of the principle.

Specific use of this method is made in the paint industry where paint droplets are electrically deposited on the object to be painted. More uniform coverage and less overspray results.

7. CHOICE OF A METHOD TO PRODUCE SMALL PARTICLES FOR WASTE EJECTION

The criteria for choice of a method for producing small particles of radioactive material for waste ejection are:

1. A method in which the waste material (radioactive waste and matrix material) is transferred as a solid mass into orbit, as opposed to

a method in which a powdered material is launched into orbit. The safety reasons are obvious.

2. A method in which the particles are formed in a continuous beam ready for acceleration purposes. This eliminates the mechanical problems associated with making the particles and then transferring them in powder form to the charging and acceleration stages.
3. A method capable of an ejection rate which is commensurate with the projected radioactive production rate of 100 tons/year from 100 power plants of 1000-MW capacity each. The dilution caused by the use of a matrix must also be considered.
4. A method in which nearly all the same size particles are produced. This would eliminate the need for reprocessing and the accelerator design would consequently be simpler.
5. A method in which the particles produced would be of the smallest size commensurate with steps 1, 2, 3, and 4. The smaller the particles, the lower the acceleration voltage required for ejection and, thus, the less demand on accelerator engineering.

In addition, the device used to produce the particles should be the least complex possible from the point of view of telemetry needs, the number of moving parts kept to a minimum because of lifetime problems, and the weight kept as low as possible to save on launch costs.

By a process of elimination, only two of the methods listed in Section 6 appear to meet the above requirements. They are the electrostatic atomizer and unstable liquid jet methods. Our system concept is based on the use of either of these methods or a combination of both.

8. SYSTEM CONCEPT

The envisioned system consists of two main sections, the accelerator section and the waste container-particle generation section.

The accelerator section would be placed in orbit and would not be recoverable. Its main function would be to supply the acceleration potential required to eject the material. In addition to the necessary electrodes and collimators, it would have a means of generating the high voltages. The emphasis should be on simplicity and reliability.

The waste container, besides holding and shielding the radioactive waste, would also possess the required equipment for generating the particles. In this manner, the most complex, and thus less reliable, portion of the entire system could be checked out on the ground before launch and, in the event of failure in orbit, it could still be recovered. Consequently, there would be no need to engineer a system possessing extremely high reliability and long lifetime, and the cost of the system would be reduced considerably. This portion of the system would dock with the orbiting accelerator section.

8.1 WASTE PROCESSING

If the electrostatic atomization and unstable liquid jet methods are to be used, the waste must be in the liquid state; yet for safety reasons, the waste material must be solid during the launch. Consequently, some means of melting the material and containing it once in orbit has to be devised. One possibility would be to take advantage of the internal heat generated by the radioactive waste itself. The waste container in such a case could possibly be made of small enough diameter so that the ratio of surface area to volume was small enough to allow the entire mass within the vessel to melt. External cooling fins could then be attached to increase the radiative heat transfer and consequently keep the mass solid on the ground and during launch. Once in orbit and linked up with the accelerator section, the fins could be retracted by some mechanical means and the mass permitted to melt. The use of a solar concentrator to supply the necessary heating should also be considered. Such a unit should be integral with the accelerator and remain in orbit. In addition, it may be possible to use part of the power from the system on board the accelerator section to supply the required heat energy.

8.2 ACCELERATOR

The accelerator design itself has yet to be looked at in detail. From the discussions in Section 4, it would appear that for a 1- μ m-diameter particle of density 3 g/cc, it might be necessary to generate an acceleration potential as high as 20 MeV. Achieving these potential levels is not a limitation in itself. The limitation appears rather to come from the practical problems associated with size and weight. From this point of view,

it behooves one to explore all the possible ways of reducing the required voltage level. This can be done, effectively, by reducing the particle size and/or increasing the charge-to-mass ratio.

One of the most important parameters determining particle size in either of the above methods is capillary size. It is possible to purchase, nowadays, collimated hole structure⁽⁵⁷⁾ fabricated from stainless steel which possesses line-of-sight capillaries down to 1- μ m diameter. In addition, porous plugs of sintered metals of extremely small pore size are available.

Various methods for increasing the charge-to-mass ratio of particles have already been discussed in Section 6. One should compare the cost associated with using these suggested methods to that associated with simply building the accelerator larger.

8.3 PARTICLE CHARGING

The most convenient method of particle charging would be that of employing electric field charging as in the electrostatic atomizer (Section 7). The fact that the high field strengths are already available for acceleration purposes makes this a desirable charging method.

In the event that the above charging method cannot be employed, the particles could be charged by electrode contact or by a crossed ion or electron beam before entering the acceleration region. The additional equipment required, however, is relatively complex and would possess a short operating lifetime. For these reasons, it would be best made a part of the recoverable waste container package.

8.4 POWER CONSIDERATIONS

The production rate of radioactive waste has been estimated at 3 g/sec, and if we assume the ratio of matrix material to actual radioactive waste material to be 1:1, the waste ejection rate from the accelerator is 6 g/sec.

Further, the mass, m , per particle assuming spheres is

$$m = \frac{4}{3} \pi r^2 \rho$$

Thus, for $\rho = 3$ g/cc and $r = 0.5 \mu\text{m}$, i.e., 5×10^{-5} cm

$$m = 1.6 \times 10^{-12} \text{ g}$$

Thus, a waste ejection rate of 6 g/sec is equivalent to 3.8×10^{12} particles/sec.

The charge on each particle, q , is given by

$$q = 4 \pi r^2 \epsilon_0 E$$

where, if we take $E = 2.5 \times 10^9$ V/m, we obtain

$$q = 6.9 \times 10^{-14} \text{ coulombs .}$$

This is a current of 0.25 amperes.

Thus, the power required to maintain a potential difference of 20 million volts with this charged particle ejection rate is 5 MW.

In addition, it will be necessary to eject charge of the opposite sign to maintain charge neutrality of the satellite. This does not involve the use of voltages greater than a few kilovolts, however, so the power needed for this function is in the kilowatt range. In such a case, however, the electrons would best be expected to be trapped in the magnetic field of the earth, since otherwise they will attempt to return to the satellite and not alleviate the electrostatic charging problem.

8.5 PARTICLE SOURCE CONSIDERATIONS

In Section 8.4, the particle ejection rate was found to be 3.8×10^{12} particles/sec, assuming 1- μ m particles of density 3 g/cc. In addition, typical drop frequencies have been found to be, for a good yield of particles of the correct size, about 1500 particles/sec in the GEES drop-forming device for fuel particle production. Thus, from these data it would appear that some 2.5 billion orifices would be required. Collimated hole structures are typically sold off-the-shelf with 750,000 holes of 1 μ m diameter within an area of 0.0014 ft².⁽⁵⁷⁾ Using these values, therefore, the capillary area which would be required to handle the above waste ejection rate would be some 5 ft², which is a circular cross section of 2.5-ft diameter. This area may have to be increased to provide more space between the capillaries, but even so, it is not unreasonable to envision the use of collimated hole structures to obtain the required number of orifices. In addition, it is likely that higher droplet formation rates can be obtained through research and development efforts.

Concerning the handling of molten oxides, this does not appear to be a particular problem. Small spherical particles containing the oxides and carbides of zirconium, thorium, and uranium are typically made in the GEES apparatus. In addition, Reference 51 describes a glass-drop-forming device. Collimated hole structures are presently manufactured from stainless steel but the potential fabrication of structures from refractory metals is indicated. (57)

9. CONCLUSIONS

This precursory study suggests the feasibility of using an electrostatic particle accelerator for the ejection of radioactive waste material from earth orbit into outer space. The major findings of this study are summarized as follows.

The main advantage appears to be in the saving in energy, and consequently cost, which results compared to using chemical rockets. Using a projected waste production rate of 100 tons/year from 100 power plants of 1000-MW capacity, experimental values for the electric field strength, and an assumed particle size of 1 μm diameter, the calculated power outlay is about 5 MW to escape the solar system.

Of the various ejection schemes considered, the one which appears best from the point of view of simplicity of design of accelerator, least cost, and high reliability is that involving ejection from earth orbit out of the ecliptic plane. An order-of-magnitude calculation indicates that the expected radioactive contamination of celestial bodies would be negligible.

Accelerator design requirements were obtained by seeking pertinent information on particle charging and acceleration reported in the literature. Such information is available from studies which are attempting to simulate micrometeoritic impact by the acceleration of μm -size particles to hypervelocities. From the experimental values of electric field strength available, the potentials necessary for acceleration of various size particles are calculated. Indications are that, for an assumed density of waste material (radioactive plus matrix material) of 3 g/cc, a potential of

20 million volts would be required to accelerate a 1- μ m-diameter particle to solar escape speed. Although not a limitation in itself, from a practical point of view it would be best to reduce this potential to a lower value. A number of suggestions are provided in the text.

An extensive literature search has been made seeking methods of producing small particles. Of all the methods surveyed, two in particular appear to be appropriate for this problem. They are the electrostatic atomization and unstable liquid jet methods. The former method is the most appealing since the particle charging is accomplished most conveniently by means of the high potential which is already required for the acceleration. The unstable liquid jet method, on the other hand, is more appealing from the point of view of generating uniform-size drops, but may require a separate charging scheme. A combination of the two methods may perhaps be the best method. A number of problems concerning waste processing must be solved before either method can be employed.

The system has been envisioned to be in two main sections, the accelerator section and the waste-container/particle-generation section. The accelerator section remains in orbit and is not recoverable. The waste-container/particle-generation section is recoverable, thereby making it possible to segregate the particle production equipment, which is the least reliable, from the more massive acceleration section. This scheme allows launch-pad checkout of the most sophisticated portion of the system, and recovery if a malfunction occurs in orbit. The waste-container/particle-generation section is docked with the accelerator section once in orbit, and no separate transfer of materials between vehicles is necessary. In essence, the waste container becomes the ion source for the accelerator stage.

10. FUTURE WORK

The amount of money remaining in the original allotment is \$6000. In this section, a program plan is presented for the expenditure of these remaining funds. The contractor will use its best efforts to:

1. Carefully survey the present-day proposed matrix materials and investigate if any of these are suitable for use with the

accelerator method proposed in this study. Particular attention is to be given to the ease with which such material can be made molten during the particle generation step and the viscous properties required in the liquid state for passage through small capillaries.

2. Investigate the materials problems which arise due to containment of the proposed molten matrix material. Present-day containment methods will be explored for suitability.
3. Investigate the various means by which the matrix can be "melted" once in orbit.
4. Investigate the problems which could arise when a large bundle of capillaries are used instead of a single capillary for formation of the liquid droplets.
5. Consider an accelerator design based on the best practical estimates of particle size and surface field strength for the particular matrix material considered. Include a crude cost and weight estimate.
6. Consider the power source needs for acceleration in more detail. This includes examining the potential use of on-board solar and thermionic converters and microwave beam transmission of power from the surface of the earth.
7. Investigate future research and development goals which include
 - a. Suggested laboratory studies needed for development of the small particle production technique.
 - b. Suggested studies needed for designing and testing the particle charging and ejection method.
 - c. Suggested research and development needed for the construction of a moderate-sized prototype.
 - d. Suggested studies required for the simulated space environment tests of the above prototype including lifetime testing of components.
8. Provide a crude cost and time schedule of the total research and development required prior to fabrication of the actual large-scale unit.

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APPENDIX 8.E

Terrestrial and Stellar Contamination from Space
Disposal of Nuclear Wastes

NASA-AMES, MS:202-8
Moffett Field, CA 94035
April 13, 1973

MEMORANDUM for Chief, Space Applications Branch

From : Lawrence C. Evans, Ph.D.

Subject: Terrestrial and Stellar Contamination from Space Disposal of
Nuclear Wastes

As we have discussed, objections have been raised concerning the use of space as a sink for radioactive waste because of the possibility of these wastes contaminating other terrestrial or stellar environments. In the case of solar disposal, it is feared that the waste will be swept out from the sun by the solar wind, and when re-encountering the Earth will contaminate the terrestrial environment. In the case of disposal by means of solar system ejection, since the spacecraft will eventually encounter another stellar system, there is the possibility that the nuclear waste will contaminate that system. It is therefore appropriate to investigate several aspects of this problem in order to evaluate the validity of such objections. The results of such an analysis are summarized here, and the documented results are available elsewhere. The primary conclusion is that such contamination, both of the terrestrial environment and of other stellar systems, is virtually impossible. The analysis leading to this conclusion with respect to solar disposal deals with the heliocentric vaporization distance of the waste, the ionization state of the atoms of waste, the effects of the juxta-solar radiation environment, the interplanetary propagation of the ions, and their access to the terrestrial environment. In the case of disposal through solar system ejection, the main considerations are the probability of encountering another stellar system, the distance traveled before such an encounter, and the time required for this travel. I will briefly summarize the solar disposal analysis, and then deal with stellar disposal.

Before the solar plasma and magnetic field can interact with the material being injected into the sun, the material must be vaporized and ionized. In order to deal with the effects of the juxta-solar environment, we must therefore estimate the heliocentric distance at which vaporization takes place, and we must also estimate the state of ionization in the atoms. The vaporization should occur within one or two solar radii above the surface of the photosphere, and all of the atoms of the nuclear waste products will be very highly ionized. For instance, on the average, Strontium atoms will be stripped of approximately 17 of their electrons,

while Cesium atoms will lose approximately 24 of their electrons. The ionization states which the atoms attain near the sun will be the ionization states which they will maintain throughout the interplanetary propagation.

Once the waste material is vaporized and ionized, it begins to be convected out from the sun with the solar wind plasma. In the meantime, however, these ions are emersed in the most intense radiation environment in the solar system, and it is critical to determine their survivability in such an environment. We can show that the ions will be within about 3 solar radii of the center of the sun for about 1 day. I have examined the survivability of the waste ions in terms of the probability of nuclear fission, and made the simplifying, if somewhat unrealistic, assumption that if a nucleus of waste material were to undergo fission, then the products would not be considered undesirable waste and their introduction into the terrestrial environment would not be considered contamination. I have only been concerned with fission induced by three types of incident radiation: solar gamma rays, neutrons, and protons. In the case of photo-induced fission, the fission cross-sections for most of the radioactive waste materials are low enough that this is not a significant effect. Lifetimes for the waste materials are probably on the order of 10^{19} seconds for photo-induced fission; compared with Sun-Earth travel time on the order of 5×10^5 seconds. The situation with particle-induced fission is, however, very much different. For proton-induced fission, it is particularly significant where the waste material is targeted on the Sun with respect to sources of charged particle radiation. If the waste materials are targeted into a quiet portion of the Sun, one might expect lifetimes on the order of 1.5×10^{17} seconds, whereas if the wastes reach the Sun at the same time and place as a moderately large solar flare, one would expect lifetimes on the order of 10^{-7} seconds. Although it is very difficult to anticipate and target for a solar flare event, it is possible to target for active regions on the Sun where the probability of flare events is much higher and where the ambient proton flux is also higher. Average lifetimes in these regions are perhaps 5×10^8 seconds. Neutrons will also induce fission, and, since they are uncharged particles, the fission cross-sections at low energies is much higher than those for incident protons. As a consequence, the probability of fission is very large even in the case of the waste materials being targeted for a quiet region of the Sun. In such a quiet region one would expect average lifetimes on the order of 10^{-3} seconds, whereas if the spacecraft were to reach an active region of the Sun, one might expect lifetimes on the order of 10^{-4} seconds. If the waste material encountered a flare, the lifetimes would be proportionately shorter, reaching 10^{-11} seconds for large flares. Thus even if the waste were to be targeted to a quiet region of the Sun, we would expect the amount of waste material to be decreased by a factor of 1 million in 0.017 second.

The problem of the propagation of the surviving waste ions from the vicinity of the Sun outward into and through interplanetary space is dominated by the fact that the ions have very low kinetic energies. As

a consequence, the spatial aspects of the motion of the waste ions in interplanetary space is determined by the configuration of the interplanetary magnetic field, and it is therefore to the stochastic and ergodic aspects of this field which we must address ourselves. A consideration of these aspects leads to the conclusion that even those waste nuclei which do survive the juxta-solar environment will be spread out into a very large diffuse cloud in interplanetary space. The fraction of these particles which would impinge upon the Earth is dependent upon the solar-longitude at which they were targeted, and this fraction varies from a maximum of 10^{-5} to a minimum of 10^{-100} .

For that fraction of the waste particles which escapes the Sun and subsequently impinges on the Earth, we finally come to the question of the contamination of the terrestrial environment. In order to determine the degree of contamination, we must determine the fraction of those particles impinging on the Earth which actually enter the atmosphere and where this contamination will occur. Since the waste atoms are highly ionized when they reach the vicinity of the Earth, their access to the terrestrial environment is determined by their interaction with the Earth's magnetic field. These particles are thus constrained to enter the magnetosphere through the low energy particle access regions far down the geomagnetic tail, and as a consequence, only a very small fraction (approximately 10^{-5}) of those particles near the Earth in interplanetary space will actually impinge upon the upper atmosphere. It can also be shown that this contamination will occur only near the geomagnetic poles.

Turning now from solar impact to an analysis of the contamination involved in disposing of nuclear waste by targeting them to escape the solar system, we ask the specific question of whether these wastes pose any realistic contamination threat to other stellar systems. Interstellar distances are so vast that one would expect that the travel times involved in encountering other stellar systems would be very large, and it is indeed so in this case. These distances are so large, in fact, that it would take several hundred decay lifetimes for the nuclear waste to reach any stellar systems. As a consequence, the amount of radioactive material would be reduced by a matter of at least 10^{100} . This essentially means that the nuclear waste cannot survive the long trip times involved in going from solar system to some other stellar system.

The net result of this analysis is, as you can see, the conclusion that disposing of nuclear waste in space by targeting them either to impact the Sun or to leave the solar system represents a final and complete disposal of these wastes and that wastes disposed of in this manner pose virtually no threat of contamination to either the terrestrial environment or any stellar environment. The details of this analysis are, as I say, contained in the more extensive report which is presently being typed and will soon be available for your perusal.

Lawrence C Evans
Lawrence C. Evans, Ph.D.

BLS

LCEvans:bh 4/13/73:5891

APPENDIX 8.F

Cost Estimate Data

TABLE F.1. Transuranic Encapsulation Capital and Operating Cost Summary(a)

	<u>\$/kg of Transuranic</u>
Operating Cost	
Direct Labor	\$ 65
Direct Material	340
Indirect Manufacturing Expense	65
Materials and Supplies	50
Utilities and Services	50
Overhead	<u>130</u>
Total Operating Cost	\$ 690
Capital Cost	
Building and Equipment Cost at 18% Per Year	
\$40,000,000 total cost; \$7,200,000 per year ÷ 2,600 kg/yr	<u>2,800</u>
Sub Total	3,490
Contingency, 30%	<u>1,010</u>
Total Encapsulation Cost	4,500
Levelized encapsulation cost charged at time of reprocessing (per Volume 1)	3,700
Levelized heat shield cost ^(b)	1,000
Total Levelized Cost for Encapsulation and Heat Shield	4,700

-
- a. Based on a plant encapsulating 2,600 kg/yr of transuranics or the transuranics from about 3,650 MT/yr of LWR uranium fuel.
- b. Based on heat shield cost estimate of \$220,000 for a unit, regardless of size (per personal conversation with John Vorreiter, NASA Ames, June 1973).

TABLE F.2. Direct Labor and Equipment Estimates,
Waste Transuranic Encapsulation for
Space Disposal

Basis: Plant to handle waste from two 1825 MT/Year
Reprocessing Plants, 2600 kg of transuranic
capacity per year, 180 operating days/year -
15 kg/day, 70% operating efficiency, 6 hr/
shift, throughput required - 4 kg/hr.

Operation	Throughput per Machine kg/hr	No. of Machines Required	Equipment Cost		No. of People Required
			Per Machine \$	Total Equipment, \$	
<u>Particle Preparation</u>					
1. Purification	5	1	20,000	20,000	1
2. Powder Consolidation	10	1	5,000	5,000	0.5
3. Crushing	10	1	5,000	5,000	0.5
4. Sizing	10	1	10,000	10,000	0.5
5. Sintering	10	1	25,000	25,000	0.5
6. Allowance for Recycle	--	--	--	65,000	2
<u>Coating</u>					
7. Coating Material Treatment	10	1	5,000	5,000	0.5
8. Magnesium Coating	10	1	20,000	20,000	0.5
9. 1st Tungsten Coating	2	2	20,000	40,000	2
10. Sacrificial Material Removal	5	1	20,000	20,000	1
11. Decontaminate	5	1	5,000	5,000	1
12. Second Phase Tungsten Coating	2	2	20,000	40,000	2
13. Aluminum Oxide Coating	10	1	10,000	10,000	1
<u>Assembly</u>					
14. LiH Treatment	10	1	20,000	20,000	0.5
15. Al Treatment	10	1	10,000	10,000	0.5
16. LiH-Al-Act. Oxide Mixing	10	1	5,000	5,000	0.5
17. Compacting	10	1	100,000	100,000	0.5
18. Clad/Prep	10	1	20,000	20,000	0.5
19. Assembly	10	1	30,000	30,000	0.5
20. Joining	10	1	<u>200,000</u>	<u>200,000</u>	<u>1</u>
TOTAL			\$ 550,000	\$ 675,000	17

Note: Using an average manpower salary of \$10,000/yr, total labor cost is
\$170,000/yr or \$65/kg of transuranics.

TABLE F.3. Direct Material Estimates, Waste Transuranic Encapsulation for Space Disposal

	Pounds of Material Per Pound of Transuranic(a)	Cost \$/lb of Material	Cost, \$/lb, of Transuranic
Magnesium	0.5	0.39	0.20
Tungsten Hexafluoride	2	10.00	20.00
Aluminum Oxide	0.2	0.14	0.03
Lithium Hydride	2.6	8.05	16.10
Aluminum	1.7	0.408	0.81
Tungsten	6.3	10.00	63.00
Stainless Steel	5.2	10.00	<u>52.00</u>
Total, \$/lb of Transuranic			\$152.14
Cost, \$/kg of Transuranic			\$340.00

a. See Table F.4 for approximate amount of materials used.

TABLE F.4. Approximate Weight Distribution of
Capsule of Transuranic Waste for
Space Disposal

<u>Material</u>	<u>Approximate Weight, kg/package(a)</u>	<u>Approximate Unit Weight, kg of Material Per kg of Transuranic</u>
Transuranic	191	1.0
Matrix	625	
Aluminum		1.7
LiH		1.7
Lithium Hydride	178	0.9
Tungsten Shielding	1,190	6.3
Stainless Steel Shell	640	5.2
Other, Re-entry Shield, Fission Products, Oxygen, Uranium	446	2.3
Total	3,270	

- a. Based on solar escape with capsule containing 0.1% of the original fission products (see Table 8.5).

APPENDIX

SECTION 9: TRANSMUTATION PROCESSING

APPENDIX 9.A

TRANSMUTATION BY ACCELERATORS

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APPENDIX 9.A

TRANSMUTATION BY ACCELERATORS

SUMMARY

The BeV proton-induced spallation device is the only accelerator transmutation concept showing promise. Other accelerator schemes can be ruled out on the basis of the criterion of energy balance. Conclusions reached are:

- (1) The inventory of intermediate half-life radioactive waste materials (Sr-90, Cs-137 and Kr-85) can be reduced significantly only by very high neutron flux levels, on the order of 10^{16} - 10^{17} neutrons/(cm²)(sec). Materials problems in the transmutation target, including its clad, and other parts of the system can be anticipated to be severe.
- (2) The energy invested per neutron produced when BeV protons strike heavy target material is the order of 50 MeV/neutron. The transmutation of Sr-90 and Cs-137 with perfect neutron utilization would thus use about 1.0 and 2.5 MeV of the energy per fission that produced them. Adding other fission products for transmutation or operation at 50% efficiency then uses at least 10% of the recovered energy from fission to transmute the fission products produced by that fission.
- (3) The spallation intense neutron source will produce large amounts of short-term radioactivity. The approximately 20 neutrons produced per incident BeV proton will come from on the order of 5 to 10 parent nuclei, almost all of the daughters being left in radioactive precursor states.
- (4) The inventory reduction is significant if long-life fission products I-129 or Tc-99 are exposed to a high thermal neutron flux. Since these materials have extremely long half-lives for natural decay (I-129 half-life is 16 million years), transmutation appears as a possible alternate to other means of handling these materials, particularly the higher hazard I-129.

9. A.2

- (5) Preliminary calculations indicate that if fission product nuclei are used directly as the target for the accelerated protons, then the transmutation might be technically feasible. The capital cost of the many accelerators which would be required, however, would be prohibitive. In order to have any practical possibility, very large advances are required in high-energy accelerator technology.

It is therefore likely that accelerators would find application only for eliminating isotopes of special concern, such as I-129.

DETAILED RESULTS

A. Charged Particle - Nuclear Reactions

It can be demonstrated from fundamental considerations that direct nuclear reactions of charged particles from accelerators with a few tens of MeV energy are not particularly attractive for radioactive waste burnup. Charged particles from fusion reactions and photonuclear reactions will be considered independently, as will BeV proton induced spallation processes.

Proton penetration at intermediate Z nuclei requires energy on the order of tens of MeV, with more energy required for heavier particle penetration. The larger energy requirement for penetration by nucleons with higher charge than protons and their larger Coulomb energy losses in material targets make them less likely candidates for transmutation of wastes.

Consider a charged particle slowing in a medium containing transmutation targets. The number of nuclear reactions N per projectile in slowing from some initial energy E_i to a reaction threshold energy E_t is given by

$$N = \int_{E_i}^{E_t} \frac{N_t \sigma_t}{\left(\frac{dE}{ds}\right)} dE \quad (A1)$$

9. A.3

where N_t is number density of transmutation target nuclei and σ_t is the cross section per nucleus. The expression dE/ds is the particle's energy loss per unit path length. An effective cross section $\langle \sigma_t \rangle$ can be defined by writing

$$N = N_t \langle \sigma_t \rangle \int_{E_i}^{E_t} \frac{dE}{\left(\frac{dE}{ds}\right)} \quad (A2)$$

The energy loss per unit path can be re-expressed in terms of a density-independent tabulated function of material and energy

$$\frac{dE}{ds} = \rho \frac{dE}{dM} \quad (A3)$$

where M is the mass per unit area of material traversed and ρ is mass density of the material.

The average of $(dE/dM)^{-1}$ for reasonable energy intervals can be taken as some multiple of a $(dE/dM)^{-1}$ at ten MeV in aluminum.⁽¹⁾

$$N = a \frac{N_t}{\rho} \langle \sigma_t \rangle \frac{E_i - E_t}{\left(\frac{dE}{dM}\right)_{10 \text{ MeV, Al}}} \quad (A4)$$

The effective cross section $\langle \sigma_t \rangle$ must be some number b of barns, while the energy range $E_i - E_t$ must be some number c of tens of MeV. The ratio N_t/ρ will be no higher than in the case of a pure isotopic target, for which case

$$\frac{N_t}{\rho} = \frac{N_0}{A} \quad (A5)$$

Here N_0 is Avogadro's number and A is atomic mass number.

Accordingly, Equation (A4) can be written

$$\begin{aligned} N &= abcd \times \frac{6.023 \times 10^{23}}{90} \times 10^{-24} \times \frac{10}{3.4 \times 10} \\ &= abcd \times 2 \times 10^{-3} \end{aligned} \quad (A6)$$

9. A.4

where d is a factor to account for departure of target atomic mass from 90. Since units were chosen to make $abcd$ of order unity, it can be concluded that only on the order of 10^{-3} of the charged particles experiences a nuclear reaction. Since the energy cost per charged particle is greater than $c \times 10$ MeV, the energy input per reaction is greater than

$$\frac{E}{N} = \frac{1}{abd} \times 5000 \text{ MeV/reaction.} \quad (\text{A7})$$

This energy per radioactive waste-eliminating reaction should be compared with 200 MeV/fission thermal energy available from the fission process. Even a multiple reaction (waste eliminating (p,n) reaction followed by waste eliminating (n, γ) reaction, etc.) does not help much. Use of energy from those waste-eliminating reactions which are exothermic does not aid materially since they tend to release less than 10 MeV per reaction.

Energy deposited by the charged particles in slowing down can be recovered only by thermodynamic means since they are quickly thermalized. Thermodynamic efficiency for the utilization of this energy would probably be no greater than 0.4 as determined by materials considerations and available reservoirs. Utilizing the thermal energy supplied to the target, an energy increment ΔE per reaction must still be supplied, satisfying

$$E = (1 - \eta) \frac{E}{N} \quad (\text{A8})$$

$$> (1 - .4) \cdot \frac{1}{abd} \times 5000 \text{ MeV/reaction} \quad (\text{A9})$$

$$> \frac{1}{abd} \times 3000 \text{ MeV/reaction.} \quad (\text{A10})$$

Since the number of fission products expected to transmute per fission is of the order of 10^{-2} to unity, the energy expenditure required with accelerator beams of charged particles of a few tens of MeV energy by direct interaction is excessive.

9. A.5

To make waste transmutation by beams of charged particles from accelerators more nearly feasible, two improvements are needed. First, the probability of a reaction must be increased. Second, the number of waste nuclei eliminated per reaction must be increased. If the energy of a proton is increased above 150 MeV, the mean free path for nuclear interaction becomes shorter than the range. This is principally because the energy loss per unit path length decreases with energy at high energy. Proton interactions at high energy in high Z targets result in the emission of multiple energetic cascade particles which in turn evaporate many neutrons.

B. Beta Decay Acceleration by Coulomb Excitation

Beta decay from certain excited states of some nuclides proceeds more rapidly than beta decay from the ground state or in other cases more rapidly from the ground state than from certain metastable states. If this is the case for a radioactive fission product, it may be possible to reduce the effective lifetime of a nuclide by inducing a transition to a more rapidly decaying state.

The FP isotope Kr-85 is representative of some of the circumstances desired. Beta decay from the $9/2^+$ ground state proceeds with a 10.76 year half-life. The $1/2^-$ excited state at 0.305 MeV decays with a 4.4 hour half-life, 77% by beta decay to the $3/2^-$ state of Rb-85 and 23% by gamma decay to the $9/2^+$ Kr-85 ground state. Figure A.1 displays an energy level diagram for Kr-85. The Kr-85 case also illustrates the difficulty in attempting to accelerate beta decay. The $1/2^- - 9/2^+$ transition is slow because of the 4 unit change in angular momentum required; hence, the β^- decay to the $3/2^-$ state of Rb-85 competes favorably with gamma emission. To excite Kr-85 to the $1/2^-$ state also requires a change of 4 units of angular momentum. Large angular momentum changes can occur in Coulomb excitation, and the cross section falls off less rapidly for higher angular momentum changes than do decay rates in gamma decay.

FIGURE A.1. Energy Level Diagram for Kr-85

9. A.7

Therefore, necessary, but not necessarily sufficient, conditions on cross sections may exist for Coulomb excitation as a possible excitation method for radioactive waste transmutation. The recoverable energy per fission energy and the cumulative yields of objectionable fission products sets an upper limit $(dE/dN)_{\max}$ to the energy expended per fission product nucleus transmuted. If it is decided to expend no more than a fraction of the recovered energy per fission in treating f fission products per fission, then

$$\left(\frac{dE}{dN}\right)_{\max} = (F\eta/f) \times 190 \text{ MeV/fission} \quad (\text{A11})$$

where η is the thermodynamic efficiency of the reactor. A charged particle passing through fission products of number density n will have a probability per unit path length dN/ds of eliminating a fission product given by

$$\frac{dN}{ds} = n\sigma \quad (\text{A12})$$

where σ is the cross section for a reaction leading to elimination of the fission product. The particle meanwhile will be losing energy at a rate

$$\frac{dE}{ds} = \rho \frac{dE}{dM} \quad (\text{A13})$$

where dE/ds is energy lost per unit path length, dE/dM is energy lost per unit mass/area, and ρ is the density. The energy expended per fission product eliminated will be

$$\frac{dE}{dN} = \frac{dE/ds}{dN/ds} = \frac{\rho dE/dM}{n\sigma} = \frac{A}{N_0\sigma} \frac{dE}{dM} \quad (\text{A14})$$

where A is atomic number of the fission product and N_0 is Avogadro's number. Requiring

$$\frac{dE}{dN} < \left(\frac{dE}{dN}\right)_{\max} \quad (\text{A15})$$

9. A.8

requires

$$\sigma > \frac{A}{N_0} \frac{dE}{dM} / \left(\frac{dE}{dN} \right)_{\max} \quad (A16)$$

This requirement on the cross section for transmutation elimination must be satisfied for at least part of the charged particle track if the elimination is to be economically feasible.

To exemplify the above criteria, consider Kr-85 with $f = 0.013$ (0.013 nuclei per fission). Assume (optimistically) willingness to expend 5% of the recovered fission energy to eliminate this isotope. Then an upper limit on energy expended per Kr-85 nucleus transmuted is:

$$\begin{aligned} \left(\frac{dE}{dN} \right)_{\max} &= (F_f/f) \times 190 \text{ MeV} \\ &= (.05 \times .4 / .013) \times 190 \text{ MeV} \\ &= 292 \text{ MeV/Kr-85 nucleus.} \end{aligned} \quad (A17)$$

Assuming a 40% thermodynamic efficiency for the power plant and using tabulated and calculated values for the stopping power of Kr-85, the cross sections required at various energies are obtained for economically competitive transmutation by protons as shown in Table A.1.

To estimate whether the cross section magnitudes needed for economical waste transmutation occur physically, the semiclassical quantum theory of the Coulomb excitation process is examined. (Semi-classical means here that the incident ion is considered to move in the

TABLE 9.A.1. Cross Section Needed For Economical Transmutation
of Kr-85 by Protons

Assumption: 5% maximum of recovered fission energy to treat 0.013
Kr-85 nuclei per fission.

<u>Proton Energy (MeV)</u>	<u>Required σ (Barns)</u>
5.	20.
7.5	15.
10.	12.
12.5	10.
15.	8.8
20.	7.0
50.	3.3
100.	1.9
200	1.0
300	0.73
400	0.57

9. A.10

classical Rutherford trajectory). The cross section for Coulomb excitation of a 2^λ order multipole transition in the semi-classical approximation is given by

$$\sigma_{E\lambda} = \left(\frac{Z_i e^2}{\hbar u} \right)^2 a^{-2\lambda+2} B(E\lambda) f_{E\lambda}(\xi) \quad (A18)$$

where

$Z_i e$ = incident ion charge

u = initial relative velocity

$\hbar$ = Planck's constant/ 2π

a = $Z_i Z_e^2 / Mu^2$

= half distance of closest approach in head on collision

$\xi = \frac{a\omega}{u} \approx \frac{Z_i Z_e^2}{\hbar u} \frac{\Delta E}{2E}$

$B(E\lambda)$ = reduced electric multipole transition probability of multipolarity λ

$$= \sum_{M_i M_f \mu} | \langle I_i M_i | M(E\lambda, \mu) | I_f M_f \rangle |^2$$

$M(E\lambda, \mu)$ = nuclear electric multipole operator of order λ

$$= r^\lambda Y_{\lambda\mu}(\theta, \phi) \rho(\underline{r}) d\tau$$

Here $\rho(\underline{r})$ is the nuclear charge density operator.

The functions $f_E(\xi)$ are approximately constant for $\xi \ll 1$ (although quite different for different λ) but fall off rapidly for $\xi > 1$. Coulomb excitation cross sections tend to be too small to observe for $\xi > 1$.

The condition $\xi < 1$ requires that the energy of the incident ion be sufficiently large to excite a given excited state. To Coulomb excite the 0.305 MeV level in Kr-85 requires protons of energy of 1 MeV or greater.

To estimate the magnitude of the cross sections for the high order multipole transitions needed for beta decay acceleration by Coulomb excitation, following J. D. Newton⁽²⁾ and using a "single particle model" estimate of the reduced electric multiple transition probability:

$$B_{sp}(E\lambda) = \frac{e^2}{4\pi} R^{2\lambda} \quad (A19)$$

where $R = r_0 A^{1/3}$ is the nuclear radius. This model gives as the Coulomb cross section of electric multipole order 2^λ :

$$\sigma_{E\lambda} = \left(\frac{Z_1 e}{\hbar u} \right)^2 \frac{e^2}{4\pi} a^2 \left(\frac{R}{a} \right)^{2\lambda} f_{E\lambda}(\xi) \quad (A20)$$

The values of $f_{E\lambda}(\xi)$ were evaluated numerically by Adler, et al.,⁽³⁾ and were used with the preceding expression to compute the Coulomb excitation cross section measurements for a nucleus with $A = 85$, $Z = 36$, and an excited state of 0.305 MeV with appropriate spin and parity for excitation by various order multipoles. The actual $9/2^+ \rightarrow 1/2^-$ excitation in Kr-85 would require an E5 transition (electric 128-pole) since an E4 transition would not give the parity change and lower order multipole

9. A.12

transitions then E4 will not give the 4 unit change in angular momentum. The cross section estimates are given in Table 9.A.2 using the $f_{E\lambda}(\xi)$ of Adler, et al. The Table stops at E4 because Adler, et al., gives $f_{E\lambda}(\xi)$ values only to $\lambda = 4$.

From Table 9.A.2 it appears that the minimal required cross sections of Table 9.A.1 will not be obtained for objectionable radioactive isotopes. An open question is whether the "single particle model" reduced transition probabilities $B(E\lambda)$ represent an adequate representation for a realistic assessment. The $B(E1)$ values found experimentally are smaller by factors 10^3 to 10^5 than the $B_{sp}(E1)$ used here, but $B(E2)$ values are frequently enhanced by factors of up to 100 over the single particle values. Enhancements of higher order pole moments probably also occur, but E3 transitions are the highest order Coulomb excitations known to have been identified. Second order quadrupole transitions E2,E2 occur and may be applicable in obtaining some desired states, but seem inapplicable to Kr-85 for two reasons: (1) the needed parity change would not be given, and (2) an intermediate state accessible by E2 transition is not known (though it may exist).

Coulomb excitation may occur simultaneously to a number of energy levels higher than the beta decaying metastable level with subsequent cascading that leads to the metastable state. Assume that five such levels exist, each having the "maximum plausible" cross section of 3 barns each. Then if all levels cascaded without loss to the known beta decaying metastable state and 77% of the excitations result in beta decay, the effective cross section for elimination would be 11.5 barns, which is close to the estimated "local minimum" cross section requirement for 10 MeV protons given here. The actual cross sections should be higher still, since compensation for

TABLE 9.A.2. Coulomb Cross Section Estimates For A Z=36, A=85
Nucleus Using Single Particle Model Reduced
Transition Probabilities

<u>Proton Energy</u>	<u>σ_{E1}(millibarns)</u>	<u>σ_{E2}(millibarns)</u>	<u>σ_{E3} (millibarns)</u>	<u>σ_{E4}(millibarns)</u>
20 MeV	3.1	0.57	0.45	0.65
10 MeV	4.4	0.28	0.06	0.02

accelerator inefficiency and the almost total loss of the last MeV of proton energy in stopping in the target material is needed. That such a fortuitous combination of physical properties exists is unlikely but cannot be ruled out.

This discussion has focused attention on numerical examples of beta decay acceleration by Coulomb excitation on Kr-85 only because it meets some of the physics requirements. It is in fact a poor candidate for transmutation, since its 10.8 year half-life and noble gas nature suggest retrievable storage. Sr-90 does not look like a good candidate for beta decay acceleration by Coulomb excitation, because its approximately 11 known excited states between 0.83 and 5.23 MeV gamma decay rapidly. The only hope for this concept for Sr-90 would be that as yet undiscovered metastable states at energy much less than 0.83 MeV exist. It is not known whether this can be ruled out from present experimental information, although the existence of such states is certainly doubtful. Cs-137 is also lacking in identified low energy fast beta decaying levels accessible by Coulomb excitation; the excitations result only in rapid gamma decay back to the ground state.

I-129 has a wealth of low lying energy levels, including a 0.0178 MeV $5/2^+$ level which should be readily excitable by a Coulomb excitation E2 transition. Unfortunately, the fairly rapid M1 (magnetic dipole) transition returns it to the ground state with a 15 nanosecond half-life and negligible beta decay. No beta decay has been observed from any of the low-lying excited levels of Tc-99.

9. A.15

It is concluded that beta decay acceleration by Coulomb excitation is probably not applicable to most of the problem fission products, although surveillance of nuclear physics developments for these materials may be warranted. It does not appear likely that any of the other fission products are amenable to beta decay acceleration Coulomb excitation. However, materials like Ho-166 whose lowest energy state decays more rapidly than a metastable state, as shown in Fig. A.2, may merit further study.

FIGURE A.2. Ho-166 Levels and Decay

C. Photon Transmutation Processes

Electron Bremsstrahlung - As an alternate approach to fission waste burnup by transmutation, one can consider bombarding targets containing objectionable waste with electron beams in the tens to low hundreds of MeV range. The electrons produce a "shower" of photons from Bremsstrahlung, from annihilation of electron-positron pairs, and from other processes. Some of the photons in the "giant" resonance energy region will undergo nuclear interactions such as (γ, n) , (γ, p) , $(\gamma, 2n)$, or (γ, np) .

The feasibility criteria for such a system are the reaction yields; i.e., number of reactions per incident electron, and the energy supplied per reaction.

The parameters for optimization of this concept are influenced by the following considerations:

- (1) Initial electron energy should be sufficiently high that radiation energy losses in matter are of the same order as or higher than ionization losses. Above some critical energy E_c radiation losses dominate. Sample critical energies are 6.9, 24, and 103 MeV for Pb, Fe, and C, respectively.
- (2) The energy spectrum of the bremsstrahlung should overlap the "giant resonance" region of the target. The photon energy of the center of the giant resonance is given approximately by

$$E_{\text{res}} = 40 A^{-0.2} \text{ MeV} \quad (\text{A21})$$

where A is the nuclear mass number.

- (3) The bremsstrahlung spectrum has as an upper bound the energy of the decelerating electron, with most of the radiation being emitted at much lower energy.

These three considerations taken together dictate that the electron beam have at least tens of MeV initial energy.

Barber and George⁽⁴⁾ suggest that an approximate yield of neutrons from thick targets bombarded by electrons can be calculated from

$$Y(Z, E_0) = K \left(\int \sigma(Z, k) dk \right) \frac{R(Z)}{I(Z) + R(Z)} \frac{E_0}{k_0^2(Z)} \frac{t_r(Z)}{A} \quad (A22)$$

where $t_r(Z)$ is the number of grams/cm² in one radiation length, A is the atomic weight of the target material, E_0 the incident electron energy, and k_0 is the photon energy at the peak of the giant dipole resonance. The integral

$$\int \sigma(Z, k) dk \quad (A23)$$

is the photon energy integral for producing a neutron by any reaction. The expression

$$\frac{R(Z)}{I(Z) + R(Z)} \quad (A24)$$

is a ratio of the electron energy loss by radiation to that by both ionization and radiation. The constant K was determined to give agreement with their experimental yield in lead with $E_0 = 34$ MeV. Taking ratios of ionization and radiation energy losses at an electron energy of 30 MeV, Barber and George⁽⁴⁾ generate anticipated yields of neutrons per incident 34 MeV electron shown in Table A.3.

9. A.18

From the data in Table A.3 it is clear that neutron yields from the electron bremsstrahlung-photonuclear reaction process increase with increasing nuclear size. Hence, any system for radioactive waste transmutation using 34 MeV electrons is likely to have a photoneutron yield less than that of uranium, which Barber and George⁽⁴⁾ estimate as

$$Y(92, 34 \text{ MeV}) = 2.59 \times 10^{-3} \text{ neutrons/electron} \quad (\text{A25})$$

A number of experiments^(5,6) indicate a roughly linear increase in yield with incident electron energy above a few tens of MeV. Hence, yield per electron can be increased by increasing electron energy, but energy expended per reaction could not be reduced significantly. Energy per reaction E/N will be of the order

$$\frac{E}{N} = \frac{34 \text{ MeV}}{2.59 \times 10^{-3}} = 1.31 \times 10^4 \text{ MeV/reaction} \quad (\text{A26})$$

Since this is very large compared with 200 MeV/fission, the concept does not meet the energy balance criterion.

TABLE 9.A.3. Calculated Z-Dependence of Neutron Yields from
34 MeV Electrons
(Alsmiller and Moran)⁽⁵⁾

Element	$\int_0^{34 \text{ MeV}} \sigma dk$ MeV - mb	k_o MeV	t_r g/cm ²	I at 30 MeV MeV gm ⁻¹ cm ²	R at 30 MeV MeV gm ⁻¹ cm ²	Expected Yield x 10 ⁴	Expected Yield (Normalized to exp Pb yield) x 10 ⁴
C	50	23	44	1.7	.44	2.45	2.84
Al	132	19.2	24.4	1.7	.79	3.53	4.10
Cu	870	18.5	12.4	1.5	1.6	8.72	10.1
Ta	3800	15	6.4	1.25	3.0	14.4	16.6
Pb	4800	13.7	5.8	1.2	3.3	18.0	20.8
U	7200	13.8	5.4	1.2	3.6	22.4	25.9

Stimulated Gamma Emission - The phenomenon of stimulated emission appears attractive on first thought as a radioactive waste management technique. A material matrix is visualized including a waste isotope emitting its characteristic radiation, with probability of emission by a given nucleus increasing with increasing intensity. The result would be the elimination of the radiation hazard on a short time scale rather than at the natural decay rate. The two major problems which tend to discourage hopes for this concept are:

- Stimulated emission is possible only for neutral Boson emission according to best available theory. For practical purposes, this means photon emission. There is no stimulated emission phenomenon in beta decay.
- The low attenuation material medium needed for stimulated gamma decay may be unattainable.

Applicability only to gamma decay, limits interest in stimulated emission for radioactive waste to a few slow gamma emitters. Metastable Ho-166 and metastable Sn-121 illustrate a situation which might be considered for stimulated emission. If a transition from the 1200 year half-life metastable state of Ho-166 to the 26.9 hour beta-decaying ground state could be induced, (see Figure A.2) the retention time for this component of radioactive waste could be reduced. Similarly, it may be desirable to stimulate a gamma transition from the 76 year half-life $11/2^-$ state of Sn-121 to the 27 hour $3/2^+$ state.

Consider the requirements of stimulated gamma transitions in an extended medium. If gamma stimulation cannot be achieved in an infinite medium, then it clearly cannot be achieved in a finite one in which diffraction and mirror losses occur. The condition for stimulated

9. A.21

emission in an extended medium is that the stimulated emission gain per unit length exceed the attenuation loss. The intensity change per unit length dI/ds will be

$$\frac{dI(\nu)}{ds} = \frac{h\nu B_{ij}(\nu) I(\nu)}{c} \left(n_i - \frac{g_i}{g_j} n_j \right) - BI(\nu) \quad , \quad (A27)$$

The first term on the right hand side represents stimulated emission gain, while the second represents attenuation. The coefficient $B_{ij}(\nu)$ is the Einstein induced emission coefficient, related to the spontaneous transition probability $A_{ij}(\nu)$ by

$$B_{ij}(\nu) = \frac{c^3}{8\pi h\nu^3} A_{ij}(\nu) \quad (A28)$$

The spontaneous transition probability $A_{ij}(\nu)$ is related to the transition probability A_{ij} from state i to state j by a line shape factor:

$$A_{ij}(\nu) = \frac{A_{ij}}{\pi} \frac{\Delta\nu}{(\nu-\nu_0)^2 + (\Delta\nu)^2} \quad (A29)$$

The preceding line shape is appropriate in the case of a homogeneously broadened line, i.e., one whose width is determined by the finite lifetimes of the upper and lower states. For the materials of interest for stimulated gamma emission, this would probably occur only at cryogenic temperatures in nearly perfect crystals. For such a homogeneously broadened line, the line width is related to the total decay constants of states i and j by

$$\Delta\nu = (A_i + A_j)/2\pi \quad (A30)$$

where

$$A_i = \sum_k A_{ik}$$

9. A.22

is the decay rate constant from state i and the sum is over states to which decay occurs. The criterion for stimulated emission at line center becomes

$$\frac{c^2}{4\pi\nu^2} \frac{A_{ij}}{A_i + A_j} \left(n_i - \frac{g_i}{g_j} n_j \right) > \beta \quad (\text{A31})$$

This expression shows the devastating effect on the requirements for stimulated emission which stem from alternate decay modes of the upper excited level. This can be seen from the alternate form:

$$\frac{c^2}{4\pi\nu^2} \frac{\phi_{ij}}{1 + (\tau_i/\tau_j)} \left(n_i - \frac{g_i}{g_j} n_j \right) > \beta \quad (\text{A32})$$

where ϕ_{ij} is the fraction of the decay from state i that goes to state j and τ_i is the lifetime of state i .

The attenuation coefficient β describes all processes which take a gamma photon out of the beam other than nuclear resonant absorption by the isotope of interest. Here any scattering process that changes the direction of the photon or the phase of its wave function effectively removes it from the coherent beam, just as do all absorption processes.

A transition from the Ho-166 metastable state to the Ho-166 ground state would produce a 7 keV gamma. For this gamma, the photoelectric effect is the dominant attenuation mechanism. Using graph values for attenuation of 10 keV gammas in lead and an extrapolation formula given by Evans,⁽⁷⁾ we estimate the attenuation in holmium as

9. A.23

$$\beta \approx 390 \text{ cm}^{-1}$$

For the desired holmium transition we estimate

$$\frac{c^2}{4 \pi \nu^2} \frac{\phi_{ij}}{1 + (\tau_i/\tau_j)} \left(n_i - \frac{g_i}{g_j} n_j \right) = w \phi_{ij} \left(1 - \frac{g_i}{g_j} \frac{n_j}{n_i} \right) \times 1.23 \text{ cm}^{-1} \quad (\text{A33})$$

where w is the ratio of density of the holmium in the material medium to the density of elemental holmium. Since each of the symbolic factors on the right side of the preceding equation is less than unity, the criterion for stimulated emission clearly cannot be met. The factor ϕ_{ij} is in fact much less than unity because most of the decay is by beta emission rather than the highly forbidden gamma transition we seek.

Clearly, stimulated emission for radioactive waste management requires some orders of magnitude reduction in attenuation of gammas in the material medium. Such a reduction in attenuation in specific crystallographic directions has been observed (Borrmann effect) and has been proposed⁽⁸⁾ for stimulated gamma emission. An electromagnetic wave of appropriate wavelength incident on certain crystals at an angle appropriate for Bragg scattering will split into a pair of transmitted waves (forward diffracted) in addition to the wave diffracted out of the crystal. If the transmitted waves interfere to give electric field minima at the planes of the atoms, attenuation is dramatically reduced. Difficulties with applying this anomalous transmission to stimulated gamma emission are:

9. A.24

- (1) Having electric field minima at the atom planes may reduce stimulated emission as much as it reduces attenuation.⁽⁹⁾
- (2) A highly perfect crystal would have to be grown in the presence of disruptive radiations.

Stimulated emission for radioactive waste elimination would require considerable development work to be applicable only to a limited set of radioactive nuclides. It may well be totally impossible. The possibility of stimulated gamma emission as a radioactive waste transmutation process depends on technical developments which cannot be anticipated and appear to be unlikely.

D. Spallation Accelerators

High energy ($\geq 1,000$ MeV), large current proton accelerators have been proposed which would provide the most intense, continuously operating source of neutrons yet attained. The largest spallation accelerator which has been seriously studied was the Intense Neutron Generator (ING) which was proposed by the Canadians⁽¹⁰⁾ (Atomic Energy of Canada, Ltd.). The conceptual design of the ING envisioned accelerating a 65 mA beam of protons to an energy of 1 GeV (1,000 MeV). The beam was to produce high energy neutrons primarily through the spallation process in a Pb-Bi target. The high energy neutrons were to be moderated in a surrounding D_2O medium to provide a thermal neutron flux of at least 10^{16} n-cm⁻²-sec⁻¹. Thermal flux values of this magnitude are required for the transmutation of low neutron cross section FP isotopes. In 1967, Steinberg, who had for some years been studying neutron transmutation of fission products,^(11,12) proposed the usage of a higher powered version of the proposed ING for this purpose. In 1971 an ORNL group⁽¹³⁾ also reported brief conclusions regarding Steinberg's proposal. A Japanese atomic energy team⁽¹⁴⁾ has also studied the use of a spallation accelerator for the transmutation of fission products. In addition, several studies have been made of the use of spallation accelerators to transmute fertile to fissile material, a concept which is closely related in technical feasibility to the FP transmutation concept.

The Spallation Accelerator - In order to discuss the technical feasibility of transmutation using a spallation accelerator it is necessary to understand the physical processes which take place in the spallation target and environs so that feasibility factors can be clearly identified.

The range of a 1 GeV proton in a heavy element is the order of 30-60 cm. In the spallation reaction of 1 GeV protons about 0.5 pi mesons with an average energy of about 100 MeV are formed independent of target material. In a thick target these pions rapidly undergo nuclear interaction or decay within a short distance of where they are formed. A few percent (~5%) of the incident protons result in a component of very high energy neutrons.⁽¹⁵⁾ These neutrons are referred to as the cascade neutron component and are characterized by an evaporation energy spectrum with a Maxwellian temperature of the order of 250 MeV for a 1 GeV proton beam.⁽¹⁵⁾ The highly excited residual nuclei formed in the primary cascade reactions can decay by the evaporation of one or more nucleons which may be followed by fission and further neutron emission. The cascade nucleus may also fragment into two or more excited fragments followed by neutron evaporation. The neutrons evaporated from highly excited nuclei are characterized by an energy spectrum with a characteristic temperature of 2.5-3 MeV. The low energy part of this neutron spectrum is degraded rapidly to a few tenths of an MeV by neutron inelastic scattering in the target material.

The dominant spallation reactions in a Pb-Bi target lead to a variety of undesirable radioactive nuclei with Z(proton) and N(neutron) values slightly less than those of the struck nucleus.⁽¹⁶⁾ These include:

9. A.27

<u>Nucleus</u>	<u>T_{1/2}</u>
Bi-208	3.68×10^5 yr
Bi-207	38 yr
Bi-206	6.24 d
Pb-205	1.4×10^7 yr
Pb-203	52 h
Tl-204	3.78 yr
Tl-202	12.2 d

Very short half life products are not shown on the above table. The sum of all of the nuclei of this type formed in the spallation reaction is 0.5 to <1 per incident proton.

The fission plus fragmentation cross section of Pb and Bi is not well known. At a proton energy of 340 MeV this cross section is about 0.2b which is about 13% of the total nonelastic cross section. The cross section for fission is expected to peak near 750 MeV. According to theoretical predictions the fission cross section will never exceed half of the non-elastic cross section. Thus near 1 GeV an estimate of 0.3 ± 0.15 would seem to be reasonable for the fraction of nonelastic processes leading to fission and fragmentation. Within a few percent, all incident protons undergo a nonelastic event in the target.

The kinetic energies of ultimate fission and fragmentation nuclei and of fission neutrons represents the exoergic reaction energy liberated in the target and environs which is gained in the spallation reaction. Kinetic energies of fragments has been measured to be 111 MeV per fission for 435 MeV protons on bismuth. By comparison with the variation of fission excitation energies with incident proton energy on other nuclei it appears that the fragment kinetic energy should be the order of 200 MeV per fission on Pb or Bi near 1 GeV.

The additional energy liberated in the target due to fission fragments can then be estimated as

$$(0.3 \pm 0.15) \times (200 \pm 50) = 60 \pm 35 \text{ MeV/p}$$

It is known from experiment that there are about 20 neutrons produced per 1 GeV proton incident on a thick lead target. Of this number, perhaps half represent exoergic fission neutrons kinetic energy. Assuming an average energy of 2 MeV for these neutrons would result in an energy augmentation of about 20 MeV. Combined with the fission fragment kinetic energy we then expect an energy liberation of some 1080 MeV per 1000 MeV proton. This energy augmentation is considerably less than has been estimated for 900 MeV protons incident on a U-235 target. In this case a value of 1295 MeV per 900 MeV proton has been estimated.⁽¹⁵⁾ The fission cross section for U-238 is, however, considerably higher than for Pb or Bi and also U-238 fissions occur from secondary neutron reactions.

The product nuclei resulting from 1 GeV proton fission and fragmentation events have a distribution with mass number which is quite different in character from that observed for slow-neutron induced fission. For protons with energies approaching 0.5 GeV the distribution of fragment masses is roughly symmetric about $A = 95$ with a near constant yield for some ± 20 mass units. At a proton energy of 3 GeV, however, the distribution of fragments is constant with A to within a factor of several. Thus, a whole host of radioactive decay products are produced in the spallation reaction, many of which are not encountered in neutron induced fission.

In the ING concept the Pb-Bi target of a few cms radius was to be surrounded by a beryllium sleeve to achieve further multiplication of the source neutrons by $n, 2n$ reactions on beryllium. The projected source strength including neutron multiplication in $\text{Be}^{(10)}$ was $1 \times 10^{19} \text{ n-sec}^{-1}$. It is important to note that this is a production of 1-1/4 moles of neutrons per day and that this figure is absolute maximum quantity of FP nuclei that could be transmuted without further neutron multiplication.

The spallation target with Be sleeve of the ING was to have been surrounded by a D_2O moderator tank to provide a maximum thermal neutron flux of $10^{16} \text{ n-cm}^{-2}\text{-sec}^{-1}$ as a source for neutron physics experiments. This flux value depends upon no absorption of neutrons other than in the D_2O moderator. The presence of any absorbing FP target material would lower this value.

Feasibility - The feasibility criteria are applied to determine the merit of using a spallation accelerator to transmute fission products.

Energy Balance

The required electrical energy, E , input to the spallation accelerator to produce N net neutrons available for fission product transmutation can be expressed as:

$$\frac{E}{N} = \frac{E_0/\epsilon_a - \eta_{th}(E_{th})}{(N' - F)\epsilon_b} \quad (A34)$$

where:

E_0 = the energy of the proton accelerator

ϵ_a = $\frac{\text{proton beam energy/proton on target}}{\text{input accelerator power/proton on target}}$

E_{th} = thermal energy deposited in target and environs

η_{th} = efficiency of conversion of E_{th} to electrical energy

N' = neutrons produced per incident proton

F = radioactive nuclei produced per incident proton

ϵ_b = fraction of neutrons absorbed in FP

A minimum energy/FP can be estimated by assuming the most optimistic conditions including the maximum conversion of thermal energy in the target to electrical energy. For the minimum value of E/N it is assumed that

$$E_0 = 1000 \text{ MeV}$$

$$E_{th} = 1080 \text{ MeV}$$

$$\eta_{th} = 0.40$$

$$\epsilon_a = 1 = \epsilon_b$$

$$N' = 24.6, F = 0$$

9. A.31

Then the minimum value

$$\frac{E}{N} \geq \frac{1000 - 0.4 (1080)}{24.6} = 23 \text{ MeV/FP} \quad (\text{A35})$$

A reasonable upper bound to E/N can be obtained from more pessimistic assumptions. For the first assumption it can readily be assumed that no thermodynamic conversion of target heat to electricity is possible. There are a number of possible reasons why this might be a realistic assumption. The ING was designed to utilize a flowing liquid lead-bismuth eutectic target without a window for the proton beam. The liquid target temperature is then limited in order that the eutectic vapor pressure not exceed the 10^{-6} torr vacuum requirement for beam transport. Besides being thermodynamically limited in temperature, materials damage problems could preclude the use of an effective heat transfer system near the target. The 100% efficiency assumed for the conversion of electrical energy to proton beam energy is clearly idealistic. Davidenko⁽¹⁷⁾ has suggested that a value of 50% would be a reasonably optimistic assumption. He also suggests a neutron utilization of $\epsilon_b = 0.8$. The number of radioactive nuclei produced per proton results from primary spallation nuclei and fission-fragmentation products. With 0.3 fissions/proton and 3 fragments fission plus 0.5-1 heavy nuclei/proton it would appear that 1.6 radioactive nuclei/proton in the target region might be a reasonable assumption. Thus an upper estimate is obtained as

$$\frac{E}{N} \leq \frac{1000/0.5 - 0}{(24.6 - 1.6)0.8} = 110 \text{ MeV/FP} \quad (\text{A36})$$

9. A.32

This value is not considered as an absolute bound but rather a practical bound. If this performance could not be attained the accelerator probably would not be built. Thus the energy required is estimated to be

$$\frac{23 \text{ MeV}}{\text{FP}} < \frac{E}{N} < 110 \text{ MeV/FP} \quad (\text{A37})$$

This required electrical energy must be less than the electrical energy produced in a fission power reactor per candidate transmutation fission product. This energy balance is strictly true only as long as electrical power is being obtained from fission reactors. The electrical energy produced per FP can be obtained from the production rates given on the Chart of the Nuclides. These values are estimated for a 1000 MW(e)-yr, slightly enriched uranium, 25,000 MWD/T fuel exposure with 85% duty cycle. The values given for candidate FP

Cs-137 41.6 Kg

Sr-90 18 Kg

Tc-99 27.5 Kg

correspond to the following values of electrical energy produced per FP nucleus:

Cs-137 900 MeV/FP

Sr-90 1370 MeV/FP

Cs-137+Sr-90 550 MeV/FP

Tc-99 985 MeV/FP

Cs-137+Sr-90+Tc-99 350 MeV/FP

Thus, the energy input to the ING required to transmute only these candidate FP seems to be clearly less than the electrical energy produced in their formation.

Quantity of FP Transmuted

The energy balance derived above can also be used to estimate the quantity of fission product which can be transmuted in this conceptual system. An ORNL study group proposed⁽¹³⁾ that 1000 MW of power to spallation accelerators would be required to handle the Sr-90 produced in 9000 MWe of LMFBR's. This conclusion is only in approximate agreement with the values derived here for LWR's. For the pessimistic assumption, i.e., 110 MeV/FP, and the transmutation of Cs-137 + Sr-90 + Tc-99 the ratio of spallation power to fission electrical power is one-third. A more optimistic assumption would be about half of that. From an economic viewpoint, however, a power ratio of 1/6 or 1/9 may still be prohibitive. The ING proposal was for an accelerator of only 65 MW proton beam power. Thus fifteen ING's would be required for 6-9 LWR's and the 1967 projected costs of an ING were \$182 M without conversion of thermal energy to electrical power.⁽¹⁸⁾ Thus projected large improvements in beam power in a single accelerator would appear to be necessary for economic feasibility.

Specific Transmutation Rate

It has been shown that the energy balance for transmutation of Cs-137, Sr-90 and Tc-99 in an ING spallation accelerator could be favorable. In order for a transmutation scheme to be attractive the induced transmutation rate must

significantly exceed the radioactive decay rate. These values are compared on Table 9.A.4 for the projected maximum thermal flux values of $10^{16} \text{ n-cm}^{-2}\text{-s}^{-1}$ of the ING. The reduction in half live for Cs-137 is seen to be decreased not very significantly even for the infinite dilute thermal flux value. In addition, if the effective flux is reduced to the order of $10^{15} \text{ n-cm}^{-2}\text{-s}^{-1}$ due to neutron self shielding in the FP sample or other factors the transmutation rate of Sr-90 becomes equally less attractive. Projected thermal flux values several times larger than $10^{16} \text{ n-cm}^{-2}\text{-sec}^{-1}$ would be required to make the spallation accelerator genuinely attractive for transmutation of Sr-90 and Cs-137 FP isotopes.

Radioactive Waste Balance

In the general discussion of the spallation accelerator some of the radioactive waste produced in the spallation of 1 GeV protons on Pb-Bi was identified. These included the production of nearby nuclei of the type:

<u>Nucleus</u>	<u>T_{1/2}</u>
Bi-208	$3.68 \times 10^5 \text{ yr}$
Bi-207	38 yr
Bi-206	6.24 d
Pb-205	$1.4 \times 10^7 \text{ yr}$
Pb-203	52 hr
Tl-204	3.78 yr
Tl-202	12.2 d

TABLE 9.A.4. Time For 99% Elimination in Thermal Fluxes With Frequent Rapid Daughter Product Removal

		<u>Time for 99% Elimination, yrs</u>		
		<u>Thermal Flux ($n\text{-cm}^{-2}\text{-sec}^{-1}$)</u>		
	<u>Thermal Capture (barns)</u>	<u>Decay Only</u>	<u>$\phi = 10^{15}$</u>	<u>$\phi = 10^{16}$</u>
Sr-90	1	191	83.	14.
Tc-99	22	1.40×10^6	6.6	0.66
Cs-137	0.11	199	170.	80.

Assumed cross sections in barns:

Sr-90 - 1

Tc-99 - 22

Cs-137 - .11

9. A.36

The production of nuclei of this type was estimated to sum to 0.5-1 per incident proton with only a small probability for the formation of a stable nucleus.

The fission plus fragmentation of the cascade nucleus was estimated to occur 0.3 times per incident proton at 1 GeV. Each such event produces about three product nuclei on the average, the probability of formation of a fragment nucleus of mass A being independent of A within a factor of several. Some specific radioactive fragments which have been identified are shown on Table 9.A.5 along with its yield, if known.

Since about one nucleus of this type will be formed per incident proton it is apparent that the short and moderate half life activity in the target environs will be orders of magnitude larger than the disintegration rates of Sr-90 and Cs-137. This observation would seem to rule out any consideration of the use of spallation accelerators for the transmutation of radioactive waste with half-life values less than a few years.

If 24.6 source neutrons are produced per proton and the utilization for fission product absorptions is 80% as previously assumed then 5 neutrons/proton are absorbed in Be, D, and structural material. These absorptions result in

In addition, tritium will be produced in appreciable quantity by fast neutron reactions

TABLE 9.A.5. Radioactive Product Nuclei Identified in the
Spallation of 1 GeV Protons Lead(16)

<u>Nucleus</u>	<u>Half-Life and Decay</u>	<u>Production Cross Section (mb)</u>
F-18	11.1s	0.04
Na-24	15 h	0.4
Mg-28	21 h $\rightarrow$ 2.25 m Al-28	0.08
P-32	14.3 d	0.09
Sr-91	9.48 h $\rightarrow$ 58.6 d Y-91	
Mo-99	66 h $\rightarrow$ 2×10^5 y Tc-99	2.5
Cd-115m	44.6 d	0.5
Cd-115	53.5 h	0.3
Ba-128	2.42 d $\rightarrow$ 3.8 m Cs-128	0.5
Ba-129	2.1 h $\rightarrow$ 32.3 h Cs-129	0.3
Ba-131	11.7 d $\rightarrow$ 9.69 d Cs-131	0.9
Ba-133m	38.9 h $\rightarrow$ 10.4 g Ba-133	

and

Thus it appears that some 6.6 radioactive nuclei with a wide range of half-lives may be former per 19.6 fission product transmutation.

Although detailed calculations do not exist to support these rather uncertain values, it does not appear that the projected reduction of about 3 to 1 in radioactive nuclei is worth detailed consideration.

Other Proposals - Steinberg⁽¹²⁾ has proposed the transmutation of fission products in a spallation accelerator of about ten times the beam power of the ING. This would improve the accelerator capital investment and the maximum thermal flux value. As the ORNL study group pointed out⁽¹³⁾ this type of accelerator is not feasible within the limits of current technology. In addition, increasing beam voltage or current has no effect on the considerations given the ING with regard to energy balance or radioactivity produced per proton.

Dr. Torao Ichimiya⁽¹⁴⁾ has proposed the use of Cs-137 target material for a ten BeV proton beam. This evades the problem of the low thermal neutron capture cross section of Cs-137 by obtaining additional transmutation from high energy n,2n reactions. The transmutation rate for this procedure cannot be deduced from physics information at hand, but Ichimiya and associates assume 100% utilization of secondaries to estimate 85 nuclei transmuted per

incident proton. At 100% accelerator efficiency, this would require almost 6 MeV per reactor fission to transmute the Cs-137 alone. Observed is: (1) this is energetically expensive, (2) the physics calculation is very uncertain, and (3) the radioactive waste production is certain to be large.

It is concluded that the spallation device is ineffective as a short term alternative in reducing the inventory in curies of radioactive materials. Its ultimate merit lies in eliminating materials whose hazard persists longer than that of human historical records. The objective is to trade the long term (10^6 year) hazards of the heavy element wastes for the shorter term (10^3 year) hazards of fission products.

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APPENDIX 9.B

TRANSMUTATION BY FISSION AND THERMONUCLEAR EXPLOSIVE DEVICES

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APPENDIX 9.B

TRANSMUTATION BY FISSION AND THERMONUCLEAR EXPLOSIVE DEVICES

SUMMARY

Because thermonuclear explosive devices are known to produce large yields of neutrons, the use of these neutrons for the transmutation of radioactive waste has been proposed⁽¹⁻³⁾. In a preliminary evaluation, PNL concluded that the concept did not appear to be technically feasible. A group of reviewers at Los Alamos Scientific Laboratory (LASL), however, reviewed the PNL study and strongly disagreed with its conclusions and concluded that the concept may be very attractive. The details of the LASL review are contained in Appendix 9.E. Because of the LASL conclusions, PNL has conducted a more extensive evaluation of this concept and this evaluation is reported in the Detailed Results of this Section. The PNL evaluation does not support the conclusions reached in the LASL review. The PNL evaluation differs from the LASL primarily in the following respects:

- The LASL estimation of the yield of fission products Tc-99, I-129, and Cs-137 is too low by a factor of about three.
- The LASL assumption that successive neutron captures in product isotopes does not occur does not appear to PNL to be physically justified. Successive neutron captures lowers the efficiency of utilization of source neutrons.
- Successive neutron captures further lowers the efficiency of the use of source neutrons due to the presence of other iodine and cesium isotopes.

9.B.2

The main conclusions of PNL of the feasibility of the concept are:

- The concept can be considered only for radioactive waste with half-lives much greater than the 12.3 year tritium because of the significant production of tritium residue in the device. Hence, the transmutation of short-lived radioactive waste is not technically feasible.
- The estimated cost of transmutation of elemental fission product technetium, iodine and cesium is almost twice the cost of the electricity produced in their production. Hence, this concept is not technically feasible.
- If isotopically separated I-129 and Cs-135 could be obtained, the technical feasibility would be within the range of uncertainty of the present analysis. More rigorous calculations of the neutron transmutation process would then be required to establish technical feasibility.
- The estimated cost of transmutation of Np, Am, and Cm waste is estimated to be less than 20% of the electrical cost and is, thus, apparently technically feasible. This concept is, however, judged to be much less attractive than that of actinide recycle in fission reactors.

DETAILED RESULTS

The thermonuclear explosive device is a potential candidate for neutron-induced transmutation of high-level radioactive waste in an underground explosion because it is a source of large numbers of available neutrons. Details of many aspects of thermonuclear devices are not available for public use because of classification for weapons purposes. However, the general features of such devices which are necessary to assess their

9.B.3

possible uses for waste transmutation appear to be publicly available. These features are given, for example, in a book by Teller⁽⁴⁾ and in many published papers given at the January 1970 Symposium on Engineering with Nuclear Explosives⁽⁵⁾. The important features of the device can be categorized as cost, radioactive waste produced and radioactive waste consumed. The characteristics of these features are summarized in the following discussions.

The thermonuclear explosive device consists of a small amount of fissionable material and the bulk of the material is that which is used to develop the (DT) fusion reaction. The quantity of fissile material in the explosive device apparently does not depend greatly on the explosive yield of the device. Most of the cost of the device is apparently due to the manufacturing process, rather than material costs. The costs of fabrication, emplacement, arming and firing the device appear to be in the range of \$0.5 million to \$0.6 million for devices in the range of 100 kiloton to 400 kiloton^(6,7) although Cohen, et al,⁽⁸⁾ used a value of \$1 million for a smaller yield device. The cost of drilling and casing emplacement holes has been taken^(6,7) to be \$33 per meter for holes 150 centimeters in diameter for depths up to 1524 meters, but Cohen, et al,⁽⁸⁾ used a cost of \$1 million for an emplacement hole 610 meters. The LASL review group proposed⁽⁹⁾ that a 100 kiloton device at a depth of 1500 meters was appropriate for the transmutation concept. These characteristics were apparently chosen in order that geographically adjacent shots be seismically decoupled. The LASL proposal estimated a total cost per shot of \$1 million assuming that the emplacement hole could be used several times. Heckman⁽⁷⁾ also proposed reuse of a hole

9.B.4

but it is apparently not a proven technology. Heckman also proposed a cost of \$200,000 for a neutron target of uranium and thorium. The LASL proposed cost apparently does not include the costs of preparing the high-level radioactive waste targets. For the purposes of this analysis, a cost of $\$1.5 \pm 0.5$ million will be assumed to cover target fabrication costs and their uncertainty and the uncertain possibility of reuse of the emplacement hole and its costs. This estimated cost figure implies that the transmutation explosions are done on a routine basis and at the Nevada Test Site since estimates of experimental shots at other locations are of the order of \$4 million.⁽¹⁰⁾

The radioactive waste created in a thermonuclear explosion results from fission products from the fission component of the device, from tritium which is created but not burned in the thermonuclear explosion, and from neutron-induced reactions in the device and in the surrounding earth medium. Estimates of these waste components are shown in Table 9.B.1 in units of moles of product. Also shown are the moles of neutrons created which are, in principle, available for transmutation. These yield estimates are primarily obtained from those of Evans and Kruger⁽⁶⁾. The neutron leakage to the surrounding media was calculated by Lessler⁽¹¹⁾ for a 100-kiloton device using a 150-centimeter diameter hole and boric acid shielding to inhibit neutron loss to the surrounding media. The values shown on Table 9.B.1 allow the following general conclusions.

- The fission product yield is a negligible fraction of the neutrons available for transmutation except for devices of very small explosive yield. The use of small yield devices is also ruled out on cost balance grounds.

9.B.5

- The radioactivity resulting from neutron leakage to the surrounding media can be made a negligible fraction of the neutrons available for transmutation.
- Since about 1 gram mole of 12.3 year tritium is produced per 4 moles of neutrons available for transmutation, the concept of thermonuclear explosives for transmutation is feasible only for radioactive waste with half-lives greater than some 50 years.

TABLE 9.B.1. Yield and Radioactive Products of Thermonuclear Explosives

	<u>Yield (gram moles)</u>
Neutrons	2.5 /kt
Fission Product	
$T_{1/2} = 15 - 50 \text{ yr}$	0.15 /device
$T_{1/2} > 50 \text{ yr}$	0.15 /device
Unburned Tritium ($T_{1/2} = 12.3 \text{ year}$)	0.67 /kt
Radioactive waste in surrounding media	<0.001/kt

To assess the feasibility of transmutation by thermonuclear explosives, it is necessary to consider the types and quantity of long-lived radioactive waste which are candidates for transmutation. Among the fission products these appear to be Tc-99($T_{1/2} = 2.1 \times 10^5 \text{ years}$), I-129($T_{1/2} = 1.6 \times 10^7 \text{ years}$) and Cs-135($T_{1/2} = 2.6 \times 10^6 \text{ years}$). The base case is taken to be the product of a 1000 MW(e) PWR for fuel processed 90 days after discharge. The calculated yields of this base case for these isotopes and their chemical elements in an equilibrium fuel cycle are shown on Table 9.B.2. The impact of stable and short-lived isotopes of the element is seen to be most important for cesium. In this case, the number of atoms which would have to be transmuted is increased

9.B.6

by almost an order of magnitude, and, because of stable Cs-133, even allowing 100 years of radioactive decay reduces the amount of material to be transmuted by less than a factor of two.

TABLE 9.B.2. Long-Lived Radioactive Waste Produced by Reference 1000 MW(e) PWR

<u>Element</u>	<u>Isotope</u>	<u>Yield (gram moles) at Cooling Time</u>	
		<u>1 yr.</u>	<u>100 yrs.</u>
Iodine	Total	72	72
	127	11	11
	129	61	61
Technetium	99	290	290
Cesium	Total	678	373
	133	260	260
	134	34.6	0
	135	82.2	82.2
	137	302.8	30.8
Total Fission Product		1040	735
Neptunium	237	70	(a)
Americium	Total	23	(a)
Curium	Total	5	(a)
Total Np, Am, Cm		98	

a. Not calculated

9.B.7

For neutron-induced transmutation of the fission-product nuclei, the assumption was made by the LASL review group that (successive) neutron capture events can be made the dominant process by proper design of the device and target configuration. There are not detailed calculations to support this assumption and it is necessary to point out that the requirements are different for the different fission product elements. For cesium, the multiple capture rate must be sufficiently high that essentially all of the Cs-133 and Cs-134 are transmuted beyond Cs-135. For I-129, the multiple capture rate in 127 and 129 must be adjusted so that no significant amount of mass 135 is produced. Whether either of these restrictions can be achieved in the face of competing $n,2n$ reactions needs more definitive analysis than has yet been made.

The need for multiple neutron capture in the chemical fission product elements iodine and cesium greatly reduces the number of thermonuclear neutrons which can effectively cause a transmutation of the desired isotope. Ignoring the complexity of competing $n,2n$ reactions, if each isotope has the same value of effective capture cross section, σ , and sees the same effective neutron flux, ϕ , the distribution of atomic masses, $A_0 + n$, after adding n neutrons to the initial A_0 nucleus is⁽¹²⁾

$$(A_0 + n) = A_0 \frac{r^n}{n!} e^{-r} \quad (B1)$$

where $r = \sigma \phi$ is the capture rate. Values of r in the range of 1 to 25 are clearly available from the device. For an r value of 5, 8.4% of the initial Cs-133 atoms would transmute to Cs-135 (or I-127 to I-129) and the average number of available neutrons used per transmutation is about 4.3. For an r value of 7 the percentage of $A_0 + 2$ transmutations is 2.2% and

9.B.8

the average number of neutrons used per transmutation is about 6.3. Thus, for a reasonable reduction in the radioactive waste only about 20% of the available source neutrons can be used for transmutation.

It is also clear from the rate equation (B1) that the neutron flux cannot decrease very much through the transmutation target. Reduction of the flux (ϕ) from a value of 7 to a value of 5 increased the production of the undesirable A_{0+2} isotope to a marginally feasible level. In order to keep the flux level to the required value in the transmutation target, the order of 1/2 of the source neutrons must escape to the device shield to be absorbed. Coupled with the estimate of the order of 5 neutrons absorbed for an effective transmutation, the overall efficiency of useage of source neutrons per transmutation appears to be no better than some 10% for FP iodine and cesium.

Also shown on Table 9.B.2 is the estimated production of the long-lived actinides neptunium, americium and curium. The transmutation concept for these elements is the reduction to fission product waste by successive captures and fission which occurs primarily at odd mass plus a neutron value. The effective reduction of neptunium to fission product differs from the iodine and cesium cases in that the average number of source neutrons required to cause a fission is probably larger than those required for I and Cs transmutation. However, the fission events in the actinide targets will cause source neutron multiplication so that the overall efficiency for transmutation may not be significantly different.

The efficiencies of utilization of source neutrons proposed here are quite different than those assumed by the LASL review group. The LASL group

9.B.9

assumed that only single neutron captures could be achieved with an overall utilization of 80 percent. Such a process cannot be achieved with eq.(B1) where each isotope has an equal neutron capture cross section. In order to achieve the LASL assumption, the neutron capture cross section of the $A_0 + 1$ isotope would have to be much less than that of the A_0 isotope. The opposite situation exists for Cs-133 and almost certainly for I-127.

The technical feasibility of transmutation by thermonuclear explosives requires a comparison of the cost of transmutation to the cost of the electricity produced in the production of the radioactive waste. For this comparison, the reference 1000 MW(e) PWR produces an assumed \$35 million worth of electricity per year based on 5 mills/kW-hr and a plant factor of 0.8. The neutron yield of a 100-kiloton device is 250 gram moles at an estimated cost of \$1.5 million. At the 10% efficiency estimated for transmutation, the cost of transmutation is estimated to be of the order of

$$\frac{\$1.5M}{25 \text{ moles}} \approx \$60,000 \text{ per gram mole of waste. (B2)}$$

The costs of transmutation of the various waste components of the reference reactor are then estimated as on Table 9.B.3 and expressed as a percentage of the value of the electricity produced in their production. Also shown is the annual number of explosions required to keep abreast of the production of one 1000 MW(e) PWR and the annual number of explosions required for a 150,000 MW(e) fission power reactor economy which is postulated by about 1980.

9.B.10

TABLE 9.B.3. Estimated Costs of Transmutation by Thermonuclear Explosives

Element	Annual Cost per PWR \$ Million	Percent of Electrical Power Costs	Annual No. of 100 kt Explosive Shots	
			per 1000 MW(e)	per 150,000 MW(e)
I + Cs	45	129%	26	3900
Tc	18	50%	11	1650
Np+Am+Cm	5.9	17%	3.5	525

From the estimated cost values given in Table 9.B.3, we conclude that transmutation of the elemental fission products iodine, cesium and technetium by thermonuclear explosives is not technically feasible. If it were possible to obtain isotopically separated I-129 and Cs-135 at not too great a cost, then the question of technical feasibility would fall within the uncertainty of the present analysis. The transmutation of the actinide waste, Np, Am, and Cm does appear to be technically feasible for the criteria of quantity, rate, and energy balance. The concept does not appear attractive, however, based on the large number of continuing explosions which would be required to keep abreast of even a static fission power reactor economy. The use of thermonuclear explosives to transmute the actinides to fission product is judged to be a much less desirable process than that of recycle in fission reactors.

The PNL analysis of estimated costs for transmutation of fission products by thermonuclear explosives differs greatly from that of the LASL review group who concluded the concept "may be very promising". The differences in the assumptions of the two analyses are shown on Table 9.B.4. The differences in estimation of cost per explosion plus the amount of fission product Tc-99, I-129 and Cs-135 amount to a factor of about five. The lar-

gest part of this difference is due to the fact that the LASL yield was erroneously estimated for a 1000 MW thermal reactor rather than the reference 1000 MW(e) PWR for which the PNL yields are based on ORIGIN calculations. The largest difference in the estimations is due to the LASL assumption of only single neutron captures. According to the PNL analysis, an average of about four successive captures is a necessary consequence of efficient removal of only a single isotope. This conclusion lowers the efficiency further due to the presence of other fission product iodine and cesium isotopes.

TABLE 9.B.4. Comparison of PNL and LASL Estimations of Transmutation of Fission Product by Thermonuclear Explosive Devices

<u>Element</u>	<u>LASL</u>	<u>PNL</u>	<u>Comment</u>
Cost/Explosion	\$1 Million	\$1.5 million	The PNL estimate contains costs of target fabrication apparently not included in the LASL estimate.
<u>Transmutations</u>			
Explosion	200 gram moles	25 gram moles	LASL assumed no multiple neutron capture. The PNL analysis indicates that multiple neutron captures are necessary for efficient transmutation of even separated isotopes.
<u>Moles of Fission Product</u>			
PWR	130 gram moles	1040 gram moles	LASL neglected the I-127 and the fission product Cs other than Cs-135. This assumption involves the assumption of single neutron captures which the present analysis indicates is not feasible. The LASL calculation of fission product yield was also erroneously based on 1000 MW(th) rather than 1000 MW(e).

Dr. Edward Teller, Associate Director of Physics of Lawrence Livermore Laboratory, was also questioned⁽¹³⁾ by PNL on the results of any studies by LLL on the thermonuclear explosives transmutation concept. Dr. Teller's reply⁽¹⁴⁾ did not indicate that the concept had received technical evaluation at LLL but he characterized the concept as "complicated and expensive" and less attractive than burial in salt domes or in chimneys of silicate rock created by nuclear explosives.

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APPENDIX 9.C

TRANSMUTATION BY FISSION REACTORS

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APPENDIX 9.C

TRANSMUTATION BY FISSION REACTORS

SUMMARY

Fission reactors produce neutrons which, in principle, can be used to transmute waste. The earliest proposal for the use of fission reactor neutrons to transmute waste was made by Steinberg et al.⁽¹⁾ In this proposal, the fission products krypton-85, strontium-90, and cesium-137 were considered candidates for transmutation in a special purpose high flux reactor. Studies conducted at Oak Ridge National Laboratory (ORNL) reported by Claiborne⁽²⁾ considered neutron-induced transmutation of fission products and actinide elements. Claiborne's work on transmutation of actinides in commercial LWRs is the most extensive study of the subject to date. Calculations of actinide recycle in a commercial PWR were made at PNL to compare results with Claiborne's study and extend the analysis to include other strategies. In reviewing the work of others and the PNL studies, it is concluded:

1. Transmutation of fission products in fission reactors is not technically feasible because it fails to meet the waste inventory and transmutation rate criteria.

2. Transmutation of actinides in fission reactors is technically feasible. Reductions of approximately two orders of magnitude in the long-term hazard potential of actinides in high-level waste can be realized by recycling actinides in fission reactors. The transmutation can effectively be accommodated in either uranium fuels (UO_2) or plutonium fuels ($\text{UO}_2\text{-PuO}_2$), and perhaps even in specially fabricated target rods selectively located in the reactors. Though existing calculational studies have been made only for a commercial pressurized water reactor (PWR), perhaps even better transmutation results are expected for other fission reactors such as High Temperature Gas Cooled Reactors (HTGRs), and Fast Breeder Reactors (FBRs).

DETAILED RESULTS

Fission reactors can, at least in principle, be applied in fission product transmutation and actinide transmutation. The technical feasibility of each is described below.

A. Fission Products

Steinberg, Wotzak, and Manowitz, made the initial study⁽¹⁾ of fission product transmutation in fission reactors. Their study was concerned with the possibility of transmutation of the fission products Kr-85, Sr-90 and Cs-137. In their analysis, they considered the competition for neutrons of other fission product isotopes of the same FP chemical elements in order to determine if isotopic separation was required. In this analysis, they used thermal

neutron cross sections. Hence, their study implicitly assumed that the transmutation reactor was a thermal reactor. They concluded that isotopic separation was necessary for Kr-85 and Cs-137 but not for Sr-90. The enrichment level which they proposed for Kr-85 (90%) is no longer valid because they assumed that the neutron capture cross section of Kr-85 was 15 barns. It is now known that the value is about 1.7 barns (see Table 9.C.1) so that much greater enrichment is required than they proposed. Their calculated transmutation rates for Kr-85 are also an order of magnitude too large because of their assumed cross section.

The transmutation in thermal fission power reactors of troublesome fission product isotopes with small values of thermal capture (transmutation) cross sections is not possible because of the low values of thermal neutron flux in such reactors. Calculated values of transmutation rates of several such isotopes are shown on Table 9.C.1 for various neutron flux levels. These results show that, with the exception of I-129, the transmutation rates differ insignificantly from the radioactive decay rates for neutron flux levels of 10^{14} neutrons/(cm²)(sec). The neutron flux levels in thermal fission power reactors are the order of 3×10^{13} neutrons/(cm²)(sec). Thus, the transmutation of low cross-section isotopes in thermal power reactors does not satisfy the criterion of specific transmutation rate.

Steinberg et al.⁽¹⁾ specifically proposed the use of special high-flux thermal reactors, such as a flux trap reactor, for transmutation of fission products. Such reactors achieve large values of thermal neutron flux, of the order of 10^{16} neutrons/(cm²)(sec), by thermalizing leakage neutrons in a medium which essentially does not absorb neutrons. Such a thermal neutron flux level gives specific transmutation rates which would be marginally feasible for some isotopes as shown on Table 9.C.1. However, this neutron flux level cannot be maintained in the presence of absorbing material. At best only a few percent of the fission neutrons could be utilized to produce an interesting specific transmutation rate. In addition, an equal or greater number of fission product isotopes of high-level waste would be formed in the fission process per transmutation event. Thus, this reactor concept does not satisfy the criteria of overall waste balance and of total transmutation rate.

Steinberg et al.⁽¹⁾ did not consider the use of a fast breeder reactor for transmutation of fission product isotopes although LMFBRs were considered in studies at ORNL.⁽²⁾ The consideration of the use of LMFBRs for fission product transmutation arises primarily from the excess neutrons per fission which are destined for the breeding of fissile material. In practical LMFBR designs the excess fraction of neutrons per fission is in the range 0.15 to 0.3. These values are approximately the same as the fraction per fission of fission product constituent of high-level waste which the LMFBR would produce during operation. Thus, the LMFBR could transmute fission product at only about a break even rate on overall waste balance and would do so at the expense of being no longer a viable breeder of fissile material. In addition, the projected fast

TABLE 9.C.1. Properties of Several Important Fission Product Nuclides and Time Required for 99.9% Reduction of Their Inventory by Decay and Neutron Transmutation

(Taken from Reference 2)

Nuclide	Sr-90	Cs-137	Kr-85	H-3	I-129
Half-life, years	28.9	30.2	10.74	12.33	1.6×10^7
Burnout cross section, barns ^(a)	1.2	0.17	1.8	nil	35
Curies/metric ton in spent fuel ^(b)	77,600	108,000	11,400	708	0.0367
Relative hazard in spent fuel ^(c)					
m ³ air at RCG/metric ton	2.6×10^{15}	2.1×10^{14}	3.8×10^{10}	3.5×10^9	1.8×10^9
m ³ water at RCG/metric ton	2.6×10^{11}	5.4×10^9	--	2.3	6.1×10^5
Time required for 99.9% decay and burnout, years ^(d)					
Decay only	288	302	107	123	1.6×10^8
$\phi = 10^{14}$ n/cm ² ·sec ^(e)	249	295	106	123	63
$\phi = 10^{15}$ n/cm ² ·sec ^(e)	112	245	98	123	6.3
$\phi = 10^{16}$ n/cm ² ·sec ^(e)	17	91	57	123	0.63
$\phi = 10^{17}$ n/cm ² ·sec ^(e)	1.6	12	11	123	0.06

- Effective thermal cross section in typical spectrum of a PWR having average thermal flux of 2.91×10^{13} n/cm²·sec.
- Per metric ton of uranium charged to a PWR having average specific power of 30 MW/metric ton and burnup of 33,000 MWd/metric ton.
- Volume of air and water potentially contaminated to RCG (10 CFR 20) by the content of a metric ton of spent fuel.
- Indicated times are doubled and tripled for reduction of inventory by factors of 10^6 and 10^9 , respectively.
- Average thermal flux assuming spectrum typical of that in a PWR.

neutron flux levels of about 10^{15} n/cm²/sec do not allow the attainment of a sufficiently high specific transmutation rate. Thus, the use of an LMFBFR to transmute the fission product component of high-level waste is not a feasible concept.

Claiborne's report⁽²⁾ gives results of work which have been conducted at ORNL on fission product transmutation. Tables 9.C.1 and 9.C.2 are taken from this report. The data in Table 9.C.1 illustrates the points made in the above arguments concerning the requirements of high flux and high cross section for effective transmutation. Flux levels of the order of 10^{17} neutron/(cm²)(sec) are needed to accomplish reasonable transmutation rates. The data in Table 9.C.2 are referred to in Claiborne's report as results of work conducted at ORNL by Nichols and Blomeke in which they studied the effect of neutron transmutation schemes on radioisotopic inventory and electrical power costs. The rate of decay is essentially unchanged in recycling Sr-90 in either PWRs or LMFBFRs. Thus the inventory is unchanged. The high flux reactor was

TABLE 9.C.2. The Inventory of ^{90}Sr and Estimated Costs Associated With the Steady-State Production of Electric Power by Various Schemes of Transmutation

(Taken from Reference 2)

	^{90}Sr Inventory (megacuries) per 1000 MW(e) of Capacity with Plant Factor of 0.8			Estimated Incremental Cost ^(a) [mills/kWhr(e)]
	In Reactors	Outside Reactors	Total	
1. PWR - conventional operation ^(b)	3.17	88.0	91.2	0
2. PWR with complete ^{90}Sr recycle ^(b)	64.6	21.5	86.1	0.1
3. LMFBF - conventional operation ^(c)	1.39	38.6	39.9	0
4. LMFBF with complete ^{90}Sr recycle ^(c)	28.9	9.6	38.5	0.1
5. High Flux Isotope Reactor - conventional ^(d)	0.11	132.	132.	24
6. High Flux Isotope Reactor - ^{90}Sr recycle ^(d)	38.3	12.7	51.0	25
7. 98% of power from LMFBFs plus ^(c) 2% of power from fusion burner	1.36 0.93	0.91 0.31	3.51	0
8. 90% of power from LMFBFs plus ^(e) 10% of power from spallation	1.54 0.16	1.03 0.51	3.23	0.8

a. In excess of the typical power generation cost of approximately 7 mills/kWhr(e).

b. Assumes thermal efficiency of 32.5%.

c. Assumes thermal efficiency of 41.7%.

d. Assumes thermal efficiency of 30%.

e. One 1000-MW(e) spallation burner reactor associated with nine 1000-MW(e) LMFBFs. The electricity generated by the burner reactor is used internally to generate a 500-MW beam of 10-BeV protons.

assumed to produce a minimum thermal flux value of about 2×10^{15} neutrons/(cm²)(sec).⁽²⁾ At this flux level, the inventory is reduced by roughly 60%. Claiborne concluded that "this type of reactor would not be an economical source of electric power, however, because of its small size, high refueling cost, and high neutron leakage." Lines 7 and 8 to Table 9.C.2 are strategies combining LMFBFs with CTRs, and LMFBFs with spallation accelerators.

In summary, it is improbable that transmutation of fission products in fission reactors could meet any of the technical feasibility requirements for the production of stable daughters. An independent evaluation of the feasibility of fission product transmutation was also made by Kubo.⁽³⁾ He concluded that "fission products are not conducive to nuclear transmutation as a general solution to long-term waste management."

B. Actinides

Actinide transmutation with LWRs appears to be quite attractive based on the preliminary analyses made to date. Claiborne's study⁽²⁾ has produced the most extensive findings on this subject to date. Kubo⁽³⁾ and Kubo and Rose⁽⁴⁾

at MIT have studied Claiborne's results and similarly have concluded that actinide recycle in fission reactors is not only technically feasible but an attractive waste management concept. Their extension to Claiborne's work has been primarily devoted to analyses of the further reduction in potential toxicity which would accrue from improved efficiency for removal of actinides higher than plutonium from the high-level waste. The principal findings of Claiborne's study are reviewed here. The extension of Claiborne's analysis made by Kubo is briefly summarized. The results of calculations made at PNL confirming Claiborne's conclusions and extending the concept to include other potential strategies are also presented.

Review of Claiborne's Work - The basis of Claiborne's study is depicted by the system flowsheet shown on Figure 9.C.1. In this system it is assumed that reprocessing of the fuel takes place 150 days after reactor discharge. The actinide elements present at that time are then recycled into fresh fuel. The recycle actinide material was simply added uniformly to every rod of a 3.3% enriched UO_2 fuel loading for a PWR. The U and Pu are kept out of the recycle stream.

ORNL-DWG 72-1436
(Taken from Ref. 2)

FIGURE 9.C.1. Flowsheet for Actinide Recycling

The effect of recycle on the inventory of actinides is seen by examination of Table 9.C.3, which lists the amounts of the various actinide isotopes found in a ton of burned fuel. (One recycle covers three years of reactor operation, with one-third of a PWR core discharged each year.) The inventory of most of the heavy elements (e.g., Np, Am, Cm) is seen to increase to saturation in about five recycles. Since at saturation these isotopes are transmuted at 99.5% of the rate at which they are produced, this represents a substantial reduction of the inventory of toxic species of actinide waste. To put these results in perspective Claiborne used a hazard^(a) index defined as the amount (volume) of air or water required to dilute for release to the environment the residual waste, following reprocessing and removal of materials to be recycled. This index was calculated for dilution of each nuclide to its Radiation Concentration Guide Value (RCG). He then defined a hazard measure (or hazard reduction factor) which is the ratio of two indices. The index in the numerator is for waste from fresh fuel, burned and reprocessed, with none of the actinides removed. The index in the denominator is for waste from fuel into which actinides have been recycled, burned and reprocessed, with 99.5% of the actinides removed for further recycling.

Table 9.C.4 presents this hazard measure as a function of the number of recycles and the time of storage prior to dilution and release. For a given storage time the numerator index is a constant and the denominator index represents the hazard associated with 0.5% of the actinide content of the fuel listed in Table 9.C.3. Thus, the hazard measure decreases as the actinide content of the fuel increases with multiple recycles. The changing isotopic content of the fuel is reflected in the changing time behavior of the hazard reduction factor with recycling. Thus, rapid saturation under recycle of the fuel content of long-lived isotopes seen in Table 9.C.3 is reflected in Table 9.C.4 in the hazard reduction factor for release after 10^6 years. The increase with recycle of short-lived isotope concentrations is reflected in the hazard reduction factor for release in the short term.

The cumulative hazard of the actinide waste from operation of a PWR for 60 years, for cases with and without recycle, is plotted in Figure 9.C.2 for the short term and in Figure 9.C.3 for the long term.

Claiborne studied the effect of extraction efficiency on the actinide recycle. Hazard reduction factors for the case assuming a 99.9% extraction efficiency are shown in Table 9.C.5 for comparison with the results for the case with 99.5% efficiency presented in Table 9.C.4. The hazard reduction factors are increased

a. We prefer to use toxicity rather than hazard since its connotation is different. However, in this section, the word hazard will be used since it was used by Claiborne in his work and because we are taking data directly from his report.

TABLE 9.C.3. Effect of Recycle^(a) on the Quantity
of Each Actinide Discharged

(Taken from Reference 2)

Recycle No.	Weight, g/metric ton of fuel										
	Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es
0	1.75×10^{-8}	1.35×10^{-3}	5.34×10^{-4}	9.56×10^5	4.82×10^2	8.97×10^3	1.37×10^2	3.95×10^1	1.56×10^{-6}	1.49×10^{-6}	4.42×10^{-12}
1	7.67×10^{-8}	2.04×10^{-3}	1.19×10^{-3}	9.56×10^5	6.56×10^2	9.21×10^3	1.57×10^2	1.16×10^2	3.60×10^{-4}	5.00×10^{-4}	4.74×10^{-9}
2	1.67×10^{-7}	2.45×10^{-3}	1.65×10^{-3}	9.56×10^5	7.19×10^2	9.29×10^3	1.61×10^2	1.68×10^2	3.10×10^{-3}	5.28×10^{-3}	8.09×10^{-8}
3	2.71×10^{-7}	2.73×10^{-3}	1.92×10^{-3}	9.56×10^5	7.42×10^2	9.32×10^3	1.61×10^2	2.01×10^2	1.04×10^{-2}	1.98×10^{-2}	3.71×10^{-7}
4	3.77×10^{-7}	2.96×10^{-3}	2.07×10^{-3}	9.56×10^5	7.50×10^2	9.33×10^3	1.61×10^2	2.23×10^2	2.32×10^{-2}	4.68×10^{-2}	9.85×10^{-7}
5	4.79×10^{-7}	3.15×10^{-3}	2.15×10^{-3}	9.56×10^5	7.53×10^2	9.33×10^3	1.61×10^2	2.38×10^2	4.10×10^{-2}	8.63×10^{-2}	1.95×10^{-6}
10	8.69×10^{-7}	3.86×10^{-3}	2.22×10^{-3}	9.56×10^5	7.55×10^2	9.34×10^3	1.61×10^2	2.87×10^2	1.73×10^{-1}	3.97×10^{-1}	1.02×10^{-5}
15	1.09×10^{-6}	4.29×10^{-3}	2.22×10^{-3}	9.56×10^5	7.55×10^2	9.34×10^3	1.61×10^2	3.15×10^2	3.10×10^{-1}	7.32×10^{-1}	1.94×10^{-5}
20	1.22×10^{-6}	4.57×10^{-3}	2.22×10^{-3}	9.56×10^5	7.55×10^2	9.34×10^3	1.61×10^2	3.32×10^2	4.18×10^{-1}	9.96×10^{-1}	2.68×10^{-5}
30	1.36×10^{-6}	4.84×10^{-3}	2.22×10^{-3}	9.56×10^5	7.55×10^2	9.33×10^3	1.61×10^2	3.48×10^2	5.40×10^{-1}	1.30	3.54×10^{-5}
40	1.37×10^{-6}	4.96×10^{-3}	2.22×10^{-3}	9.56×10^5	7.55×10^2	9.33×10^3	1.61×10^2	3.53×10^2	5.88×10^{-1}	1.42	3.87×10^{-5}
50	1.37×10^{-6}	5.01×10^{-3}	2.22×10^{-3}	9.56×10^5	7.55×10^2	9.33×10^3	1.61×10^2	3.56×10^2	6.08×10^{-1}	1.47	4.01×10^{-5}
60	1.38×10^{-6}	5.02×10^{-3}	2.22×10^{-3}	9.56×10^5	7.55×10^2	9.33×10^3	1.61×10^2	3.56×10^2	6.12×10^{-1}	1.48	4.04×10^{-5}

a. Based on recycling of 99.5% of actinides other than U and Pu and cooling 150 days before processing.

TABLE 9.C.4. Effect of Recycle of 99.5% of Actinides Other than U and Pu on Hazard Measure of Waste^(a) from PWR Spent-Fuel Processing

Recycle No.	Water Required for Dilution to RCG, ^(b) Ratio of Standard to Recycle Case for Decay Times (years) of:						
	10	10 ²	10 ³	10 ⁴	5 x 10 ⁴	10 ⁵	10 ⁶
0	40.4	12.3	15.3	18.5	22.8	27.9	52.3
1	22.5	9.30	12.4	13.4	16.0	19.7	45.7
2	19.3	8.20	10.0	10.8	14.5	18.0	43.6
3	17.5	7.57	8.43	9.29	14.2	17.4	42.8
4	16.5	7.15	7.35	8.25	14.0	17.1	42.5
5	15.8	6.77	6.57	7.53	14.0	17.0	42.5
10	13.4	5.76	4.72	5.75	13.9	17.0	42.5
15	12.1	5.32	4.16	5.53	13.8	17.0	42.5
20	11.4	5.08	3.78	4.89	13.8	17.0	42.5
25	11.0	4.95	3.63	4.73	13.8	17.0	42.5
30	10.7	4.89	3.56	4.63	13.6	17.0	42.5
40	10.5	4.83	3.49	4.55	13.6	16.9	42.5
50	10.3	4.80	3.46	4.39	13.6	16.9	42.5
60	10.3	4.80	3.46	4.39	13.6	16.8	42.5
Eff., %	25.5	39.0	22.6	23.7	59.6	60.2	81.5

a. 0.5% Pu and U sent to waste.

b. The recommended default RCGs in the Code of Federal Regulations (ref. 8) were used for unlisted nuclides.

by approximately five times due to the increased extraction efficiency. (In the 99.9% case only one-fifth of the fuel and actinide waste release of the 99.5% case takes place.) Clearly, obtaining higher partitioning factors (e.g., 99.99%), would proportionately increase the hazard reduction factors. In addition, the increase in partitioning factor also results in more cycles before the asymptotic reduction factor is reached as indicated by the line drawn between the values in Table 9.C.5.

Thus, these results show that the potential hazard can be reduced by one or two orders of magnitude by the recycle of actinides in fission reactors. Improving the technology of extracting actinides from the waste stream has substantial payoff in terms of further reducing the potential hazards.

Kubo's Analysis - Chapter 13 of Kubo's thesis⁽³⁾ is devoted to a review and extension of Claiborne's work on actinide recycle. Kubo illustrated, in Figure 13.6 of his thesis, the relationship between relative waste toxicity and the efficiency of extracting nuclides in fuel reprocessing (partitioning). This curve is reproduced here as Figure 9.C.4. As shown, for a constant stripping fraction, the toxicity increases with recycle because the discharged

FIGURE 9.C.2. Short-Term Cumulative Hazard of Actinide Waste from 60-Year Operation of a Typical PWR

FIGURE 9.C.3. Long-Term Cumulative Hazard of Actinide Waste from 60-Year Operation of a Typical PWR

FIGURE 9.C.4. LWR Waste Toxicity for Differing Extraction of Actinides with Recycle Equilibrium Fuel Cycle Assumed
3.3 w/o U-235, Burnup = 33,000 MWDT/MT
(Taken from Reference 3)

TABLE 9.C.5 Effect of Recycle of 99.9% of Actinides Other than U and Pu on Hazard Measure of Waste^(a) from PWR Spent-Fuel Processing

Recycle No.	Water Required for Dilution to RCG, ^(b) Ratio of Standard to Recycle Case for Decay Times (years) of:						
	10	10 ²	10 ³	10 ⁴	5 x 10 ⁴	10 ⁵	10 ⁶
0	199	57.5	73.1	88.8	110	137	256
1	116	43.7	58.9	64.2	77.7	95.9	224
2	94.8	38.4	47.7	51.6	70.8	87.3	213
3	85.8	35.5	40.1	44.2	68.4	84.4	210
4	80.7	33.4	34.8	39.3	67.5	83.4	209
5	77.2	31.7	31.1	35.8	67.2	83.2	208
10	65.7	27.0	22.1	27.0	66.9	82.7	207
15	58.7	24.7	19.1	23.7	66.4	82.3	206
20	54.5	-	17.6	22.2	66.1	82.1	206
25	52.0	-	16.8	21.4	66.0	82.1	206
30	50.6	-	16.5	20.9	65.8	82.1	206
Eff.,%	25.4	-	22.6	23.5	59.8	59.9	80.5

a. 0.1% of all actinides sent to waste.

b. The recommended default RCGs in the Code of Federal Regulations (ref. 8) were used for unlisted nuclides.

quantity of actinides increases (as illustrated in Table 9.C.3). However, as Kubo points out, "if actinide recycle at 99.99w/o extraction is accomplished, the relative toxicity after 1000 years would closely approach unity, the level of 'non-toxic' wastes," as he defines these in Chapter 6 of his thesis. He concludes that, "it appears that long-term reduction of actinide toxicity may be accomplished using recycle in a LWR." However, he states that a fast neutron spectrum such as in an LMFBF, would be a better device for actinide burnup.

PNL Studies - Survey calculations were repeated for the strategy used in Claiborne's study. In addition, a case where the same actinides were recycled in every tenth UO_2 rod was investigated. Finally, recycle of these actinides in a plutonium recycle fuel (UO_2 - PuO_2) was also studied. The calculations were made using the ALTHAEA code.⁽⁵⁾ This code communicates with the QUICK economics code⁽⁶⁾ which was used to calculate the incremental enrichment fuel cycle costs given in Section 9.3.

The calculated amount of each actinide element during recycle for the strategy of recycling in every UO_2 rod is presented in Table 9.C.6 along with the values obtained by Claiborne. The comparison shows the PNL results to be consistently larger than those of Claiborne's study. Of the numerous possibilities existing to cause differences in calculated values, the most logical is that the cross sections that were used in the calculations were different. In addition, the

TABLE 9.C.6. Comparison of Actinide Quantities in Discharged Fuel at 33,000 MWD/MTM

Recycle Number	Element Weight, g/metric ton of fuel									
	U		Np		Pu		Am		Cm	
	A*	B**	A	B	A	B	A	B	A	B***
0	9.56×10^5	9.51×10^5	482	521	8970	9325	137	144	39.5	32.0
1	9.56×10^5	9.50×10^5	656	784	9210	9592	157	176	116	92.0
2	9.56×10^5	9.50×10^5	719	921	9290	9719	161	183	168	135
3	9.56×10^5	9.50×10^5	742	993	9320	9779	161	184	201	161
4	9.56×10^5	9.50×10^5	750	1031	9330	9811	161	184	223	176
5	9.56×10^5	9.50×10^5	753	1052	9330	9827	161	184	238	184
10	9.56×10^5	9.50×10^5	755	1073	9340	9844	161	184	287	193

* A is Claiborne's Results (Table 20 of reference 2).

** B is this study.

*** Does not contain curium isotopes heavier than 245.

analysis made by Claiborne may not have included neutron spatial self shielding effects, since he used the ORIGEN code⁽⁷⁾ which does not have provisions for this. The comparison of neutron multiplication values shown on Figure 9.C.5 tends to bear this latter point out. The multiplication values obtained by Claiborne are consistently lower, but with the same trend as those obtained in the PNL study. Since spatial self shielding in fertile nuclides is a major effect in the neutron multiplication, the larger concentrations of these isotopes in Claiborne's analyses would lead to lower multiplication values. The differences in concentrations indicate that the reductions in toxicity values due to transmutation of actinides from Claiborne's study might be overestimated. Nevertheless, the difference is not so large that technical feasibility is jeopardized.

The mixture of isotopes for those elements given in Table 9.C.6 will present some fabrication problems due to decay heat, gamma dose rate and neutron dose rate. Thus, additional incremental cost for fabrication can be expected for the production of UO_2 fuel rods bearing these actinides. It might, therefore, be more profitable to recycle the actinides in a small fraction of the fuel so that only this fraction is subjected to the fabrication penalty.

A second series of calculations assumed recycle of the actinides in ten percent of the fuel rods. The higher concentration caused additional self-shielding of the resonances, requiring a larger inventory of each isotope before the burnout rate equaled the production rate. Calculations were made assuming that the actinides were recycled in every tenth UO_2 rod rather than

FIGURE 9.C.5. Comparison of Neutron Multiplication Values

in every rod. The calculated isotopics for Np, Am and Cm for this strategy are presented in Table 9.C.7 along with the values calculated for the other strategy. Comparison of the data in Parts A and B of this table shows that the actinide inventory does not get reduced as much by the recycle of actinides concentrated in a few rods as in the recycle in every UO_2 rod. After the ninth recycle, the Np-237 content is about 28% greater for the concentrated case. The Am-243 and the Cm-244 contents are likewise higher by ~13 and ~27%, respectively. Thus, concentration of actinides in 10% of the rods leads to a larger actinide inventory and reduces the transmutation rate. Nevertheless, the technical feasibility criteria are met by this strategy.

The increase in uranium enrichment required to achieve the same energy output as for 3.3 wt% UO_2 fuel without actinides is shown in Figure 9.C.6 for

TABLE 9.C.7. Comparison of Actinide Inventories for Two Recycle Strategies

Part A - Actinides Recycled in All Rods

<u>Actinide Inventory (Gms/MT of Heavy Metal)</u>								
Recycle No.	NP237	AM241	AM242	AM243	CM242	CM243	CM244	CM245
0	521.28	64.61	.58	78.63	7.89	.12	22.90	1.07
1	783.92	76.41	.77	100.41	9.95	.28	76.66	5.75
2	921.49	77.51	.79	104.67	10.15	.33	116.20	9.63
3	993.84	77.81	.80	105.12	10.15	.34	139.81	12.02
4	1031.99	77.95	.80	104.94	10.15	.34	152.96	13.37
5	1052.15	78.03	.80	104.72	10.15	.34	160.08	14.10
6	1062.81	78.06	.80	104.58	10.14	.34	163.88	14.49
7	1068.45	78.08	.80	104.49	10.14	.34	165.90	14.70
8	1071.44	78.10	.80	104.44	10.14	.34	166.97	14.81
9	1073.02	78.10	.80	104.42	10.14	.34	167.53	14.87

Part B - Actinides Recycled in One Rod in Ten

<u>Actinide Inventory (Gms/MT of Heavy Metal)</u>								
Recycle No.	NP237	AM241	AM242	AM243	CM242	CM243	CM244	CM245
0	521.28	64.61	.58	78.63	7.89	.12	22.90	1.07
1	811.78	73.37	.71	103.43	9.68	.27	76.55	5.96
2	994.46	74.98	.75	111.98	9.90	.32	120.63	10.44
3	1118.27	75.85	.78	115.43	9.98	.34	152.73	13.75
4	1192.93	76.31	.79	116.81	10.01	.35	175.55	16.05
5	1253.06	76.68	.80	117.66	10.04	.35	193.78	17.88
6	1299.96	76.96	.81	118.01	10.06	.36	203.55	18.95
7	1334.71	77.15	.81	118.16	10.07	.36	208.71	19.57
8	1360.02	77.27	.82	118.22	10.08	.36	211.28	19.91
9	1379.70	77.35	.82	118.26	10.09	.36	212.44	20.10

both strategies. Present PWR fuel management practices are to discharge one-third of the core each year. Thus, at the end of a cycle each 1/3 fraction of fuel has exposures of 33,000 MWD/MT, 22,000 MWD/MT, and 11,000 MWD/MT. The core reactivity roughly equivalent to the value when the core has an average exposure of about 22,000 MWD/MT. Thus, the neutron multiplication value obtained for UO_2 fuel (no recycle) at 22,000 MWD/MT ($k_{\infty} = 1.026$) was used as a target for setting the enrichment of the recycle fuel. To achieve the same cycle length, the enrichment for recycle in every rod must be increased from 3.3 to about 3.43 wt% U-235 for equilibrium. (The rods containing the actinides were enriched increasingly in each recycle to achieve the desired cycle length.) This incremental enrichment was used for calculating the fuel cycle cost penalty which is around \$14 million dollars per year (see Section 9.5.2). Recycling actinides in ten percent of the rods also requires an increase in

FIGURE 9.C.6. Enrichment Requirements for Actinides Recycle

core average enrichment. The added enrichment represents an energy balance factor, namely, the amount of energy expended in the enrichment process to obtain the higher U-235 content. Although these added factors were not calculated, increases of the energy input for these incremental separative work requirements do not appear to jeopardize the energy balance criterion.

Survey calculations were performed to make a preliminary assessment of recycling actinides in the context of a self-generation plutonium recycle strategy for PWRs. Self-generation plutonium recycle means that approximately one-fourth of the fuel in the core is mixed-oxide ($\text{UO}_2\text{-PuO}_2$). Either one quarter of all fuel assemblies in the core are mixed oxide or one fourth of the rods in an assembly are mixed oxide.

The scheme used here was to assume three quarters of the assembly contained enriched UO_2 rods at 3.3 wt% U-235 and one fourth contained mixed oxide. The

enrichment of UO_2 - PuO_2 rods necessary to achieve equal cycle length was calculated to be 3.75 wt% fissile (i.e., U-235, Pu-239 and Pu-241) which yields an assembly average enrichment of 3.41 wt% fissile. Recycling actinides in these mixed-oxide rods requires increasing the plutonium content in the mixed-oxide rods to 4.43 wt% fissile to achieve equal cycle length.

An alternative loading would be to have two types of mixed-oxide fuels in each assembly if the radiation level of actinide-bearing fuel is such that costs could be reduced by manufacturing a lesser number of these rods. For this alternative, most of the mixed-oxide rods would be the normal Pu recycle rods with 3.75 wt% fissile material. The remainder, which would be of special (remote or semi-remote) manufacture, would contain Pu and actinides with 5 wt% of more fissile material. When the fuel from the first recycle is reprocessed, Pu and actinides are extracted separately from the enriched U rods and from the mixed-oxide rods. It is assumed that the Pu from the enriched U rods is mixed with natural UO_2 and fabricated into 3.75 wt% fissile mixed-oxide fuel. The actinides from the enriched U rods and the Pu plus actinides from the mixed-oxide rods are mixed into Pu-actinide-natural U mixed-oxide rods. The radiation level of these rods would probably dictate remote or semi-remote manufacture. Calculations indicated that at 6 wt% fissile, the desired exposure of 33,000 MWD/T could not be obtained for the actinide recycle rods. Only a trivial improvement was calculated for an increase in fissile content to 7 wt%. The necessary increase in reactivity could be obtained by mixing the Pu and other actinides with enriched U. However, this results in calculated power densities in this fuel which were deemed too high for PWR operation. A solution to this problem of high power density might be to dilute the mixed-oxide fuel with a relatively inert material such as MgO .

This line of thinking goes as follows. The Pu-actinide-natural U mixed-oxide rods are conceived as follows. The fissile plutonium concentration was set at 0.438 gm/cc (which is equivalent to 5% enrichment in normal fuel). The results of calculations indicate that natural uranium at 0.955 gm/cc will yield sufficient reactivity to permit a burnup of 33,000 MWD from 115,290 cubic centimeters of fuel (the volume of one metric ton of fuel in the reference cycle). The remainder of the fuel volume would be filled with some inert material such as MgO .

For the third recycle the plutonium from the enriched uranium region is again mixed with natural uranium and fabricated in the glovebox facility. The plutonium from the plutonium region and the plutonium from the actinide region plus all the actinides is fabricated in the remote facility. A fissile plutonium concentration of 0.438 gm/cc is again selected and calculations indicated that a natural uranium concentration of 0.168 gm/cc can be used.

For the fourth and subsequent recycles the plutonium and actinide mixture does not have sufficient reactivity to reach the desired burnup. Three alternatives are immediately obvious. First, is the addition of highly enriched

uranium. This unnecessarily complicates the chemical reprocessing step and adds to the volume to be fabricated in the remote facility. Second, is the addition of some of the more reactive plutonium from the enriched uranium region. This also adds to the volume to be fabricated in the remote facility. Third is to increase the enrichments in one or both of the other two regions to drive the actinide region to the desired exposure. For the purposes of this study, it is assumed that the third alternative is feasible and that the increase in enrichment of the uranium region does not alter the quantity of plutonium or actinides to be added to the actinide region each cycle. The resultant actinide inventories for the first nine recycles and an equilibrium case are given in Table 9.C.8. The quantities of the uranium and plutonium isotopes recovered in each recycle for the plutonium recycle strategy are given in Table 9.C.9. Included as the last two entries are the recoveries from the ninth recycle for the strategies of recycling actinides in every UO_2 and in 10% of the UO_2 rods. Small amounts of U-232, Pu-236 and Pu-244 would be present, but these were not included in the calculation, since they do not contribute significantly to the actinide inventory. The U-235 available for recycle decreases through the third recycle and then increases as the enrichment must be raised to compensate for the absorption of neutrons by the actinides. The U-238 decreases each cycle because the actinide region which contains little or no uranium increases in size. This leads to a decrease in Pu-239 inventory in the third and fourth recycle steps. Beginning with the fifth recycle, the Pu-239 again increases as the absorptions in Pu-238 become more significant. Note that the Pu-241 and Pu-242 approach equilibrium slowly leading to the slow approach to equilibrium of the americium and curium as indicated by the data in Table 9.C.8. Recycle of U-236 was not considered.

The economic penalty of the recycle of the actinides, exclusive of the cost of the remote fabrication which may be required, is given on Table 9.C.10. The plutonium value was assumed fixed at \$10.00 per gram fissile, and the other economic parameters were assumed as given in Table 9.C.11. It may be argued that the value of the plutonium in the actinide region is essentially zero from the fourth recycle since the concentration of Pu-238 and Pu-242 are high and since the plutonium actinide mixture requires external enrichment to drive it to goal exposure. The compositions of the plutonium recovered from the actinide region are given in Table 9.C.12. If it is assumed that the value of the plutonium added to the actinide region goes from \$10 to \$0 upon mixing, this cost will be slightly higher than the burnup cost used. However, the interest charge for fuel inventory goes to zero with the net result that the average incremental cost is about 45% of the values shown. If a plutonium value loss is attributed to the actinide recycle in the first recycle case the indicated penalty would increase (possibly to 0.15 mills), the amount depending on the loss and method of assessment. For the second recycle, the penalty would

increase to about 0.062 mills if the value loss occurs in the first recycle or to as much as 0.136 mills if it is assessed against the second recycle. For the third recycle the penalty could decrease to as little as 0.030 mills if the plutonium value loss is assessed against the first two recycles or could increase to as much as 0.166 mills if it is assessed against the third recycle. If fast reactors have no affect on the price of plutonium an independent series of plutonium recycle calculations should be done to evaluate prices for use in these calculations.

TABLE 9.C.8. Actinide Inventory In Grams Per Metric Ton Of Fuel
Plutonium Recycled and Actinides Recycled In Multiply
Recycled Plutonium Rods

Recycle No.	EXPOSURE	NP237	AM241	AM242	AM243	CM242	CM243	CM244	CM245
0	33000.	521.28	64.61	.58	78.63	7.89	.12	22.90	1.07
1	33000.	799.12	214.37	3.11	246.52	23.02	.42	138.54	13.00
2	33000.	987.75	359.82	6.08	444.38	37.50	.81	325.69	33.22
3	33000.	1068.93	424.04	7.30	647.12	50.44	1.28	583.14	55.19
4	33000.	1125.13	472.37	8.39	839.01	55.35	1.60	862.52	77.65
5	33000.	1166.45	509.27	9.27	1018.09	58.12	1.81	1147.23	99.54
6	33000.	1195.53	536.55	9.93	1184.29	60.18	1.95	1429.60	120.01
7	33000.	1216.04	558.31	10.46	1337.74	61.71	2.05	1703.76	138.96
8	33000.	1230.60	576.93	10.91	1479.07	62.96	2.12	1966.00	156.45
9	33000.	1241.04	593.53	11.31	1609.13	64.05	2.18	2214.20	172.54
Equilibrium	33000.	1279.38	772.21	15.45	3181.79	74.83	2.57	5340.75	352.83

TABLE 9.C.9. Uranium and Plutonium Recovered in Gram Per Metric Ton of Fuel

Recycle No.	<u>U-234</u>	<u>U-235</u>	<u>U-236</u>	<u>U-238</u>	<u>Pu-238</u>	<u>Pu-239</u>	<u>Pu-240</u>	<u>Pu-241</u>	<u>Pu-242</u>
0	178	8792	3931	938074	140	5358	2161	1214	452
1	158	7811	3343	933802	463	7174	3622	2320	1088
2	155	7245	3125	879017	861	7278	4329	3019	1803
3	158	6945	3002	840212	1295	6873	4539	3327	2606
4	167	7177	2995	836419	1679	6778	4636	3520	3370
5	174	7463	2988	834145	2007	6819	4702	3652	4061
6	181	7625	2976	830543	2284	6872	4759	3750	4685
7	186	7721	2964	827280	2517	6920	4817	3832	5249
8	190	7821	2955	824400	2712	6962	4875	3905	5762
9	194	7897	2946	821871	2876	6998	4931	3972	6228
Equilibrium	214	8569	2872	800161	3883	7201	5756	4726	11793
9th Recycle in every UO ₂ Rod	193	9767	4048	935643	496	5547	2151	1216	433
9th Recycle in 10% of the UO ₂ Rods	194	10069	4049	935395	568	5537	1241	1222	440

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TABLE 9.C.10. Incremental Fuel Cycle Costs For Increased Enrichment
Due to Recycle of Actinides When Also Recycling Plutonium

<u>Recycle No.</u>	<u>Incremental cost mills/kwhe</u>	<u>Volume Fraction of Actinide Region</u>	<u>Incremental Enrichment in U Region</u>	<u>Core Averaged Incremental cost mills/kwhe</u>
1	.16	.169		.027
2	.71	.080		.057
3	.70	.104		.073
4	.68	.106	.034	.081
5	.64	.108	.075	.089
6	.61	.112	.101	.095
7	.59	.115	.118	.100
8	.57	.118	.135	.103
9	.55	.121	.148	.107
Equilibrium	.42	.144	.271	.132

TABLE 9.C.11. Economics Parameters Used

Reactor Life	30 years
Reactor Cost	\$400.00/KW
Interest on Reactor Cost	10%
Fabrication Loss (Recycled)	1%
Reprocessing Loss	0.5%
UF ₆ Conversion Cost	\$1.50/kgU
Separative Duty	\$32.00/kg
Tails Composition	0.3% U-235
Plutonium Value	\$10.00/gm fissile
Fuel Cycle Working Capital Interest	10%
Fuel Inventory Interest	8%
UO ₂ Fabrication Cost	\$80.00/kgU
Mixed Oxide Fabrication Cost	\$85.00/kg Heavy Metal
Reprocessing Cost	\$40.00/kg
U ₃ O ₈ Cost	\$17.64/kg

TABLE 9.C.12. Composition Of Plutonium Recovered From Actinide Region

Recycle No.	Plutonium Composition, Wt.%				
	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242
1	4.95	39.57	26.37	18.88	10.22
2	12.19	24.10	24.92	22.04	16.75
3	15.39	15.25	23.10	21.75	24.51
4	17.50	11.72	20.53	20.53	29.72
5	18.59	10.72	18.52	19.19	32.97
6	19.18	10.27	17.17	18.10	35.27
7	19.48	9.97	16.25	17.28	37.01
8	19.62	9.73	15.61	16.65	38.40
9	19.64	9.52	15.14	16.17	39.53
Equilibrium	16.54	7.38	13.28	13.46	49.34

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APPENDIX 9.D

TRANSMUTATION BY FUSION (CTR) REACTORS

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APPENDIX 9.D

TRANSMUTATION BY FUSION(CTR) REACTORS

SUMMARY

The unique features of a fusion reactor (otherwise called a Controlled Thermonuclear Reactor CTR) as a waste transmutation device are the high energy of the neutrons available, and the high flux anticipated. Due to the high energy, (n,2n) reactions can contribute significantly to the process, while the high flux and high source strength makes waste transmutation at reasonable rates appear possible a priori. Analyses indicate:

- (1) For the specific isotopes analyzed, calculations indicate that transmutation decreases the radiological toxicity. That is, the ultimate daughter products created are non-radioactive stable isotopes. This is necessary for the feasibility of any transmutation scheme. Waste transmutation is a function of the neutron flux spectrum and this conclusion does not necessarily apply to all neutron transmutation schemes. A particularly attractive transmutation rate for these fission products (FP's) has not been demonstrated by the analysis to this point. However, it appears that detailed parametric studies performed on specific target loading techniques may indicate ways to achieve transmutation rates several times larger than the natural decay rates of these isotopes.
- (2) It appears that there will be adequate physical space in a CTR blanket for fission product loadings, and that the thermal burden of these fission products on the blanket will not be too great.

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- (3) It has been shown that a few CTR power plants can, in principle, transmute all of the Sr-90 and Cs-137 created by an essentially all fission-reactor electrical power economy.
- (4) The daughter products produced appear to be approaching well defined equilibria after a year or so of operation. They should, therefore, cause few perturbations on the characteristics of operating CTR power plants.
- (5) The question of the economics of CTR power plants as fission product burners was found to be most interesting. First of all, it was shown that a transmutation scheme which is a net absorber of neutrons (n, γ) would be relatively expensive. Secondly a CTR transmutation scheme based on the $n, 2n$ reaction could be a net producer of neutrons for long irradiation periods. This observation has a number of implications. First of all, it indicates that the process could be a net producer of income. Secondly, it shows that the process may not require early separation of the daughter products. Because certain fission products may prove to be of value to CTR power plants, this finding has some implications as to what strategy should be used in applying retrievable storage concepts. The real economic factors for the transmutation of radioactive waste in a CTR would appear to be not in the neutronics but in the manufacture of FP irradiation targets. Among other considerations are possible increased capital costs associated with using the CTR for this purpose and increased operational costs for handling and safety.

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- (6) The transmutation of heavy elements in a CTR is a separate consideration. As a rule the longer the natural half-life of a particular isotope and the larger the cross section of that isotope, the more attractive that isotope is for transmutation. Consequently, while Sr-90 and Cs-137 have been found to be possible transmutation targets, heavy element wastes are more attractive for transmutation in a CTR. This is because their neutron cross sections for transmutation are significantly higher. The CTR will be better for neutron-induced transmutation than other known approaches if it attains the higher neutron flux and excess neutron capacity currently predicted. An additional reason why transmutation of heavy isotopes appears particularly attractive in a CTR has to do with the specific neutronic characteristics of a CTR. Analysis has demonstrated the importance of high-energy neutron threshold reactions in CTR transmutation. It is concluded that the $(n,2n)$, $(n,3n)$, and fission reactions could be of major importance in the transmutation of heavy isotopes in a CTR. These reactions would result in either the transmutation of isotopes back up the chain (i.e., $\text{Pu-240} \rightarrow \text{Pu-239}$) or fission. In either case, the net effect could be a rapid decrease of the potential radiological burden on the environment. In addition, the average thermal energy deposited in the CTR blanket per fusion neutron will be enhanced through fission. Thus, the confinement conditions on the fusion plasma would be several times lower than the conditions required for energy breakeven without the presence of fission.

DETAILED RESULTS

The CTR is of interest for the possible transmutation of radioactive waste because of both the large neutron source and high neutron fluxes which have been projected for fusion reactors. The most troublesome fission product nuclei are those which have relatively small neutron reaction cross sections. Consequently, high values of neutron flux are required for reasonable transmutation rates. The requirements on neutron flux-level are obtained by comparing neutron-induced transmutation rates with radioactive decay rates. If the assumption is made that, as a minimum, the transmutation rate needs to be about equal to the natural radioactive decay rate then

$$\lambda \approx \sigma \phi \quad (D1)$$

where λ is the radioactive decay constant and σ and ϕ are the effective transmutation cross sections and fluxes, respectively. The minimum required flux for transmutation is then

$$\phi = \frac{0.693}{\sigma T_{1/2}} \quad (D2)$$

where $T_{1/2}$ is the half-life for radioactive decay. For a $T_{1/2}$ value of thirty years and a reaction cross section of one barn (i.e., 10^{-24} cm^2), the minimum required neutron flux level is about $10^{15} \text{ neutrons}/(\text{cm}^2)(\text{sec})$.

The initial justification for analyzing a CTR as a fission product burner is that it could potentially provide this neutron flux level.

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As previously shown in Section 2, for a wide range of power reactor wastes, Sr-90 and Cs-137 account for almost 85 percent of the total waste activity in curies, and 95 percent of the beta activity. Cs-137 provides 75 percent of the total waste activity from gamma decay. Therefore, Sr-90 and Cs-137 have been identified as primary targets in studies of possible transmutation in CTR blankets.

A. The Transmutation Chain

The neutron-induced transmutation of radioactive fission product isotopes occurs primarily via two processes. Which process is dominant depends upon the energy spectrum of the neutrons considered for the transmutation. In a neutron flux which is predominantly thermal the only significant process is neutron capture. The neutron capture generally results in a product isotope which beta decays with a very short half-life (minutes to days) to a stable nucleus. This product nucleus and nuclei formed by successive neutron captures will generally have thermal neutron capture cross sections comparable to or greater than that of the original target nucleus. Thus, if the target nuclei are present in sufficient quantity to effect the thermal neutron flux, as the numbers of product nuclei increase, they will eventually compete with the target nuclei for the thermal neutron captures. The result of this competition is that the transmutation rate of the target nuclei will decrease with irradiation time. The decreased transmutation rate will depend upon the assumed initial concentration of the target nuclei and the details of radioactive decay and neutron capture cross sections of the nuclei in the transmutation chain. Thus, detailed neutronic

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calculations as a function of initial concentration and irradiation exposure are necessary to establish the possible need for chemical separations at intervals during the irradiation process.

In a fast (i.e., MeV) neutron flux the dominant transmutation process is the $(n,2n)$ reaction. As is the case with neutron capture reactions, the $n,2n$ reaction on the target fission product nuclei usually leads to product nuclei which beta decay with a short half-life to a stable isotope. The $(n,2n)$ cross sections of radioactive fission product isotopes and their $(n,2n)$ product and radioactive decay product nuclei do not differ greatly in the fast neutron flux spectra produced in the blanket of a deuterium-tritium (DT) CTR.

Thus, the same consideration of target nucleus concentration and neutron irradiation effects on transmutation rates and chemical reprocessing apply as for the neutron capture process.

In projections of irradiations of fission product in CTR blankets where neither the capture nor $n,2n$ process is clearly dominant the transmutation chain which must be considered is much more extensive and complex.

The consideration of the transmutation of actinide nuclei is based on the assumption that the ultimate goal of the transmutation is neutron-induced fission in the actinide nuclei thereby creating fission products. In a fast neutron flux the typical actinide nucleus primarily undergoes three transmutation reactions; $(n,2n)$, $(n,3n)$ and fission. In contrast

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to the fission product transmutation process, the $(n,2n)$ and $(n,3n)$ processes do not lead to more desirable nuclei from a radioactive waste standpoint. The product nuclei simply become other target nuclei which ultimately must be fissioned. Consequently, the merit of the possible irradiation of actinides in a purely fast neutron flux of a CTR blanket will depend upon the assumed initial isotopic content of the actinide mixture and of their neutron cross sections. Detailed studies of the fission rates, radioactivity, toxicity, and retention rates as a function of neutron irradiation time, are needed to establish the relative merits of this possible type of transmutation

In a thermal neutron flux the dominant transmutation processes are capture and fission. In this case, the capture reaction and successive captures do not produce desirable nuclei but simply compete with the fission process. The same factors must be considered as for irradiation in a fast neutron flux to determine the relative merits of transmutation in purely fast, purely thermal, or mixed spectrum neutron fields.

Preliminary studies have been made at PNL of the transmutation of fission product nuclei and actinides in different types of neutron irradiation fields of CTR blankets. These studies have encompassed:

- The isotopic content versus irradiation of dilute Sr-90 and Cs-137 targets in a fast neutron flux spectrum.
- The isotopic content versus irradiation of dilute targets of Sr-90 fission product Sr, Cs-137, fission product Cs, Kr-85, I-129, and a mixed actinide target in a predominantly thermal neutron flux spectrum.

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- The initial transmutation rate versus the Cs-137 concentration to a neutron moderating blanket.

The results of these investigations are summarized in the following sections.

B. Transmutation Calculations

The Fast Neutron Flux Transmutation of Sr-90 and Cs-137 - Wolkenhauer has reported⁽¹⁾ the results of calculations of the isotopic content versus irradiation time of dilute targets of Sr-90 and Cs-137 in a predominantly fast neutron flux spectrum of a DT fusion reactor. A detailed discussion of these calculations is given in a BNWL report.⁽²⁾ The neutron flux spectrum which was used to calculate the reaction rates for the isotopes in the transmutation chains was that calculated by ANISN⁽³⁾ for the dominant lithium zone of the standard calculational benchmark blanket.⁽⁴⁾ The integrated fast neutron flux level was taken to be 5×10^{15} neutrons/(cm²) (sec). This flux level is obtainable in approximately the first 10 cm of the lithium zone for a 14 MeV neutron power inner wall loading of 10 MW/m². The isotopic contents versus irradiation time were calculated including neutron capture and (n,2n) reactions and radioactive decay using the transmutation program ALCHEMY.⁽⁵⁾

The calculated fractions of the isotopic products of the irradiation for times up to three years are shown for the Sr-90 target on Figure 9.D.1 and for the Cs-137 target on Figure 9.D.2. For the Sr-90 target the n,2n reaction on Zr-90 was not included in the calculation. Since this reaction produces

FIGURE 9.D.1. Buildup of Sr-90 Daughter Products with Time

-9.D.10-

FIGURE 9.D.2. Buildup of Cs-137 Daughter Products with Time

Zr-89 which decays shortly (78.5 hour $T_{1/2}$) to Y-89 the net result is only a stable nucleus and the Zr-90 fraction shown on the figure contains some Y-89. Similarly, the transmutation chains for the Cs-137 target were terminated at Ba-135 and La-139 so that the fractions shown on the figure for these isotopes also contain product nuclei.

The neutron production rates per neutron absorbed in the mixed nuclide samples of the initial Sr-90 and Cs-137 targets were also calculated as a function of irradiation time. The values of neutron production per neutron absorbed were about 1.75 for the Sr target and about 1.50 for the Cs target. Neither value changed more than a few percent over the three year irradiation which was calculated. Thus it was concluded that in dilute targets the FP nuclei contributed neutrons to the economy of the CTR blanket.

Thermal Neutron Transmutation of Dilute Target Samples - Preliminary studies sponsored by the USAEC Division of Controlled Thermonuclear Research have been made at PNL, in which the intent was to establish an approximate upper bound to the highest transmutation rates which could be achieved utilizing the neutrons produced by a controlled thermonuclear fusion reaction. The results of these studies help to establish the types of radioactive waste targets which are worthy of more serious and practical investigation. The detailed results of the calculations have been published in a BNWL document⁽⁶⁾ and summarized in an American Nuclear Society presentation.⁽⁷⁾

9.D.12

Consistent with the intent of the study as given above, calculations have been made using a moderating blanket, the initial portion of which is beryllium, to obtain multiplication of the fusion plasma source neutrons through the beryllium $n,2n$ reaction. The geometry chosen for the calculations was essentially that of the CTR calculational standard benchmark,⁽⁴⁾ a TOKOMAK with 200-centimeter inner vacuum wall radius and a blanket 100 centimeters thick. Calculations have been made for both D-D and D-T fusion plasmas, each with a fusion reaction rate producing a neutron power loading at the inner wall of 10 MW/m^2 . The neutron distribution in the blanket was calculated using the ANISN code.⁽³⁾ A maximum thermal flux value of almost 3×10^{16} neutrons/(cm^2) (sec) was found to occur at the inner surface of the beryllium moderator in the absence of any absorbing vacuum wall material. The transmutation calculations which were made utilized the neutron energy spectra and flux values representative of that of the first centimeter thickness of the Be blanket.

Radioactive waste inventories representative of an expected LWR economy were established using results obtained from the ORIGEN code⁽⁸⁾ for chemical isotopic mixtures of fission products Sr and Cs and of actinide isotopes. As before, the transmutation code ALCHEMY was used to calculate the isotopic concentrations for dilute targets as a function of irradiation time. A computer code was written to transform the calculated isotopic activities to dispersal toxicity values.

The results of these calculations are shown on Table 9.D.1 for both D-D and D-T neutron sources and for neutron wall loadings of 1 and 10 MW/m^2 . The

TABLE 9.D.1. Toxicity Half-Life^(a) as a
Function of Neutron Wall Loading (P_w)

		Toxicity Half-Life, Years		
Target	Plasma	$P_w = 0.0$	$P_w = 1.0 \text{ MW/m}^2$	$P_w = 10.0 \text{ MW/m}^2$
Sr-90	D-T	32.0	7.6	1.3
Sr ^(b)	D-T	32.0	8.0	7.4
Sr-90	D-D	32.0	4.7	0.48
Cs-137	D-T	30.0	19.0	5.6
Cs ^(b)	D-T	14.0	12.0	4.0
Cs-137	D-D	30.0	16.0	4.4
Kr-85	D-T	11.0	8.6	6.1
I-129	D-T	1.6×10^7	0.35	0.27
Actinides	D-T	460 ^(c)	0.60	0.056
Actinides	D-D	460 ^(c)	0.23	0.037

- a. Defined as the time required for the toxicity index from a particular isotope and its daughters to decay to one-half of its original value. The toxicity index itself is the volume of air or water required to dilute the nuclide to its Radiation Concentration Guide value.
- b. This is isotopic mixture of Sr or Cs from LWR fuel at 33,000 MWD/MT and 10 years cooling.
- c. Am-241 only.

expected large enhancement in reduction of actinide toxicity due to the neutron irradiation is apparent. The actinide toxicity values do not include the effects of fission product created in the fission process. A similar drastic reduction in toxicity values is obtained for a target of I-129. The reduction in toxicity values for D-D source neutrons relative to D-T source neutrons for the same neutron wall loading arises primarily from an increase of about a factor of two in thermal neutron flux. The relative merits of isotopic separation of Sr and Cs cannot be clearly deduced from just the hazard values listed on Table 9.D.1. More detailed study of economic and other factors is required to produce any value judgments in this area.

Transmutation of Quantitative Amounts of Cs-137 in a Moderating Blanket

The results of the preceeding studies of irradiations of Cs-137 have shown that there are some potential hazard reductions when dilute samples are irradiated in either fast or thermal neutron flux spectra. The indicated reductions were marginal in benefit, and further studies were considered to be desirable. The studies of dilute targets had not quantitatively assessed the quantity of Cs which could be subjected to transmutation. As a result, it was decided to study the transmutation rates as the loading of Cs in a Be moderating blanket was progressively increased. The results of this study have been summarized in an American Nuclear Society presentation⁽⁹⁾ and are published in more detail elsewhere.⁽¹⁰⁾

The calculational model of this study was intended to represent a more realistic blanket configuration which was, as before, built upon the standard calculational benchmark model with a D-T fusion plasma with

a neutron wall loading of 10 MW/m^2 . In these calculations an inner vacuum wall of 0.5-centimeter thickness of niobium was placed at the 200-centimeter radius as shown in Figure 9.D.3. This was followed by a 3-centimeter coolant zone (void in the calculations) and a second 0.5-centimeter Nb wall. The target zone was the next 20-centimeters annular thickness of the blanket. The target zone was followed by an annular zone of beryllium of 70-centimeter thickness followed by a 6-centimeter thickness of lithium. The calculated transmutation rates in the target zone were found to not be very dependent on whether the 70-centimeter zone of beryllium was replaced by graphite or whether its thickness was significantly reduced.

In the initial calculation the 20-centimeter thick target zone was filled to 80% volume with beryllium and a 20% volume void space to allow for gas cooling channels. The infinite dilution reaction rates of Cs-137 were then calculated using the ANISN code. The calculated maximum and volume averaged reaction rates for the neutron capture and $n,2n$ processes which were obtained are shown as the first entry of Table 9.D.2. The Cs-137 concentration was then progressively increased, a given volume fraction of Be being replaced by Cs-137 at the atom density of elemental Cs as indicated on the following entries of the table. The total specific transmutation rates for Cs-137 are seen to be not very dependent on Cs concentration. the main effect being an increase of the $(n,2n)$ reaction rate to almost compensate for the decrease in the neutron capture rate as the Cs concentration is increased.

In view of the above results, the beryllium moderator was removed from the target zone and the effect of filling this void with different volume

9.D.16

FIGURE 9.D.3. Modified CTR Standard Blanket

TABLE 9.D.2. Calculated Transmutation Rates of Cs-137 in the Blanket of a Tokamak Fusion Reactor

Volume Fraction %Cs/%Be	$\phi(1/v) \times 10^{-16}$ Neut/(cm ²)(sec)	Average				$\phi(1/v) \times 10^{-16}$ Neut/(cm ²)(sec)	Maximum		
		Reaction Rate < $\sigma\phi$ > _{n, γ} fraction (year)	Reaction Rate < $\sigma\phi$ > _{n, 2n} fraction (year)	Total Effective T-1/2 ^(a) (years)	Total Transmutation Rate ^(b) kg/m (yr)		Reaction Rate < $\sigma\phi$ > _{n, γ} fraction (year)	Reaction Rate < $\sigma\phi$ > _{n, 2n} fraction (year)	Total Effective T-1/2 ^(a) (years)
0/80	1.19	0.0428	0.0115	9.0	0	1.26	0.0450	0.0266	7.3
5/75	1.16	0.0419	0.0119	9.0	20	1.23	0.0440	0.0267	7.4
25/25	1.05	0.0379	0.0137	9.3	96	1.13	0.0402	0.0273	7.7
50/30	0.874	0.0321	0.0168	9.7	185	0.958	0.0349	0.0281	8.1
50/0	0.743	0.0275	0.0241	9.3	191	0.763	0.0282	0.0292	8.6
80/0	0.671	0.0250	0.0221	9.9	291	0.702	0.0262	0.0290	8.9
100/0	0.633	0.0238	0.0209	10.3	352	0.668	0.0252	0.0289	9.0

a. Includes radioactive decay activity of 0.0229 per atom per year.

b. This device produces 200 MW(t) per meter of CTR device length.

fractions of only Cs-137. From the calculated results of this study as shown on Table 9.D.2 it is again apparent that the total specific activities are not very sensitive to the amount of Cs loaded or to the presence of beryllium. The maximum practical amount of Cs which can be transmuted is the 291 kg/yr per meter length of blanket as obtained for an 80% volume of Cs-137 in the target zone. This quantity of transmuted Cs-137 has been compared with the amount of inventory Cs-137 by considering the Cs-137 produced in a LWR. For this particular Cs-137 loading, the Cs-137 production rate in a LWR would equal the transmutation rate in a CTR blanket of the same thermal power if only one percent of the CTR blanket were loaded for the Cs-137 transmutation.

The conclusions of this study are that interesting quantities of Cs-137 could be transmuted under the projected CTR blanket loading conditions. The reductions in Cs-137 toxicity are, however, still projected to be at most a factor of about three. In addition, the buildup of product nuclei has not yet been studied in order to establish the requirements of periodic chemical processing and associated costs.

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APPENDIX 9.E

COMMENTS OF PEER REVIEWERS

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APPENDIX 9.E

COMMENTS OF PEER REVIEWERS

SUMMARY

Seven people technically expert in the various areas reviewed the draft copy of the study, High-Level Waste Disposal Alternatives: TRANSMUTATION PROCESSING BNWL-B-301 Section 9, and provided their comments.

The peer group met with PNL and AEC personnel in Seattle on January 14th to discuss the report, give their views, and resolve differences of opinion. Those in attendance were:

Peer Group

Dr. J. L. Crandall
Dr. R. E. Hellens
Prof. A. S. Kubo
Prof. H. N. Lefevre
Prof. C. J. Poncelet
Prof. B. H. Spinrad
Dr. D. G. Foster, Jr.

Savannah River Laboratory
Combustion Engineering, Inc.
U.S. Military Academy
University of Oregon
Carnegie-Mellon Institute
Oregon State University
Los Alamos Scientific Laboratory

PNL

C. M. Heeb
B. R. Leonard, Jr.
R. C. Liikala
T. I. McSweeney
K. J. Schneider

AEC

R. W. Ramsey

In summary, there was general agreement with respect to conclusions on technical feasibility. Those areas where consensus or near-consensus opinion exists and where the report was modified to reflect these opinions are as follows:

1. Actinide recycle in LWRs seems technically feasible but not necessarily attractive.
2. The LMFBR is probably more attractive for actinide recycle. An immediate Research and Development goal should be an evaluation of the concept.
3. If actinide recycle in LWRs is to be done, the need for immediate irradiation experiments is urgent.

9.E.2

4. The actinide recycle concepts in LWRs calculated by Claiborne and by PNL almost certainly represent a worst case in terms of reactor operation and economic penalty.
5. The concept of actinide recycle in LWRs as a burnable poison needs to be evaluated.
6. The use of a captive fission reactor, perhaps special-purpose, perhaps part of the National Repository, deserves serious consideration as an alternative to actinide recycle throughout the fission power reactor industry, LWR or LMFBR.
7. Actinide recycle in LWRs needs to be evaluated as a hazard relative to plutonium recycle as a base case.
8. The concept of the transmutation of actinides in a near-critical fission assembly driven by an accelerator neutron source is not attractive since the same job can be accomplished in fission reactors at much less cost.
9. The utilization of thermonuclear explosives to transmute radioactive waste is probably not publicly acceptable, even if feasible.
10. Considering transmutation of waste in fusion reactors, it should be emphasized that CTRs do not presently exist.
11. Transmutation of waste in fusion reactors should be based on a captive, isolated CTR--not a CTR power economy.

Additional analysis and discussion has resolved the main difference of opinion regarding the feasibility of using accelerators and nuclear explosives as transmutation devices. This report reflects the comments provided by the peers.

9.E.3

E. I. DU PONT DE NEMOURS & COMPANY

INCORPORATED

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January 10, 1974

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Dear Dr. Leonard:

At your request I have reviewed BNWL-B-301, Section 9, the Transmutation Processing section of the Battelle investigations of high level waste disposal alternatives for their Advanced Waste Management Studies. The report accomplishes its main purposes in admirable fashion, identifying those transmutation processes worth a detailed study and providing a comprehensive and systematic review of essentially all the possibilities. Actually I must admit it was not until I had worked through the report references that I realized just how necessary such a unified study was to correct the many misconceptions in the literature. I am in general agreement with the report's conclusions that few processes except transmutation in inexpensive neutrons from the still-to-be-developed fusion reactors or actinide transmutation in fission reactors hold current promise. However, as a small quibble, there might also be limited promise to byproduct transmutation in nuclear explosions performed primarily for gas stimulation or similar Plowshare purposes, even though the report quite properly demonstrates that the nuclear explosions would not be attractive for waste transmutation alone.

As usual, in a basic study of this sort the key questions are more philosophical than technical, and I would like to raise two of these questions. The first concerns the urgency for the actinide recycle. As clearly developed in the report and in most other waste management studies, the hazards from the byproduct actinides (excluding plutonium) are several orders below the fission product hazards for the first few hundred years of waste storage, but do become limiting (along with some of the long-lived fission products, such as ^{129}I) at very long times. The current consensus seems to be that the fission products can be safely put into retrievable engineered storage for 50-100 years pending development of ultimate disposal methods, and the question becomes why should this not also be done with the byproduct actinides, which, due to their lower heat generation, are probably much easier to store in the interim period. There seem to be three possible reasons why the actinides might merit special treatment. All are connected with the fact that the actinides need to be safeguarded for more than a million years whereas most of the fission products will

9.E.4

have decayed to safe levels in less than a thousand years. First, the ultimate disposal methods chosen for the fission products may not be suitable for the longer time demands of the actinides. Second, the combined costs of interim plus ultimate disposal of the actinides may exceed that of immediate transmutation. Third, the actinide hazards may be greater than should be accepted for interim storage.

I believe that the first two possibilities can be eliminated. In the first regard, many of the disposal methods being proposed for the fission products, such as disposal in space, do apply to the actinides and, even if special methods are required, it is almost certain that in 50-100 years less expensive alternatives can be developed than transmutation in light water reactors. Similarly, in the second regard, both the possibility of relatively inexpensive future disposal methods and time-value-of-money considerations say that immediate actinide transmutation will be less expensive than interim storage and later disposal only if it is in fact less expensive than the interim storage alone, which is demonstrably untrue. Thus the real question comes down to whether interim storage is safe for the byproduct actinides, which might contaminate the earth for a million years if they were released. A direct answer can be given only by detailed biological and transport studies. However, the problem can also be put into perspective by recognizing that, on a gram-for-gram basis, the plutonium produced in the reactors offers approximately equal hazards to the byproduct actinides and that any extraordinary circumstances which could lead to actinide release from engineered storage - war, technological breakdown, etc., - are even more likely to lead to plutonium releases. Thus in viewing the hazards reduction from byproduct actinide burnup, the percentage base should not be from the byproduct actinides alone but from all actinides including plutonium. Since the byproduct actinides are only produced at about a tenth the rate of the plutonium, this change in bases greatly reduces the importance unless a fuels recycle program is proceeding to burn up the plutonium without touching the byproduct actinides.

My own prejudices on actinide treatment in view of this analysis are that partitioning of the actinides may be justified at the present time since it greatly broadens the fission product storage options and might be much more difficult to perform at a later date. Also, the separated actinides may eventually be valuable products in their own right as radiation, heat, or neutron sources. Likewise, it would be desirable to have more data on fast reactor transmutation of the actinides. However, little additional work seems justified on light water reactor transmutation at this time, particularly since some pertinent data are still coming in from the various transplutonium programs (Savannah River, for example, expects to publish an evaluated multigroup cross section set by June).

The other philosophical question I would like to raise is on the use of dilution to RCG as a hazard index. Certainly it makes a simple and definite index, and it is used sensibly and effectively throughout the report. However, as a general purpose index it has obvious disadvantages since it takes no account of half-life or dispersibility. Worse, if the ultimate disposal method effectively eliminates the waste, say by shooting it into the sun or even by guaranteed geologic dispersal, such a hazard index has no meaning at all, the required index being merely a yes-no index of those nuclides that have to be removed and those that don't.

The following are a few pertinent notes I made while reading the report.

Page

- 9.3 As noted in text, hazards index considered is probably too simple for general use.
- 9.5 Need economic criteria as well as physical ones.
- 9.20 Seems to consider that undesirable products are formed on a ratio of 1 per fission. If plutonium is not considered, more like 0.1-0.2 per fission.
- 9.23 Repeated in my copy.
- 9.25 ^{129}I could probably be transmuted quite effectively in a spallation reactor.
- 9.47 Some of the required cross sections are available in new SRL studies (in press) for ^{252}Cf production.
- 9.55 I personally put the partitioning studies at the highest priority in the research programs, since the text shows clearly that good partitioning is basic to most other handling methods.
- 9.60 Potential reduction in hazards by two orders of magnitude is true only if plutonium produced in reactor is excluded from consideration.
- 9.61 Since these programs always seem to cost more than anticipated, it is doubtful if economics will be attractive under even the "better" methods.
- 9.66 I believe costs increase by ~1%/year, not 0.1%/year.
- 9.76 Two rather than to.
- 9.A.4 Apparently the energy calculation is performed on the basis of one undesirable product per fission but since accelerator electrical efficiency is also neglected overall balance, it is still approximately correct.
- 9.A.28 U-235 for U-238?

Sincerely yours,

J. L. Crandall, Director
Advanced Operational Planning

JLC/fy

TITLE: REVIEW OF PNL STUDY ON TRANSMUTATION PROCESSING OF HIGH
LEVEL WASTE

AUTHOR(S): D. Graham Foster, Jr., Editor

SUBMITTED TO: Presented at meeting held at the Battelle Seattle
Research Center for preliminary study of alternative
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Transportation, January 14, 1974.

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REVIEW OF PNL STUDY ON TRANSMUTATION
PROCESSING OF HIGH LEVEL WASTE

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SUMMARY

We have reviewed briefly a draft of BNWL-B-301, Section 9: "Transmutation Processing, Advanced Waste Management Studies, High Level Waste Disposal Alternatives," at the request of Pacific Northwest Laboratories. This document describes a preliminary study of transmutation concepts (accelerators, thermonuclear explosives, fission reactors, fusion reactors) for the management of fission-product and actinide waste generated in the nuclear-energy economy. We strongly support the principal conclusion of the study; i.e., that disposal of higher actinide nuclides by recycling in fission power reactors appears to be attractive with near-term, high-likelihood technology, and that major R & D emphasis is appropriate in that area (page xi). Effective, large-scale applications of this concept will depend on R & D which affects the economics of recycle utilization of the actinides including plutonium. With respect to disposal of fission-product nuclides, we feel that the work done to date may have discarded fission reactor transmutation of selected nuclei (after chemical separation) prematurely, whereas a more detailed study may demonstrate feasibility for special cases.

We support the PNL conclusion that transmutation both of actinides and of fission products in fusion reactors appears to be sufficiently attractive that a modest R & D effort is projected in this area. We feel, however, that the case for transmutation by particle accelerators is stronger than suggested and deserves an R & D effort to determine the utility of GeV proton bombardment with or without fission boosting. In this connection we describe a fast actinide burner, either stand-alone or driven by a proton accelerator, which appears to have economic potential.

Contrary to the PNL conclusion, we suggest that transmutation of high-level waste by neutrons from an underground nuclear explosion may be very promising, rather than infeasible.

9.E.8

I. INTRODUCTION

This report was prepared in response to a request from the Pacific Northwest Laboratories of the Battelle Memorial Institute for a review of a draft of BNWL-B-301, Section 9: "Transmutation Processing, Advanced Waste Management Studies, High Level Waste Disposal Alternatives." This section discusses transmutation processing for disposal or utilization of radioactive wastes from nuclear-energy sources. It was not possible in the time available to prepare a review which is appropriate to the importance of the problem. Accordingly, what follows is a composite of views expressed by the contributors, grouped into sections devoted to related concepts. Comments expressing the views of one or more contributors are arranged as a lettered sub-section. Some of these views are somewhat disparate in detail, reflecting both the short time available for editing and technical differences in judgement. There is a general consensus, however, on the conclusions stated in the summary.

II. CONCEPTS FOR EVALUATING TRANSMUTATION AS A METHOD OF WASTE MANAGEMENT

- A. Most of the contributors feel that the dilution "hazard index" is an appropriate indicator for beginning exploration of disposal methods. However, it ignores some vital details and may be misleading in making a final decision. The effective volume of air or water which dilutes an accidentally released radionuclide is difficult to predict. In order to determine the hazard to the general population, the transport properties of the material released need to be considered in detail, and the pathways by which it can affect the general population explored individually to determine which one presents the greatest hazard. A simple change in chemical or physical form can drastically alter the effective hazard index.
- B. There is a semantic problem in the words "hazardous" and "waste." At present, certain hazardous fissile and fertile nuclides are considered useful, whereas other actinide nuclides are considered waste or a "nuisance." The very long-lived products of reactors have such low specific activities that it may be misleading to label them as problems. For example, ^{99}Tc is treated by PNL as a waste to be disposed of, although $^{99\text{m}}\text{Tc}$ is routinely injected into humans for gamma radiography of the brain, and apparently causes no biological problems after it decays to ^{99}Tc . In this respect it appears no more desirable to transmute it to some shorter-lived species than to transmute the ^{232}Th used in the mantles of gasoline lanterns.
- C. Table 9.3 gives the hazard index for some important fission products and for tritium. We feel that the discussion of transmutation would benefit if the hazard index for each nuclide was included when it is discussed. It would place these in better perspective to tabulate also the hazard indices of ^{232}Th , natural and slightly enriched U, and fuel-grade ^{233}U and Pu. The latter are hazards, but certainly not "waste."

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- D. In considering transmutation, it is instructive to factor the hazard index,

$$h = I(1/\tau),$$

where I contains the biological information and $1/\tau$ is the mean decay rate per nucleus. Then the hazard index integrated over all time is

$$H = \tau(I/\tau) = I,$$

which is independent of τ . Thus, unless transmutation changes the biological considerations the cumulative hazard is not changed by shortening the half life. The primary advantage of such a transmutation is then in decreasing the time span over which the waste must be protected. Note, however, that in fissioning actinides the predominant radiation is changed from short-range α emission to more penetrating β^- and γ , which complicates further handling.

If a nuclide with mean life τ_0 is taken from storage where it has a mean probability per unit time of catastrophic release P_0 and placed in a transmutation device which shortens its hazard mean life to τ_t with a catastrophic release probability P_t , the time-integrated hazard to the population is changed from $P_0\tau_0$ to $P_t\tau_t$. Clearly we would like to require

$$P_t\tau_t < P_0\tau_0 \quad \text{or} \quad \frac{1}{\tau_t} > \frac{P_t}{P_0} \frac{1}{\tau_0}$$

If we hypothesize that in a reactor, as opposed to a salt bed, $P_t/P_0 = 100$, then we must require that the mean life be shortened by at least a factor of 100 (rather than "several times") before transmutation can be justified. This makes the transmutation of such fission products as ^{137}Cs and ^{90}Sr , which have the highest hazard indices, even more unattractive than estimated by PNL, but makes transmutation of the very long-lived wastes more attractive even with low conversion rates. We must also require that the transmutation products be either stable (or so long-lived as not to be considered radiological hazards) or else have half lives so short that they decay essentially completely in acceptably few years or decades.

- E. These considerations underline the fundamental fact that any processing of wastes involves temporarily increasing both the hazard to workers in the operations of refueling and processing, and the probability of catastrophic release. In marginal cases, this would favor long-term storage over transmutation.
- F. Our contributors feel strongly that the feasibility of any transmutation cannot be proved or refuted without a detailed examination of each nuclide in question and of its possible pathways to the population and environment in the event of an accidental release from any point of the processing/refueling operations. Since each nuclide may require its own strategy of management, we concur with the PNL emphasis on the importance and value of chemical, and perhaps even isotopic, partitioning.

- G. It is too early to assess accurately the prospects for laser-induced photochemical separation of elements and of isotopes. However, we feel that this possibility might become very important within, perhaps, a decade, and is certainly more promising than the gamma-ray laser. It might be expected to lower the cost of partitioning, improve the stripping of actinides from the fission-product stream, and perhaps profoundly alter the economical strategies of reactor design and fuel recycling. Even without such a breakthrough, Kubo and Rose have pointed out the substantial advantage in pushing the concentration of Pu in the fission-product stream below 10^{-4} a/o by conventional means. The draft report does not explain why the separation of fission products from Pu is so much more complete than the separation of Pu from the fission products.

III. ACTINIDE RECYCLE IN FISSION REACTORS

- A. Our contributors are virtually unanimous in endorsing the promise of actinide recycle in fission reactors. The PNL report emphasized actinide recycle in fission power reactors, primarily LWR's, but special-purpose reactors also may have promise. Certain actinides produced during fission power reactor operation, e.g., plutonium isotopes, are already projected for recycle as power reactor fuels.

The effective, large-scale recycle of higher actinides in fission reactors will depend on the successful resolution of a number of technical and economic factors. Many of these factors are common both to problems of plutonium recycle and to higher-actinide recycle and should be examined together. The design of actinide-recycle systems presents a large number of options, and it is not clear that the economic costs and benefits can be reliably projected without careful optimization. In general, it appears that among the penalties of actinide recycle are additional shipping hazards and costs, additional hazards in the event of a loss-of-coolant accident, and additional complexity and cost of the design process. Among the benefits of higher-actinide recycle are reduced costs for long-term actinide storage and reduced fuel-separation costs. It is not completely clear that there is an economic penalty associated with inclusion of higher actinides in plutonium fuel. It is possible that, with optimized reactor design, increased uranium enrichment and increased control-rod swing can be avoided. The cost of alternative actinide disposal must be borne by the power producer if the benefits of actinide recycle are to be apparent.

- B. It is worth underscoring the PNL caution that any transmutation blanket or loading added to a reactor, whether fission or thermonuclear, acts as a parasite in competition with breeding. The loading of waste which would still permit breeding is not necessarily large enough for the reactor economy to consume its own higher actinides at equilibrium. Clearly calculations directed at this question should have a high priority in Phase 2 of the program.

9.E.11

- C. Kubo and Rose have claimed that the "relative waste toxicity" of the fission products in the early decades of decay exceeds that of all actinides together by a factor of more than 500. Thus, the immediate problem of high-level wastes is overwhelmingly that of the fission products. This fact will probably have to be borne in mind in establishing funding priorities for developing all-actinide recycling technology.
- D. We emphasize that every industrial process generates waste. Processing recycled actinides currently generates wastes which are sometimes inordinately expensive to recover chemically, and they may be in forms more dangerous than before processing. To prevent accumulation of such nominally low-level waste the definition of "scrap" must be set low enough to discourage discarding inconvenient forms as "waste"--a conclusion which runs counter to the present trend. In fact, the economic penalty of actinide wastes may have to be increased artificially (or a bonus established for recovering scrap) at the taxpayers' expense in order to insure that commercial power plants will accept the modest penalties incurred by recycling, as projected by the PNL study.
- E. In present-day ^{235}U -fueled reactors the buildup of higher actinides is due mostly to the presence of ^{238}U as a fertile material. If isotope separation becomes dramatically cheaper in the near future, it might be useful to compare the economics of highly-enriched uranium burners with the possible cost reduction in managing the higher-actinide waste. The short-term effect would be to delay the buildup of higher actinides until the technology for managing them becomes economically competitive.
- F. It is often forgotten that ^{232}Th and ^{238}U are probably the only long-lived actinides which do not have finite critical masses. In other words, it is quite possible that an assembly fueled by the higher actinides alone could be made to go critical and consume the undesirable nuclides with a bonus of additional power production. The spectrum would probably have to be much harder than contemplated for current designs of fast breeder, and the delayed-neutron fraction does not appear easy to predict, so control might be more difficult than envisioned in present fast-reactor designs. However, as a "waste burner" which is not required to make economical power, such a reactor could be sited in a remote area and optimized for safety rather than electrical efficiency. The total thermal power of enough burners to consume the actinide waste of an equilibrium recycle economy would be in the range of a few to 10 percent of the thermal power in the nuclear-power industry as a whole, in direct proportion to the fraction of higher actinides in the overall actinide inventory. The extremely hard spectrum at reasonably high flux might also be attractive for testing components and as a neutron source for other measurements.
- G. As long as the delayed-neutron fraction is sufficient to provide a clear margin between delayed and prompt critical such a burner should be controllable. To increase the margin of control, it might be worth returning to the slow-fast coupled-reactor concept. A thermally-critical region with its characteristically long generation time would be used to dictate the response time of the entire assembly, which would be subcritical without the thermal region, but still have a very hard spectrum in the burner region. Similar

9.E.12

designs have also been studied for the thorium thermal breeder, in which power level (and neutron economy) was to be controlled by adjusting the coupling (neutron leakage) between the regions, rather than by parasitic absorption. The multiplying blanket might be either inside or outside the thermal seed, or merely adjacent to it.

- H. A still safer design would involve a subcritical assembly fueled with the unwanted actinides and driven by a high-power proton accelerator. This variation resembles one cited by Claiborne. A very rough study at LASL suggests that $k = 0.97$, so that $(1-k)^{-1} = 30$, is an appropriate value for the actinide assembly. An efficient accelerator of the LAMPF design would produce neutrons by spallation directly in an actinide target at a cost of 50-100 MeV/n. Realistic efficiency estimates (see Section VII) suggest that electricity generated by steam from the multiplying region at this rather modest multiplication would just supply the required 100 MeV/n in the accelerator beam. Control would be achieved by adjusting the ion source of the accelerator. Conservative cost estimates, assuming that production-line accelerators would cost half as much as the prototype, yield a capital cost of about 20% of the capital cost of the power reactors serviced by the actinide burner. The possibility of offsetting unexpectedly high energy costs by higher multiplication in the burner region is obvious. Similar designs have recently been studied for plutonium breeding and as neutron sources for experiments.

IV. FISSION PRODUCT RECYCLE IN FISSION REACTORS

- A. We concur with the pessimistic PNL view on transmutation of fission products in fission reactors. However, we are not satisfied that the study to date has conclusively disproved the feasibility for all prominent fission products. Studies done originally for the prediction of reactor after-heat have disclosed many complex and unexpected transmutation chains. We suspect that chemical and isotopic partitioning of the fission-product waste will be required for useful lowering of hazard index.
- B. In particular, thermal radiative capture in the rare earths from Ce through Dy is clearly unprofitable, since most of the products are no less hazardous than the targets. Placing ^{151}Sm in an epithermal flux might deserve further study, but the yield of this nuclide is small. From the arguments given in Section II-D, even thermal transmutation might be useful on the fission products with half lives greater than 10^4 y. It is not clear whether these were studied by PNL.
- C. PNL appears to have limited consideration of sources of troublesome radionuclides to the fuel itself. Neutron interactions with cladding and structural materials can also produce long-lived products such as ^{94}Nb and ^{93}Zr . Zr-clad fuel elements at typical burnup, for example, will have had about 0.5% of the cladding converted to ^{93}Zr .

V. TRANSMUTATION IN FUSION REACTORS

- A. CTRs are expected to be relatively "clean," in the sense that their burden of radionuclides will be much smaller than in fission reactors. They appear attractive for siting near population centers for this reason. Use of such reactors to transmute large loads of radioactive waste from fission reactors would greatly increase the hazard to the population, relative to the hazards associated with underground storage. As with fission reactors, the parasitic blanket of unwanted nuclides competes with breeding--in this case tritium--which may prevent usefully fast destruction of the waste nuclides.
- B. Useful destruction of nuclear waste by plasma devices need not wait for commercial power-producing CTRs. Dense-plasma-focus devices at present produce fluxes too small to be attractive, but seem likely to be capable of being scaled up before self-sustaining CTRs become available. Laser-pellet concepts may also prove useful in producing a non-self-sustaining plasma device for transmutation applications.

VI. TRANSMUTATION USING NUCLEAR EXPLOSIONS AS A NEUTRON SOURCE

- A. We feel that this alternative has been discarded prematurely. The basic PNL assumptions were so far from optimum that the conclusions were unreasonably pessimistic. Most questionable are the assumptions that 5 kt is an appropriate size of device and that transmutation would be limited to one cycle.

Studies at LASL suggest 100-kt devices at a depth of 1.5 km as an attractive base case. The ground motion would be quite acceptable at this depth. The cost of drilling holes much larger than 60 cm is not prohibitive. The cost has been estimated at roughly 500 k\$ for the device and an equal amount for site preparation (if the cost is prorated over several explosions). The device can be relatively clean, with a small fission yield. The cavity can probably be reused repeatedly, with the products from each shot being recovered, partitioned, and recycled. The above parameters lead to a cost of about \$5,000/mole of fission products transmuted. If we set out to transmute the worst 10% of the fission products from reactors, the cost of transmutation comes to 0.4% of the value of the electricity produced by the reactors. Thus, this technique looks quite promising for ^{129}I , ^{135}Cs , and ^{99}Tc .

- B. An old Plowshare concept is again under study, aimed at producing economical electric power from steam using underground explosions as the heat source. If this concept can be combined with burning the most offensive fission-reactor products, the process may prove to be very attractive

VII. TRANSMUTATION BY CHARGED-PARTICLE ACCELERATORS

- A. We have described in Section III-H an accelerator-driven barely-subcritical actinide burner which looks more attractive than any purely accelerator-induced process. We have also referred briefly in Section V-B to non-self-sustaining plasma devices which might loosely be classed as accelerators.

9.E.14

In addition, we feel that the case against direct accelerator transmutation is enough weaker than suggested by PNL to justify further exploration before abandoning this approach.

- B. The initial conclusion of the PNL study is, of course, clearly correct. Charged particles of a few 10's of MeV show no promise at all.
- C. We offer two comments on specific details of the discussion of spallation-neutron sources. The tritium production in the Be moderator proposed for ING is a bonus, not a penalty. Tritium breeding is the heart of first-generation CTR concepts. Also, the penalty attributed to radionuclides produced in the spallation target is overstated, by an amount that remains to be explored. While it is true that roughly 7 radionuclides would be produced per 19 fission-product nuclei consumed, under the basic assumptions of transmutation waste-management only those new nuclides with half-lives greater than those of the waste to be transmuted can be counted as additional nuisances. The entire strategy is based on reducing the half lives of the waste nuclides.
- D. Spallation itself is a rather interesting form of transmutation. Studies with an intranuclear-cascade-plus-evaporation code show that the product nuclei may range from the target nucleus down to about half the original mass for proton or neutron energies of about 1 GeV. Unlike fission products, spallation products are predominantly on the neutron-poor side of the valley of stability, because the Coulomb barrier inhibits the evaporation of charged particles. Although most codes omit the fission channel, it is certain that direct bombardment of actinide targets above 1 GeV leads mostly to fission, which is the desired effect. If the proton energy is high enough, the yield is probably close to one fission per incident proton, plus still-uncalculated further transmutations from secondary particles. We shall outline below a brief calculation which suggests that this is too low a yield to be attractive.
- E. In computing the energy cost of spallation-induced transmutation, the PNL study reaches a lower limit by assuming that 100% of the input electrical power can be converted to beam power. Surprisingly, this is much less unrealistic than it seems. LAMPF produces 0.8 MW of beam power at about 15% overall efficiency, which could probably be raised to about 50% without stretching current technology unreasonably. LAMPF cost about \$4/W for the accelerator alone. This figure could be cut by a factor of 2 to 10 in mass production.
- F. The PNL study gives almost no consideration to direct spallation of radioactive waste by protons in the GeV range. The Japanese proposal to use 10-GeV protons on ^{137}Cs is mentioned briefly in an appendix. From Monte Carlo calculations at 0.8 GeV, we extrapolate a cost of about 50 MeV/nucleon emitted, which implies about 200 new nucleons per incident proton at 10 GeV. In an infinite Cs target, 100% of the neutrons and a substantial fraction of the protons will transmute ^{137}Cs nuclei into products which have either a shorter half life or a lower specific activity than the target. Radiative capture of either a neutron or a proton, and most reactions which emit one nucleon, lead directly or quickly to stable nuclei. Unfortunately, even an optimistic calculation shows that the 50 MeV/transmutation required is 1.5%

9.E.15

of the thermal power generated in the reactor which produced the Cs, or about 10% of the original power when realistic efficiencies are included. The terawatt nuclear power economy envisioned for the pinnacle of the fission-power age will generate about 1 kg/s of ^{137}Cs . For the 65-mA ion sources proposed for the Canadian Intense Neutron Generator the yield at 10 GeV would only be of the order of 1.5 kg/d, so that 5×10^4 accelerators would be needed.

VIII. EXOTIC FORMS OF TRANSMUTATION

We suggest that, for transmutation of ^{85}Kr by raising it to the shorter-lived excited state, inelastic scattering of 14-MeV neutrons from a CTR or other d-t device would have a much higher yield than Coulomb excitation.

IX. COMMENTS ON THE PROPOSED FUTURE PROGRAM

- A. We have remarked above that certain particular topics deserve a more thorough study. We feel that the present study has dismissed a few possibilities prematurely. Of these, accelerator-driven subcritical burning of a higher-actinide core and transmutation of partitioned fission products by underground explosions look particularly promising.
- B. It is our feeling that the time allotted to development of transport methods (Task 3.0) is grossly overestimated. Adequate codes already exist, and are estimated to require only a few man-years to collect and adapt. The time allocated for evaluation of nuclear data is more realistic. We question however, the omission of a modest program of new microscopic measurements, primarily on selected fission products, in tandem with some additional development of nuclear models for fission yields, decay systematics, and cross sections.

X. COMMENTS ON THE PRESENTATION

- A. Many of our contributors found the present draft rather repetitious. Most of them thought that at least parts of it give insufficient detail to make clear how much exploratory work has actually been done. If the detail of exposition matches the detail of investigation, parts of the study are too shallow to meet the weeding-out goal of Phase 1.
- B. The draft is replete with minor grammatical errors.
- C. We have found that the symbol MT is more likely to be interpreted as megaton than as metric ton. In the *Système Internationale* $1000 \text{ kg} = 1 \text{ t}$. The corresponding English unit is properly designated 1 short ton (sh tn). Similarly, there are other sloppy uses of units, such as KW for kW.

Review Comments on BNWL-B-301, Sec. 9: Advanced WasteManagement - Transmutation Processing

Dr. Robert L. Hellens

January 17, 1974

I enjoyed very much participating in the review of your draft report and in the discussions which took place subsequently in Seattle. The group, no doubt, came to some specific conclusions before the end of the meeting, and I gather that my comments during the discussion were recorded. In summary, I would like to emphasize several points.

1. Section 9 of your report does not go far enough in distinguishing which actinides and which parts of the chains are the main contributors to the long-term hazard. As Bernie Spinrad pointed out, it is suggestive that Claiborne's hazard curves showed a marked change with the amount of U and Pu separated from the waste stream. Unfortunately, this can be interpreted in a variety of ways and I think this needs to be spelled out much more clearly in your review of the ORNL work.
2. I believe that actinide recycle is feasible in power reactors but the anticipated gains need to be made more explicit in your report by the removal of some of the ambiguities which are mentioned in my notes and which were also emphasized by other members of the panel. I don't think that it has been shown that adequate gains could be made in hazard reduction in a practical application to warrant the appreciable effort which would be involved. If it should turn out to be an efficient approach, I would suggest that you devote more effort in the proposed program to the evaluation of methods of inserting actinides into a power reactor and to the evaluation of the changes in reactor operation, safety considerations and in reactor maintenance operations which would result.
3. The concern expressed by Claude Poncelet regarding the distribution of actinides among the number of power reactors could be substantially allayed if it can be shown that at the time of irradiation the hazards arising from the actinides are substantially smaller, even with recycle, than those normally arising from the

plutonium and fission products in the fuel. I suspect that the latter might well be the case and this would strengthen your proposed solution to the problem in an important area.

If I can be of any further assistance to you in connection with this study, particularly in those aspects which are concerned with power reactor utilization, please let me know and I will be more than happy to assist insofar as I can.

January 14, 1974

Time limitations have allowed me to read with care only those sections of the report that deal with the recycle of actinides and fission products through light-water reactors. Since the report views actinide recycle as the most promising form of waste hazard reduction by transmutation, and, in fact, speaks of this approach as both feasible and economic, concentration on this area seems warranted. The concept is not new, having been studied some years ago at BNL and more recently in ORNL, but the idea of using a power reactor as an irradiation facility for radioactive wastes is unconventional, to say the least, from the point of view of the reactor designer. I suspect that the Brookhaven concept grew up around the HFBR and envisioned a special purpose reactor devoted to waste hazard reduction; in such a reactor, the energy and waste balance (p. 9.3) would have to be very favorable and my intuitive feeling tends to favor looking for such an optimized "garbage incinerator" to avoid the dispersion of wastes among many utility-operated power reactors.

1. General Comments on Report Form

On the whole, I found the report well organized and relatively easy to read. There are some ambiguities in language which can be attributed to either haste or too much familiarity with the subject on the part of the writers, (for example, p. 9.30 on the distinctions between the 99.5% and 99.9% recovery curves of Claiborne is very unclear - and quite unnecessarily so). Beyond these normal difficulties, the report stops short of distinguishing between the various chemical

elements whose isotopes produce the various components of the net waste hazard. It may well be that further chemical separations, so that smaller but more toxic components could be subjected to higher flux levels, would not aid in reducing waste levels, but the question is, I believe, worthy of comment for the audience familiar with reactors but not with the hazards of actinide components; this latter audience comprises most reactor design experts.

The discussion of placement of the actinides in the core is limited to mixture with either uranium or mixed-oxide fuel. This approach would be considered with great reluctance by most reactor designers because of (i) the importance of achieving rated power and any change which could be prejudicial to the fuel performance could not be tolerated; (ii) inclusion of radioactive materials in the fuel, such as Pu, is expected for some time to increase the rod fabrication cost of $\sim 100\%$ until pellet manufacturing and fuel rod loading is automated; (iii) the reprocessing of fuel containing unusually high actinide content may well increase the costs for the large masses of fuel in the core to prohibitive levels. With these points in mind, one is led to consider the use of target rods composed, for example, of uranium tailings or inert diluents and high actinide loadings. Some appraisal of the core rod fraction necessary and the resulting actinide mass evolution (certainly larger than in the uniform dispersion case) would be helpful in providing a rough limiting case which would be viewed as more acceptable in not interfering with the fuel performance. It might also prove to produce so small a reduction in waste hazard that the approach through irradiation in power reactors could be eliminated from consideration.

2. Development Program

At first sight, the calendar length of the development program seems excessively long, 15 years, in view of the Figure 9.11, which shows that if the program started in 1975 we could expect to have 500 Te of actinide inventory on hand to deal with by the completion of the program. At that point the rate of construction of fission reactors should be nearing its maximum value and the

problem would seem to be well out of hand. I can accept the possibility that a reactor can reduce its own output of actinide waste without serious redesign of the core, added safety and maintenance problems, etc., but dealing with an initial inventory of this size in addition to recycle tends to prejudice the evaluation of the approach. It would help in evaluating the program plan to have this question of timing gone into in more careful detail.

As mentioned in the text, improved separation of the heavy metals from the reprocessing waste stream can result in substantial gains in the waste hazard reduction. Is enough time devoted to this question in the program? Perhaps the technology is already available, but no clue as to its state seems to be given in this volume of the report.

In the safety evaluation of the actinide recycle, the impact of the increased loading of radioactive heavy metals in the core on the inspection, maintenance, repair and possible carry-over into the steam side of the plant should be addressed under conditions of assumed failures of the target rods. It may be possible simply to show that compared to the hazards and/or contamination allowed from failed uranium or plutonium recycle rods, the actinide target rods can be expected to release so much less additional toxic material that the problem is second order either because of the number of rods, their low probability of failure, or because of their content. If the content is more hazardous than a Pu rod, one would have to consider a common tendency for failure of target rods and this would entail an extensive target rod test program which is expensive and lengthy. These comments are all predicated on a mode of operation in which heavy loadings of actinides are incorporated in a small number of displaced fuel or poison rod locations so that fuel and actinide rods are entirely separated.

In reviewing the Research and Development Program Schedule, Figure 9.13, it appears to me that the irradiation tests performed in the fifth year are too late and too brief to serve the required purpose of demonstrating that irradiation in typical flux levels is an efficacious means of reducing the actinide hazards by significant margins. The program should demonstrate that inventory

rise rates are indeed as estimated by calculations, as in Table 9.4. Thus, if it is at all feasible, the irradiations should start, say, in the second year of the program and continue for at least six years with typical fresh loadings of actinides being introduced along with the already irradiated material to support the theoretical contention of the approach to an equilibrium content in a fixed number of target rods in a given reactor.

During the irradiation program, the test rods should be measured for reactivity worth in a facility like the ARMF relative to boron glass standards, to ensure that no unforeseen problems of uncertainties in the actinide chain structure, decay constants, cross sections, competition between various (n, x) reactions, etc., are present. Measurements should be made at the end of each irradiation on a time schedule guided by calculations so that the observed reactivity worth can be translated back to the value applicable during the irradiation if short half-life components are present with appreciable content and cross sections. After the recycled actinides have been mixed with the fresh component, selected to represent the output from a processed uranium or Pu recycle batch, a second set of reactivity worth measurements needs to be made to provide a check for calculations of the isotopic and reactivity worth evolutions during the next cycle.

In my opinion, the reactivity worth measurements and accompanying calculations are vital to the success of the intent of the program if the use of power reactors is seriously considered as a source of neutrons for the irradiation and subsequent reduction of the actinide inventory. It would be quite difficult to persuade reactor vendors or utility operators to incorporate such recycle rods in a core without some assurance that the perturbation of the fuel enrichments, the cycle length, and refueling interval were well-known. In addition, the influence of such rods on core power pattern would need to be predicted with reasonable assurance to satisfy both operators and AEC regulatory staff that plant down-rating would not occur. It could be argued that the basic

measurements mentioned above would be made unnecessary by the demonstration irradiations scheduled to start in Year 12 and extending into Year 15. However, this phase is really one of final proof-testing, and by this point all questions of feasibility, efficiency, degree of hazard reduction, value of process, should have been disposed of at a much earlier stage.

There are two other types of measurements which I feel should be incorporated at an early stage in conjunction with the irradiation sequence mentioned above for the years of roughly 2 - 8. The first are measurements of the activities of the actinide pellets which contribute to the radioactive hazard at the beginning and end of each of irradiation cycles. Admittedly, these measurements would yield activities far different from those of concern in the long term, judging from Figure 9.C.14, but they could possibly provide data for confirmation that the calculated reductions shown in Figures 9.C.14 and 9.C.15 could be expected and that the process is efficient in hazard reduction. The possible value of such measurements should at least be examined. The second type of confirmatory measurement that would have fairly obvious advantages in showing that the transformations induced by neutrons and decay among the actinides are properly understood is mass spectrometer determinations of isotopic populations. The area of isotopic measurements with a large conglomeration of radioactive nuclei of closely spaced mass values may be difficult to deal with, but judging from the Yankee-Rowe data the measurements are possible and would provide additional substance to the actinide recycle data being generated in this program. Because of the recycle the population of remote isotopes, which do not figure prominently in uranium or even Pu recycle cores, should become large and the verification of the predictions concerning the content and reactivity worth of these constituents should be a primary goal of the development program outlined in this report.

Having mentioned these additions to the proposed development program, I recognize that they could be very costly extras and would require more extensive justification for inclusion than given above. It seems to me that the

program would be proceeding on a rather weak foundation without support of an experimental nature to show beyond reasonable doubt that the theoretical expectations would be achieved in practice.

Before discussing some reservations I have on the reactor design implications of actinide recycle, I would like to reinforce the comments made in the report (p. 9.54) concerning the development of strategies for insertion of recycled actinides into the reactor without appreciably degrading the power-producing capability of the plant. Of course, the lowest actinide level should be achieved in a uniform mixture of fuel and waste. If this configuration can be achieved in an economical fashion at all, it will probably have to be deferred until the time when all fabrication processes are remote and when contamination of the production line by highly toxic material can be accepted. This has obvious impact on the conditions for maintenance, repair, inspection, clean-up, etc., operation required on the production line.

Since the bulk of fuel fabrication until 1990 will probably be uranium, there is not a high incentive for complete automation of the fuel production line at a much earlier stage, unless it can be shown to be cheaper. Consequently, an approach using target rods in the unused control rod locations of PWR cores is most attractive. The actinide content of the rods will need to be higher, no fuel can be included since little cooling is provided in these channels, and the depletion of actinides will be reduced due to flux self-shielding. The whole fuel management pattern of the core will probably be strongly affected by the presence of the target rods and their change in worth during irradiation can roll the power enough during one fuel cycle to degrade the plant operation unless their characteristics are quite accurately predictable. The actinide content from roughly 50,000 - 60,000 fuel rods will have to be inserted into roughly 2000 rods of slightly smaller diameter in a (rod cluster control) RCC core or into about 400 rods of larger diameter in a Combustion Engineering PWR core. Thus, the concentration of actinides should be increased by about a factor of 40 for each

fuel batch contribution when transferred from the fuel to the target rods. The increase sounds large, but when the relatively small reactivity worth of the actinides, other than U and Pu, is recalled, one suspects that the target rods would not be very black, particularly if the actinide oxides are mixed with a low cross section diluent such as Al_2O_3 or MgO . As later actinide wastes are cycled into the target rods, the blackness will increase and flux depression will occur, with a corresponding loss in the efficiency of recycle to reduce the waste hazard level. How far these various effects will change the results of earlier calculations will need to be evaluated in detailed fuel management calculations in at least two-dimension to see whether stable fuel management patterns can be found with acceptable power distributions.

One further point should be mentioned in connection with the large number of 1200 MWe plants expected to be commissioned in the early 1980's. If the proposed recycle program is well along, these reactors could start reducing the backlog of wastes in first-core loadings where no previous inventory from the particular reactor itself needs to be included. The difficulty is that data from the proposed development program would need to be available to the core designers well in advance of the start of waste recycle so that the planned loadings can be reflected in uranium enrichment orders and in fuel loading studies. Once again it strikes me that the program, as outlined, is too slow to attack the waste build-up during the time when large quantities of neutron flux are becoming increasingly available and the buildup of waste is proceeding at an accelerating rate.

The last comment on the development program, as outlined in the report, is that the most serious problem of actinide production has not been specifically noted. This, I believe, occurs in the plutonium recycle mode of fueling for thermal reactors since the contents of Am and Cm are substantially increased by the repeated irradiation of the various Pu isotopes. It would be interesting to see Curves 9.8 and 9.9 redrawn for a reactor operating in the SGR (self-generating recycle) mode--perhaps these have already been generated but have

not been included in the report. If the uranium and SGR modes do indeed have different actinide hazard consequences, the measurements program connected with the experimental sequential irradiations may have to be substantially expanded to demonstrate that the actinide recycle scheme does, in fact, provide sufficient reduction to justify its full development and use in power reactors.

3. Comments on Appendix C

This appendix provides some additional detail to that given in earlier sections of the report, but not quite enough to be as useful in forming an opinion as it might be. The PNL inventories, using only 10% of the fuel rods rather than all, are useful in showing the effects of higher actinide densities in the rods and deviations from Claiborne's calculations. However, the choice of fuel enrichments, both uranium and later plutonium, to provide the same energy output from a target rod as from a normal fuel rod is not a particularly good or valid way of estimating enrichment penalties for recycle in a full core. Nor is the idea, as I have mentioned before, of mixing fuel with recycle waste a very desirable choice.

The section (p. 9.C.23) dealing with the recycle of actinides with plutonium fuel is not very clear in the presentation of the recycle schemes, either for the plutonium or for the actinides. However, the weakest point is that the primary emphasis seems to be on the cost increment of waste recycle, rather than on the very large increase in the actinide hazard associated with Pu recycle. Seeing this increase in Table C8, one wonders first if this is representative of the entire core or only the actinide loaded Pu rods and, second, what impact these very large increases would have on the amplitude and shape of the hazard curves 9.8 and 9.9. To my mind, an evaluation of this sort could have an important bearing on the choice of this program as a vital part of the waste management scheme.

In early calculations of plutonium recycle, we have observed the marked increase of the Am and Cm isotopes on the reactivity loss at end of cycle. The

calculations were primarily aimed at evaluations of some of the low mass actinide concentrations. The attached table of the reactivity worth of various actinides from cell calculations, though old, is probably indicative of the magnitudes of the changes in reactivity worth to be expected as the mode of refueling is changed. It should be kept in mind that these calculations assume complete removal of all actinides other than U and Pu isotopes in recycling, so that the reactivity loss due to Am and Cm are due to buildup only in the cycle in question. I gather from the tables in the report that very substantial further reactivity losses could be anticipated in later stages of plutonium recycle if the actinides were to be recycled.

4. Conclusions

The comments I have made have largely been critical in tone with the intent of improving the perspective provided by this proposal. The importance of actinide waste reduction needs to be emphasized and it appears that it may be most valuable in dealing with wastes generated in the late 1970's through 1990 and beyond by plutonium recycle in thermal reactors. The proposed development program seems minimal, particularly in the area of the measurements which would demonstrate the validity of the approach.

If I were making the proposal, I would suggest two phases. A short first phase would be defined to confirm the need, feasibility and efficiency of actinide recycle in power stations. If the first phase were successful and compelling, the second phase should be considerably more substantial in some of the areas I have outlined above than that now proposed. If the problem is serious and the solution effective, the job should be thoroughly done.

REACTIVITY WORTH OF SOME HEAVY ISOTOPES

at 33,000 Mwd/te

<u>Isotope</u>	<u>3.3 w/o UO₂</u>	<u>1st Gen. SGR</u>	<u>Equil. SGR</u>
Np-237	0.51	0.14	0.15
Pu-238	0.15	0.03	0.03
Am-241	0.12	0.71	0.89
Am-242	0.01	0.10	0.13
Am-243	0.09	0.89	1.95
Cm-244	0.01	0.19	0.41
Cm-245	0.01	0.10	0.22
TOTAL	0.90	2.16	3.78
U-236	1.04	0.20	0.22
Pu-240	6.27	12.51	14.05
Pu-242	0.34	1.64	3.37

COMMENTS ON

Advanced Waste Management Studies
High Level Waste Disposal Alternatives
Section 9: Transmutation Processing
BNWL-B-301, August 1973

I. General Comments:

1. Accelerators, Explosives, and CTR's:

I agree with your dismal prognosis for accelerators and explosives as potential waste processing units. The prospects of a CTR waste burner appear promising but as yet premature depending upon its own technological break through. In this I do agree that at present a continuous though modest interest be maintained in CTR's to provide coordinated development of both waste management and CTR technologies.

2. Feasibility and Order of Merit:

It seems clear what four factors comprise the feasibility criteria; however, it is not so clear what elements comprise the order of merit criteria to be used in comparing transmutation schemes with each other and with other strategies described on pages iii and iv. It is assumed that Section 9.4 through 9.6 generally make up the order of merit criteria. If this assumption is valid, the following suggestions are offered:

a. Hazard Index: The reduction in long-term waste toxicity, as measured by the hazard index, using nuclear transmutation will be achieved by improved technology and a more complex fuel cycle. To appropriately use this toxicity measurement, it should properly be bounded on either end. On one extreme, as is pointed out, are the untreated high-level wastes. On the lower extreme no such limit presently exists. Some reasonable lower limit should be identified, either technological or economic. Thus bounded,

it makes more meaningful your results of a two-order of magnitude improvement in long-term waste toxicity costing about 0.2 mills per KWHe for actinide recycle in a LWR.

b. Environmental Impact: Two impacts of actinide recycle not discussed in detail but deserve more attention are dilution of the high-level wastes due to chemical processing, and changes in the actinide waste forms. The former presumably is discussed in Section 4 (Potential for Waste Partitioning), while the latter rightfully seems appropriate in this present section. Current estimates by ORNL (Sources of Transuranium Solid Waste and their Influence on the Proposed National Radioactive Waste Repository, ORNL-TM-3277, February 2, 1971) indicate about 0.5 w/o of the heavy metal will be lost at both fuel preparation and manufacturing facilities (also the value assumed in Table C11, p 9.c.31). These losses reappear predominantly as low-level alpha contaminated wastes currently termed alpha-wastes by the AEC. Because of the higher levels of alpha and neutron activity of the trans-plutonium isotopes, these low-level wastes will no longer be as benign and easily handled as a purely plutonium contaminated waste.

3. Hazard Index:

The use of the hazard index as defined is a most convenient measure of waste toxicity; however, it is disproportionately sensitive to the fraction of Np237 alpha decay chain present in the waste. Only two of the eleven radioactive daughters of Np237 have specified RCG's; the remainder assume the default values assigned by 10CFR20. This circumstance is not as extreme for the other three alpha decay chains. Future research could be directed towards resolving this 'imbalance' but for the nonce some clarification is needed.

4. Useful Actinides:

As conceived and if implemented, routine actinide recycle might emerge circa 1990. During the interim, it is projected that Np237 (as a progenitor of Pu238), and possibly the curium and californium isotopes, may become commercially valuable materials. If these prognostications are reasonably true, commercial use of the extractants might partially off-set fuel preparation, manufacturing, and reprocessing costs; reduce neutron radiation difficulties; and modify to some extent core physics and in-core fuel management. This aspect of actinide commercialism certainly does not lessen the ultimate requirement for disposal of the 'useful' actinides nor should it be made pivotal in determining physical feasibility of actinide recycle in fission reactors, but it is a possibility worth investigating.

5. Waste Partitioning:

The technological-economic limits of actinide-fission product partitioning should be of major concern and one of the first milestones to the Recycle-Transmutation Program. Some reasonable lower limit (for extraction) should be delineated (see 1.a. above) from which competing options can be judged. My studies indicate that long-term toxicity reduction is not as optimistic when applied to LMFBR fuels. This circumstance obtains because of the more plentiful quantities of trans-plutonium isotopes generated in the fast reactor coupled with a poorer extraction efficiency for these isotopes (about 0.99) with current technology. However, waste partitioning is the only currently feasible option expanding alternative and should be considered on its own merits.

6. System Model

There is a need to look at waste management on a total system basis. It is quite possible that less than optimal solutions can obtain if too narrow a view of the fuel cycle is framed.

II. Specific Comments:

<u>Page</u>	<u>Comment</u>
9.12 (3-6)*	Heretofore, workers in the field have estimated the final disposal cost for high-level wastes at but a few percent of the nuclear fuel cycle cost. Thusly, the exact strategy and timing
Note: '*' (lines from top of page inclusive)	
	seemed not as important as it now appears if waste management costs approach 15-20 percent (your estimate) of the fuel cycle cost or as much as 100 percent if more exotic means are needed.
9.27 (18-20)	As is rightly observed, there is incomplete/inconsistent data at present to definitively predict recycle in fast reactors. This shortcoming, as is pointed out in Section 9.3.2, should receive major emphasis in future actinide-recycle research programs.
9.32 (Figure 9.8)	There is some question if any advantage exists in the short-term by recycling actinides as Figure 9.8 implies. If the fission product hazard for the comparable time period is superimposed, it dominates. Additionally the increased handling problems and neutron generating alpha-wastes makes such near-term savings moot.
9.70 (11-13)	Space disposal competes with nuclear transmutation as a 'now' method of waste disposal, albeit more costly.
9.77 & 9.78	It is not entirely clear why the actinide release and attendant environmental insult from a fabrication plant should decrease so dramatically over the current U-Pu fabrication facility. The bulk of the fuel would remain the standard U-Pu fuel, while

9.E.31

the added transuranics tend to be more toxic per unit weight, thereby exacerbating the problem.

9.82
(1-3)
(23-25)
9.83
(1-9)

The impact of extracting the actinides from the high-level wastes on transportation appear (in addition to your discussion):

a. Although the actual waste volume/weight will decrease with actinide extraction, the waste-inert material volume/weight most possibly will increase. This is due to the more difficult chemical processing necessary to make the actinide-fission product cut.

b. The reduction of toxicity due to extracting the actinides from the high-level wastes is almost inconsequential vis-a-vis the fission products on the near-term, in particular during transportation of the wastes. Additionally the neutron source strength does not vary appreciably between the no-recycle and recycle high-level wastes due to the isotope composition of the actinides.

c. Recycling actinides will result in a reduction of actinides in the solidified high-level wastes but trace amounts of this contaminant will be present in the fuel manufacturers' solid alpha-wastes. Previously considered alpha-wastes were inconsequential neutron sources (e.g. 3440 n/sec-gm Pu238). Conversely, the heavier trans-plutonium isotopes are extremely active alpha and neutron emitters (e.g. 2.3×10^7 n/sec-gm and 3320 (α) Ci/gm Cm242). High energy alpha emitters are problematic in handling because of the (α ,n) reactions (note: (α ,n) reactions in oxides are of the order of 0.5×10^4 n/sec-Ci; J.P. Vaane, "Hazards Connected with the Handling of Transuranium Elements

and Methods used for the Protection of Personnel", Actinides Review, 1, 1969, pp 337-370). Presently, alpha-wastes can be shipped using DOT approved containers of negligible shielding. With actinide recycle, this may not be possible.

9.82
(4-17)

The concept of a cycle is not entirely clear. It is inferred that a cycle extends from one fueling to the next, much as is the practice in nuclear fuel management. Claiborne uses the total irradiation period as a cycle (three refueling periods for a PWR). Since actinide recycle takes so very long to achieve equilibrium, it seems some distinction should be made.

9.85
(7-13)

The comments of the second paragraph appear inconsistent with Tables 9.10 and 9.11 (see previous comment).

9.A.7

The term 'F' is not clearly defined in equation A11.

9.C.12 & .16

It is not clear what the heavy lines indicate on the tables, possibly quasi-equilibrium.

9.c.22

Figure C5 has no scale for the abscissa.

9.E.33

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January 8, 1974

Dr. Bowen R. Leonard, Jr.
Battelle Northwest
Richland, Washington 99352

Dear Bo:

I approached section 9 of BNWL-B-301 with some hesitancy since it is a massive document. After going through it several times, I find myself comfortable with it. It strikes me as a well done study which should lead to easy management decisions. It seems to me though, that those decisions must be influenced strongly by sections 1 through 8 which I have not seen.

Let me first comment on transmutation schemes other than reactor processing of the Actinides. I can find no fault with Appendix A except that I think most energy balance and transmutation rates are stated optimistically. I think that your conclusions could be strengthened if there were any need to strengthen them. On page 9.20 however, the factor of 25 obtained by comparing 200 MeV per fission with 5000 MeV per proton produced transmutation is overstated if one limits attention to a particular species such as ^{137}Cs . The appropriate first number should then be 900 MeV/ ^{137}Cs (page 9.A.32) giving a ratio closer to 5.

Since you discuss coulomb excitation using ^{85}Kr as an example, it might be worth mentioning that neutron inelastic excitation of the same state by a fission neutron spectrum might be more feasible. One can manage with $\lambda = 3$ neutrons, and in either a fission spectrum or a CTR spectrum there should be plenty of those. Also, have you considered the possibility that neutron proximity might significantly alter beta decay rates by angular momentum coupling? (Since you consider Nobel Prize level schemes like GRASERS I thought that I should slip that in.)

I wouldn't be surprised if someone eventually defends the proposition that transmutation of hazardous materials in large volume can only be done

Dr. Bowen R. Leonard, Jr.
January 8, 1974
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safely with neutral radiation. The surface to volume ratio of material to be processed, and protection of cladding integrity both point to that conclusion.

I will pass by Appendix B without comment. Appendix D is incomplete because much more nuclear data, and a better defined CTR is needed to make that section complete. I would regard Appendix D as an interesting possibility which should not be used to distract either the people working on that program or the general public. Everyone "knows" that CTRs will be "clean". Don't spoil that illusion. I think that I would worry some about a CTR loaded with 50 kg of ^{137}Cs .

Now on to the Actinides. I think that the scale of the problem is well stated by Table C 6, p. 9.C.18. The actinide of most significance is clearly ^{239}Pu and it is also one which will persist for a time which is long compared to human records. We clearly have accepted the long term hazard associated with storage of ^{239}Pu in our weapons inventory. If we accept the one hazard, does it make all that much sense to devote a rather large effort to eliminate a small part of the total long term hazard? (I suspect it does.) Metallic storage of actinides like weapons are stored should be possible. The heat burden from actinides is probably also small enough to allow salt bed disposal of at least them. The energy balance arguments aren't very convincing for two reasons: 1) ^{238}U can't be worse than say ^{237}Np as a breeding material, 2) from a reactor operations point of view, more time would be lost from a jam on loading an actinide rod than would be lost on a jam with a clean rod. In summary, if I had to make the management decision, before I would recommend proceeding with this program I would spend a large amount of time with sections 1 through 8 of the report.

Sincerely yours,

Harlan

Harlan W. Lefevre
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HWL/js

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January 7, 1974

Mr. B. R. Leonard, Jr.
Staff Scientist
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Dear Mr. Leonard:

I greatly appreciate this opportunity to review the Battelle study on high level radioactive waste transmutation as documented in the draft report, "Advanced Waste Management Studies - High Level Waste Disposal Alternatives; Section 9: Transmutation Processing". The management of long-lived radioactive wastes from fission nuclear power plants is a crucial problem and transmutation offers the only possibility of elimination as opposed to disposal. I therefore highly commend you and your staff for your efforts in this area.

I concur with the study's conclusions regarding the lack of technical feasibility for transmutation processes involving the use of accelerators or nuclear explosives. The technical feasibility of burning actinides and long-lived fission products in a fusion system appears to be well established. However, the concept of burning the wastes in central station CTR's seems to me very objectionable, since it would introduce in a relatively clean system high level and long-lived radioactive material. This would negate one of the major advantages of fusion systems and could lead to substantial public health hazards. I would suggest that fusion systems be designed for the sole purpose of burning high level radioactive wastes, thus leading to the possibility of siting at the reprocessing plant and to the possibility of achieving engineering feasibility independent of CTR power plant engineering feasibility.

I have strong reservations regarding the desirability and real feasibility of actinide recycle in central station fission power reactors. The benefits are limited and these do not appear to outweigh the added risks accruing to plant operation, as well as the increased difficulties in fuel fabrication and fuel reprocessing, which have real influences of plant reliability and public health hazards.

Specific and detailed comments on the draft report follow.

1. The study only considers existing or anticipated devices, whose major purposes are other than waste transmutation. This probably limits both feasibility and neutron source characteristics. For example, a fusion facility could be developed for the specific purpose of waste transmutation. Research and development programs should be recommended in this area. Note that engineering feasibility of such a facility could be reached sooner or independent of CTR power plant feasibility.

2. The conclusions drawn in the study favor those processes, such as actinide recycle in fission reactors, that have relatively immediate, although limited, applications. More emphasis should be placed at this time on more comprehensive and effective long-range methods.

3. The concept of burning highly radioactive wastes in central station power plants is questionable. It may be better, as a matter of principle, to decouple radioactive waste transmutation from electrical power generation. That is, the neutrons required for transmutation should not necessarily have to come from processes whose function is to generate electrical power. The public and the power industry might much more readily accept a concept where radioactive wastes would be burned in a facility located at the reprocessing plant, apart from electrical power generation.

4. The recycle of actinides in fission power reactors may prove unacceptable to the electric utility industry and perhaps the fuel manufacturers. This is essentially based on a cost-benefit argument.

- The benefits of actinide recycle in fission reactors are limited. The arguments advanced in the study to explain the non-feasibility of burning long-lived fission products in fission reactors, that is if they could be burned effectively this would imply they would burn in situ at the time of production, are partly applicable to actinides. The study shows that the actinides reach an equilibrium in a recycle scheme after about 15 years. Essentially, given that the neutron fluence nvt is the parameter of importance in transmutation processes, the concept here is to stretch the time of exposure t . However, as depicted in Figures 9.8, 9.9, and 9.11 of the report, the reduction in the hazard index is about a factor of 10, whereas many orders of magnitude are probably necessary to cause a significant hazard reduction. That is, with a continuing increase in central station fission power plants, the total hazard coming from actinides would still be very significant even with actinide recycle.

Mr. B. R. Leonard
Battelle Northwest Laboratories

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- With actinide recycle, the actinide inventory at any given time in a power reactor is increased. The necessity for remote fabrication of the recycled fuel rods implies a probable decrease in quality control. These considerations imply additional burdens on both the safety and reliability of the plants, and the prospect of severe licensing problems. Given the limited benefits, these additional problems may make actinide recycle very unappealing to the electric utility industry. Note that the concepts proposed in the study would burden the entire fission reactor economy with actinide recycle.
- Actinide recycle would impose relatively severe manufacturing, logistical, and quality control problems on the fuel manufacturing industry. It may be difficult to justify the cost of special actinide recycle fabrication plants.

5. Considering long-range goals, it may be necessary to rely on more than one process for effective transmutation. For example, the wastes could be exposed sequentially to a number of different neutron spectra.

6. The recycle of actinides in uranium and/or plutonium bearing fuel rods has serious implications in terms of fuel manufacturing, quality control and reliability, safety and operations. Fuel fabrication research and development requirements may be much more extensive than envisioned in the study. An alternate concept would be to recycle the actinides as specially fabricated poison rods that could be utilized in the same way as the burnable poison rods currently in use in LWR's. This would minimize fabrication cost, increase quality control, and alleviate safety, reliability and licensing concerns. Another concept would be to locate the actinide targets in the reflector of LWR's, which experiences a very high thermal neutron flux, and essentially utilize leakage neutrons. The feasibility of locating targets in the reflector space is, however, open to question.

7. The energy balance argument used in the discussion of accelerator devices on pages 9.20, 9.23 and 9.24 of the report, appear incorrect. The energy released in the power reactor where the specific waste products to be transmuted were created is more than 200 Mev/waste. That is, the specific fission product yields and the neutron balance leading to actinide production must be considered.

8. I fail to fully understand the explanation for the differences between Clairborne's results and the PNL results as far as the actinide concentrations in the recycle scheme are concerned. The actinides are present in relatively small quantities in the fuel and I would not expect spatial self-shielding to be very significant. Certainly it cannot explain a difference of 0.05 in k_{∞} of the fuel. Are the actinides even worth this much? The difference in Pu concentrations (~5%) are also significant.

Mr. B. R. Leonard
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9. The enrichment penalty in actinide recycle first mentioned on page 9.34 should be worded as a 4% increase in enrichment, and not a 4% additional U-235 content. Note that this implies a roughly proportional increase in kw/ft. Also, a 4% increase in U-235 enrichment implies a correspondingly larger increase in total additional uranium requirement, as opposed to the comment made on page 9.83 of the report.

10. In recycling actinides in a few rods as opposed to every rod, one must consider the effect on local peaking factor in the fuel assemblies.

11. The results for actinide and fission product transmutation in fusion systems appear very promising and significant. The reduction by a factor of 3 for Cs-137 (page 9.38) is probably significant, given the relatively short half-life of Cs-137.

12. The strategy of recycling actinides in fission power plants until fusion power plants come on line, and then to feed actinides and selected long-lived fission products to the fusion plants, appears questionable. Since the burning of actinides in fission plants is only partial and since long-lived fission products must be stored in any event, why not also store the actinides and await the development of fusion systems, thus avoiding the problems attendant to recycling in fission power plants.

13. The total cost for the proposed R&D program to develop actinide recycle in fission reactors may be much higher than the \$20 million arrived at in the study. Major cost may accrue to the fuel fabrication and re-processing technology R&D.

14. Will government subsidies to the electric utilities have to be considered to make up for the economic cost penalty that actinide recycle in fission power plants would imply, particularly during the demonstration stages?

15. With reference to the research and development requirements, I question the need for methods development. Existing methods are probably more than adequate. On the other hand, uncertainty and/or lack of cross section data is probably a much more important consideration. A substantial effort would therefore be required in the area of nuclear data. Verification will not be an easy task, given that the existing data on actinide isotopes from irradiated fuel measurements are not that good and plentiful.

16. The neutronics and fuel management analyses will have to consider all the various engineering design and safety factors, including: effect on kw/ft and hot channel factors; effect on shutdown margin; effect on reactivity coefficients; effect on control rod worth; effect on accident analyses. It is likely that engineering design and safety considerations would dictate the recycle strategy, and not fuel cycle or management considerations.

17. The experiments briefly discussed on page 9.53 of the report in relation to the research and development requirements are not defined or specified in the study. Neutronics experiments are probably not required or

Mr. B. R. Leonard
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meaningful. Experiments to directly measure basic cross section data, and irradiation tests in reactor during the demonstration program, are probably the more important requirements. It should be noted that irradiation tests in operating fission power plants would require extensive and costly design and safety analyses and licensing procedures.

18. Safety may be much more of an issue in actinide recycle than the study makes it to be. In particular, as opposed to the statement made on page 9.85 of the report, it is felt that the risk associated with actinide recycle in fuel elements of central station fission plants would be greater than that associated with normal reactor operation.

19. With reference to the comment made at the bottom of page 9.60 of the report, there is currently no Pu recycle in LWR's, safety and licensing issues having yet to be resolved.

20. With reference to the fuel cycle strategies discussed in section 9.5 on Capital and Operating Costs, the differentiation between fuel rod fabrication and fuel assembly fabrication does not come through. That is assemblies, not fuel rods, are loaded into the core, and individual rods cannot be removed from spent fuel assemblies except at the reprocessing plant. Thus, if actinide bearing fuel rods are fabricated at the reprocessing plant site, the entire fuel assembly may have to be fabricated there. Also, it would not be feasible to expose actinide bearing fuel rods longer than other fuel rods in the assembly in which they are inserted. Individual rods could not be shipped from the reprocessing plant to the power plant, since they must be fabricated into fuel assemblies.

21. The economics analyses appear favorable. However, there may be hidden cost in added safety and licensing problems and in a potential decrease in plant reliability.

22. It is suggested in the study that the fuel fabrication and reprocessing plants be located at the same site. Although there is a great deal of merit in this concept, it is questionable whether the limited benefits accruing from actinide recycle would justify modifications of current industry plans for both fuel fabrication and reprocessing plants, given the high capital investments involved.

23. As indicated in the study, the effects of actinide recycle on accidents in fuel processing plants are probably minimal.

24. Transmutation of long-lived fission products in fission reactors indeed appears to be non-feasible.

25. The rationale used on top of page 9.C.23 is not clear. The volume-averaged core burnup at the end of a cycle is about 22,000 MWD/MT. The replacement of the 33,000 MWD/MT fuel with fresh fuel increases the core k_{eff} to about 1.1, which is higher than the value when the core has an average exposure of 22,000 MWD/MT ($k_{\infty} = 1.026$ in the study).

Mr. B. R. Leonard
Battelle Northwest Laboratories

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26. Recycle of actinides in plutonium bearing rods would probably lead to engineering design and safety problems more severe than for recycle in normal UO_2 fuel.

27. The use of novel fuel designs, such as the MgO fuel rods proposed in the study, is questionable because of the many potential problems that can be expected with the use of novel fuel types.

28. The necessity to rely on one or more core regions which drive regions of the core with low k_{∞} imply very poor power distributions, particularly in PWR's.

29. The concept of burning fission reactor radioactive wastes in a central station CTR plant is not appealing because it introduces actinides and long-lived fission products in a system which otherwise is free of these materials. It would make more sense to develop special facilities based on the fusion process to burn fission reactor wastes.

30. The very encouraging results obtained with the fusion processes indicate that major R&D efforts should be concentrated on these processes, as opposed to fission processes.

I look forward to discussing these matters more fully with you and your colleagues and the other reviewers.

Sincerely yours,

Claude G. Poncelet, Chairman
Nuclear Science and Engineering

CGP:GB

HIGH-LEVEL WASTE DISPOSAL ALTERNATIVES

A Review by Bernard I. Spinrad

GENERAL COMMENTS

- 1) As is recognized in the study, the case for transmutation processing rests primarily on the ability to partition nuclear wastes. This seems to be self-evident, but the study provides adequate objective confirmation.
- 2) The Kubo and Rose studies cited, and the draft Section 4 of the BNWL study, make a sufficient case for partitioning as a valid technique and a desirable one. The major advantage on the one hand is the possibility of separating out those fission products of significant activity whose half-lives are less than about 50 years. Particularly in the Kubo and Rose study, it is shown that a variety of techniques which have been proposed and criticized for ultimate disposal of mixed wastes can be successfully justified for 1000-year disposal, during which time the radioactivities of this short-half-life set of nuclides will decay to harmless levels.
- 3) Given the assumption that the results of partitioning and disposal of "decades-long" half-life fission products can be achieved, transmutation of the actinides becomes extremely important as an indicated technique for disposal of the next most dangerous set of by-products of fission power.
- 4) Although not part of the review of Section 9, my opinion is that Section 4 is unduly timid with regard to both the potential of various chemical processes for realizing high degrees of separation, and the costs of so

doing. The conservatism seems to arise from the fact that the potential and economics of such processing seems to be based on past and current chemical-engineering practice, rather than on that basic chemistry which permits future engineering to be developed. For processes to be developed, this line of thought does not seem appropriate.

5) I would guess that some further qualitative differences exist between the effect on the biosphere of medium-lived nuclides such as most of the actinides, and of long-lived nuclides among the fission products such as Tc and I¹²⁹. At first glance, I am somewhat surprised at the concern over these latter, and would think that their effects would be very hard to discern against background radiation effects. An in-depth study as to whether concern over these long-lived nuclides is justified would seem to me appropriate to any follow-on to the current project. At a minimum, studies defining that value of half-life which is a "threshold for concern" ought to be initiated. Clearly, U-235 @ 7×10^8 yr. is a "safe" radioactivity, U-236 @ 2.4×10^7 yr. is "safe" in small quantities and U-233 @ 1.6×10^5 yr. is "unsafe" in large amounts, even setting aside criticality hazards.

6) Transmutation of actinides within the fission cycle is exactly equivalent to recycle of actinides. I do not think that the term "transmutation" improves the public image of the process, which factor is a significant one if "recycle" is an equally valid describing term.

7) In line with my comment #4 above, I am uneasy about accepting current practice of permitting ca. 0.5% of the Pu to be lost to waste during the reprocessing cycle. I think recovery can be improved without major cost penalty, and in this case there is the additional incentive of diminishing

the possibility of diversion of significant quantities of Pu from the MUF in the waste stream. Thus, I think it's worth a lot more R & D. Even though the current standard 200-fold reduction in Pu content seems to be sufficient to make Am and Cm the major "villains" among the remaining actinide content, I think that any large total amount of Pu sent to ultimate waste would be unacceptable for a large nuclear industry.

8) As a reactor physicist, I am concerned with the elaboration of reactor physics projects in the suggested development program for actinide recycle. There is a lot of room for improvement of cross-section data, but beyond that existing methods and computer programs are adequate for answering such general questions as may be posed (such as, for example, the order of magnitude of cost penalties to the nuclear fuel cycle which actinide recycle would bring about), while detailed questions about particular recycle schemes are best answerable within the context of specific scheme proposals.

9) There are policy questions as to what administrative or legal steps would be effective and appropriate in order to achieve actinide recycle within our nuclear industrial framework. While not a part of the present BNWL study, they should be addressed by AEC in any future follow-on studies.

SPECIFIC COMMENTS ON SECTION 9

9.1 - In 9.1.2, it is assumed in the text and illustrated in Figs. 9.2 and 9.3 that partitioning is to be performed after "waste" has been rejected from the chemical reprocessing plant. I have registered skepticism as to the ultimate validity of this assumption. In any case, the assumption is not necessary to the conclusions of this subsection, and ought therefore be edited out.

9.E.44

The part of 9.1.2 labeled "Transmutation Cycles" is unnumbered. I believe it is the most salient part of the subsection, and as such ought to be put into the front of 9.1 as a numbered subsection (perhaps 9.1.1).

9.2 - On page 9.30, the text does not indicate whether Claiborne's strategy recovers residual U and Pu from the waste stream to the same recovery factor from that stream as the other actinides. This uncertainty beclouds subsequent discussion. In fact, I interpret Fig. 9.9 to suggest that its bottom curve is dominated by the hazard of the 0.5% plutonium not removed.

On page 9.37, I believe that the last two sentences of 9.2.3 could be much more optimistically and positively worded. There is no question in my mind but that fast reactors are much more efficient actinide burners than are thermal reactors. Indeed, fast breeders would produce far less actinide by-product (at least, trans-plutonic) than thermal reactors, a matter which could be ultimately of great economic advantage to them.

Section 9.2.4 seems to me to be correct as regards fission products, but potentially very misleading as regards actinides. The reason is that the very high neutron fluxes cited for CTR devices are not much more effective for actinide destruction than are the much lower fluxes in fission reactors. The important matter is to reduce burnup half-lives to the order of a year or less. The use of "hazard half-life" as a relative figure of merit gives inordinate weight, for the actinides, to processes which burn them up in unnecessarily short periods.

In section 9.2.5, I believe care should be taken not to assume that CTR is an established technology. I would add appropriate qualifying comments to

the references to CTR if the subsection remains organized as in the draft. Better would be to segregate the fission and fusion recycle schemes into separate subsections, indicating the independent strategies.

The sentence beginning on the bottom of page 9.39 contains an editorial mistake.

In Fig. 9.10, why are long-lived FP to be returned to the fuel cycle?

Fig. 9.12 is a good illustration for the "mixed" cycle, but the fact that it is such a cycle, or the words "CTR" or "fusion" should be incorporated into title or legend. Another figure should be added to Section 9.2 to illustrate the workings of a pure CTR economy, cutting into the by-products of a preceding fission economy.

Summary Comments on 9.2

(a) It is not clear what is the fate of that portion of the Pu not recovered in reprocessing, during that part of the cycle in which "actinides" are recovered.

(b) A discussion of the improvement in actinide burnup due to the use of fast, rather than thermal, reactors as burners should be included. The gains are demonstrably substantial, and, even without detailed calculations, the generalities of fast-neutron cross-sections permit a definite and positive statement.

(c) Discussion of the potential usefulness of CTR techniques should be introduced by qualifications throughout. Such machines do not exist, and our estimate of how we would cycle wastes through them is consequently

vastly vaguer than is the case for fission machines. Relative reduction of hazard half-life is a poor figure of merit to employ.

9.3 - In Section 9.3.2, I believe that the data evaluation task should be clearly divided into separate subtasks, one for actinides and one for FP. I believe it would be useful and appropriate to recognize that many aspects of the task are already under way in other AEC-supported programs, that coordination with these programs is needed, and that with such coordination the work to be done is likely to be not so enormous. The appropriate AEC programs are, of course, those of the Reactor and Research Divisions.

In Section 9.3.3, I recommend the title "System Modeling" over "Methods Development". It is more correct, and does not over-generalize the task which is contemplated.

I do not think that the task of "Neutronics Analysis" described in 9.3.4 should take as long as scheduled, nor should it be very costly. Given cross-sections, all the codes which have been validated for reactor design should be routinely useful.

I do not think that the review of actinide production data should be included in the task described under 9.3.5. That review seems to me to be an important subtask under "Data Evaluation", being the major validation of the cross-sections adopted.

Section 9.3.6 describes a number of activities in "catch-all" fashion. Many of these are not independent jobs, but add-ons to programs which will, for other reasons, be on-going in both industry and government. This thought, and the cost savings which may be achieved by recognizing it, ought to be expressed.

Section 9.3.8 has questions associated with it which are covered in the summary comments below.

Summary Comments on 9.3

(a) I would be happy if the write-up of the neutronics programs, and particularly those involving large experiments, would be in the spirit of "bowing to the inevitable". As written, it is largely "programming the unnecessary". That is, it ought to be clearly stated that reactor physics is now a well enough based discipline that its predictions, given valid cross-section data, are qualitatively unexceptionable. I recognize that pseudo experts will question this and that some group of reactor physicists who are concerned about their employment will support the question; but I would rather see the issue joined than have it conventionally included in recommendations.

(b) As written, the development program is much more elaborate than I think it ought to be. Insufficient attention is paid to work in collaboration with other AEC programs. As a practical matter, it is conventional to propose "fat" programs, but I here register my objection to this convention.

9.4 - I do not see why the task labeled "demonstration" in Fig. 9.13 could not be in fact the beginning of commercial operation. However, since LMFBFR's are likely to be the optimum vehicles for actinide transmutation, and since they are likely not to be in commercial use much before 1990, the total schedule would not be affected by official demonstrations.

9.5 - The high costs associated with remote fuel fabrication are used in 9.5.1. These costs are expected to be subjected to very large reductions, particularly as the LMFBR program progresses into recycling high-burnup Pu in quantity. The uncertainty which this comment illustrates is, however, much reduced by the strategy proposed by BNWL, and the conclusions of the section are therefore valid.

For reasons cited above, I suspect that the fabrication cost penalty cited in Table 9.6 is too high. For reasons given in my comments on Appendix C, I also do not think there should be as large an enrichment penalty as is cited.

On page 9.66, electricity is priced at 23 mills/kwh. Current base-load costs of generation are ca. 10 mills/kwh for new nuclear plants.

On page 9.69, the interesting matter of HTGR use for actinide recycle is brought up. Prospects may be worth further elaboration, or incorporation in the program described in 9.3.

Also on page 9.39, mention is made of the possibility of fuel reuse without reprocessing (LWR-LMFBR). I have been mentioning this possibility in public lectures for about 15 years, so it can hardly be described as a recent idea.

9.6 - This is in general a well-reasoned section. Risks from transportation may be minimized if nuclear parks are used for all operations of the nuclear industry, as suggested by Weinberg, or even if operations other than power generation are isolated in parks.

As opposed to accident hazards, the milling operations of the nuclear industry routinely increase the potential exposure of the biosphere to

significant quantities of radium. Should recovery and transmutation of Ra be considered? Should mill-tailing practice be compared with actinide disposal practice?

This section may be the proper place to discuss the reciprocal impacts of actinide recycle and safeguards on each other. The net results will probably be favorable to both. However, a separate partitioning facility would incur incremental costs for safeguards - both for technological security as represented by analytical facilities, and for plant security. This latter would have to be particularly stringent in consideration of the contained hazards of the facility.

Appendix A - There is, of course, a real question of cost feasibility for spallation devices. Even ING, based on promotional cost estimates, has too high a cost per neutron for any uses except research.

Appendix C - Comments have already been expressed on the need for removing Pu along with the other actinides. It seems that Claiborne did not contemplate Pu extraction - to which I attribute the (to me) disappointingly low hazard reduction factors of Table C-4. The 1000-year decay time is the touchstone to me, and a factor of 3.5 activity reduction does not seem worthwhile on which to base a whole new technology.

Even the factor of ca. 5 in Fig. C-3 is not exciting. The numbers for 99.9% actinide recycle (Table C-5) are more significant, and justify that number as a target.

The reasons cited for differences between PNL calculations and those of Claiborne seem reasonable, and give confidence to many of the calculations generated.

I am instinctively skeptical about the results of C-5 with regard to the enrichment penalty to be incurred by incorporation of actinides in fuel. My reason is that the actinides are quite valuable burnable poisons. Their incorporation ought to be credited against reduction of boron and rare-earth requirements in the reference fuel, and I wouldn't be surprised if optimum use resulted in an actual enrichment reduction. It is also likely that insufficient credit was taken for fast fission of the actinides: all the higher ones are much more easily fissionable (lower effective threshold energies and higher cross-sections) than U-238 with fast neutrons.

CONCLUDING REMARKS

In spite of the detailed criticisms noted above, the report has merit. I would urge that more emphasis should be given to certain recommendations:

(a) Follow-up studies on detailed schemes - similar in coverage to Claiborne's work - for actinide recycle in LMFBR's.

(b) More complete fuel cycle studies on actinide recycle in LWR's, looking to optimization of cost with actinide recycle. The optimization to be emphasized is that of the whole cycle, rather than any single part. There is a strong tendency for reoptimization to reduce sharply the effect of a cost penalty on a single part of the cycle; and on this basis I expect the cost penalties for actinide recycle which have been derived in this report to be upper limits rather than order-of-magnitude estimates.

May I repeat the observation that the recommended R & D program for actinide recycle seems to me to be overly elaborate, requiring perhaps more money and time than is necessary? Consideration should be given to the possible

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effect that such a recommendation might in consequence adversely affect the chances of the program be adopted at all.

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