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WASTE MANAGEMENT AND PROLIFERATION: AN ASSESSMENT OF TECHNOLOGIES AND POLICIES RELEVANT TO NUCLEAR POWER

FINAL REPORT
JUNE 1975 - MARCH 1977

MASTER

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ROBERT MALONE, WALTER A. SEVIAN, AND RICHARD LESTER

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ABSTRACT

This study presents some of the long-term hazards from radioactive waste management and the problems in safeguarding plutonium in a world moving toward a plutonium economy. To ameliorate these problems, several alternative fuel cycle options are presented: homogeneous reactor, denatured thorium, open, tandem, accelerator-regenerative, co-processing, plutonium, spiking, and partitioning. An assessment is made of a variety of separation technologies applied to these options, including a review of 32 different reprocessing methods.

The effects of these options on both U. S. and transnational policies regarding waste management and proliferation are examined. The history, characteristics, capabilities, weaknesses, and advantages of each technology are described relative to these issues. This study points out how, by considering both problems simultaneously, a fuel cycle might be developed that concomitantly reduces the risks from proliferation and improves the margin of safety in waste management. It addresses the transnational environmental policy issues created by a worldwide nuclear industry and suggests the need for two international organizations: one to manage spent fuel, perhaps even reactor operations in emerging countries, and the breeder fuel cycle; the second to protect the global environment.

Technical and economic feasibilities of photochemical methods are analyzed. Two photochemical schemes for improving existing reprocessing technology by reducing wastes and materials unaccounted for (MUF) are presented. The applicability of this technology, along with column chromatography, Talspeak, and other separation methods, is examined relative to various waste management alternatives including the partitioning and transmutation option. A computer

model to determine the effectiveness of transmutation as a function of separation efficiency has been developed and employed. To estimate health impacts from various fuel cycle options, the Brookhaven energy system network simulator has been integrated with an atmospheric dispersion and pathway analysis model. Using revised ^{222}Rn emission data, it is estimated from the linear hypothesis that the number of excess cancers is slightly less for the open than for the closed cycle. More importantly, the number of excess cancers induced by mill and mine tailings is from one to two times that caused by the rest of the entire fuel cycle.

PREFACE

This study was commissioned under the Office of Energy R&D Policy, National Science Foundation (NSF), in June of 1975. At that time this office was functioning in support of the Presidential Science Advisor, then H. Guyford Stevers, who was serving in the dual capacity as head of NSF and Science Advisor to then President Ford. When Congress created the Office of Science and Technology Policy a little over a year ago, the Office of Energy R&D Policy was eliminated. At that time this work was transferred under G. Graves of the Office of Policy Research Analysis, NSF, and the original scope of this program was curtailed.

This study was requested to cover a broad category of analysis and research on separation technologies, safeguards, biomedical and environmental impacts, as well as societal issues. An assessment of the separation technologies and their possible applications in developing alternate fuel cycles was conducted. The ability of these fuel cycles to reduce proliferation and waste management problems associated with the development of the nuclear power industry was examined. This report is merely a summary of the work performed as much of the detailed work is published as BNL reports or other publications.*

To accomplish the many tasks involved in this investigation, several approaches and methodologies had to be developed. A systems approach was taken in the analysis portion of this study. A detailed process level reference

*Note: It should be pointed out that this work happens to fit in with the total review of our international nuclear policy which occurred after the recent election even though most of this work was completed at about that time. It is anticipated that this document will be used in the current international policy review.

energy system was developed to calculate the emissions efficiencies and costs from various fuel cycle options. To determine the effect on the U.S. economy, this model was developed to be compatible with existing Brookhaven econometric models. This reference energy system was then coupled with an atmospheric transport and pathway analysis model to obtain population dose. Next, health effects were obtained from NRC and EPA-accepted linear dose response curves. Excess cancers induced by various fuel cycle options were estimated. Currently, analyses of health effects from the lightly enriched uranium open cycle and the plutonium-uranium reprocessing and recycle options have been performed.

A systematic scientific review was made of the photochemical literature applicable to actinide separations. Extrapolations based on scientific principles were then used to develop improvements for current purex technology. Some of these suggestions have since been verified by Bell, Toth, and Freedman at Oak Ridge National Laboratory. It appears that these photochemical modifications might reduce the limits of error of material unaccounted for (LEMUF), i.e., in reprocessing plants a small percentage of fissile material might always be diverted within certain limits of error. The limits of error in current technology allow diversion of substantial amounts of fissile materials; i.e., an entire nuclear arsenal could be equipped within a few years of operation even with existing International Atomic Energy Agency (IAEA) safeguards.

Assessment of laser technology applied to reprocessing (both isotope and chemical separation induced by photons) was performed with the aid of Los Alamos Scientific Laboratory. This work established costs in dollars per joule as a function of wave length for laser and lamps applicable to separating isotopes and for separating radioactive wastes. The time frame for feasibility of applying lasers to isotopic and elemental separation was analyzed as well.

Results indicate that for the next several decades there is only a small probability of using this method for producing strategic quantities of weapons-grade material.

An engineering survey of 32 reprocessing methods was conducted with the aid of consultants with engineering expertise in a variety of relevant fields. This survey was performed to determine the applicability of various reprocessing technologies to ameliorate proliferation and waste management problems via advanced fuel cycles.

This summary paper was conducted from the viewpoint of producing a document useful for decision makers; i.e., the technical options available were outlined relative to various policy alternatives.* No attempt was made to select the best alternative. Technical complexity and detail were at times sacrificed for simplicity in order to relate in a concise manner how these technical alternatives could be utilized in policy decisions to reduce societal impacts.

One aim of this study has been to point out the most promising technical alternatives that could produce significant benefits to society, thus allowing decision makers to determine appropriate future energy R&D funding allocations. Results imply that several alternative fuel cycle technologies could substantially lessen the risks from radioactive waste and proliferation.

*Note: The more technical aspects of the issues are dealt with in the list of papers on the following page.

1. "Calculation of Nuclear Health Effects Using ESNS & RAGBAD." (In preparation).
2. "A Photochemical Technique for Reduction of Uranium and Subsequently Plutonium in the Purex Process." BNL Report No. 22443 (September 1976).
3. "Partitioning of Actinide Elements from High-Level Waste Using Photochemical Methods." Proceedings of USNRC Technical Workshop on Waste Partitioning as an Alternative in the Management of Radioactive Wastes (June 1976).
4. "Technology Assessment and Impact Analysis of Separation Methods Applied to Reactor Waste Management.:" BNL Report No. 50628 (March 1977).
5. "The Production of Heavy Water by Photodesorption." BNL Report No. 21691 (Proc. 20th Symp. Soc. Photo-Optical Inst. Eng., August 1976).
6. "Simulation and Optimization Capabilities at Brookhaven National Laboratory, Related to the Operation of the Nuclear Fuel Cycle." BNL Report No. AC/EgP-12 (January 1976).
7. "Projected Benefits from Actinide Partitioning." (To be published as proceedings of USNRC technical workshop on waste partitioning as an alternative in the management of radioactive wastes, June 1976).
8. "Survey of Alternate Approaches for Reprocessing Spent Nuclear Fuel." BNL Report (in preparation).
9. "Photochemistry in Nuclear Fuel Reprocessing." BNL Report No. 22724 (May 1977).
10. "Low-Proliferation-Risk Breeder Reactor Fuel Cycles." BNL Report No. 22656 (June 1977).
11. "A Methodology for Comparative Evaluation of Nuclear Fuel Reprocessing Techniques for Advanced Low-Proliferation-Risk Fuel Cycles." BNL Report (in preparation).
12. "Photochemistry Relevant to Nuclear Waste Separations: A Feasibility Study." BNL Report (in preparation).

I. SUMMARY AND CONCLUSIONS

This work deals with the technical solutions to the proliferation and waste management problems. However, the nature of the proliferation issue is such that motivational methods of control are of at least equal importance. A world without a long-range fission option (nuclear breeder) could mean that there would be excessive tension and competition for an inadequate world fuel supply. Such severe energy shortages could threaten the survival of many nations and thus could lead to a more militant world climate which would, in turn, increase the proliferation risk. (Since proliferation is not directly coupled to nuclear power, isotope separation or production reactors could be used to produce weapons-grade material.) On the other hand a plutonium economy in a world of adequate energy, when compared to no long-range nuclear option, could lessen this proliferation threat as well as the likelihood of war if the proper climate of world cooperation is maintained. Certainly the successful development of another long-range energy alternative could change this rationale.

There may exist, however, a middle ground where some technical modification of the long-range fission option could give the world an adequate energy supply while ameliorating the risks from a plutonium economy. This study is primarily directed toward those technical "fixes" for the proliferation problem as well as the waste management problem. Several fuel cycle options and their reprocessing technologies are assessed with regard to their impacts on radioactive waste management and proliferation. Some of the fuel cycles investigated, summarized below, are (1) partitioning and transmutation,¹⁻⁵ (2) co-processing,⁶ (3) denatured thorium,^{7,8} (4) homogeneous,^{9,10} (5) tandem,¹¹ (6) open,¹²⁻¹⁵ (7) plutonium,¹⁶⁻²⁰ and (8) accelerator breeders.^{21,22}

1. Partitioning and Transmutation

The technical feasibility of actinide partitioning^{1,2} and recycle is examined as a waste management alternative. Actinide recycle is shown to be technically feasible but at considerable cost, perhaps doubling the reprocessing charges.²² Neutronics cost, however, has been shown to be insignificant.²³ The process would probably increase short-term hazards while reducing long-term hazards from radioactive wastes. Thus we have a moral question as well as a technical and economic one: Should the society receiving the benefits accept the risks? This partitioning option, although adaptable to LWR and LMFBR reactor technologies, does not ameliorate the proliferation problem.

2. Co-Processing Fuel Cycle

Co-processing all the actinides and then fabricating them into a recyclable fuel is an option that might reduce the risks of proliferation and result in transmutation of the actinides concomitantly. The proliferation advantage arises because the separation of weapons-grade plutonium from this recyclable actinide fuel element is nearly as difficult as obtaining it from spent reactor fuel. Therefore, the mixed oxide fuel would be somewhat easier to safeguard. Because this technology is only a small perturbation on the existing Liquid Metal Fast Breeder Reactor (LMFBR) fuel cycle, it is likely to be more acceptable to the energy establishment. Several of the reprocessing methods that could be applied to the co-processing cycle include pyrochemical, pyrophysical, and aqueous processing.

Pyrochemical and pyrophysical have several advantages over the aqueous method in reducing the volume of total waste, cost, and proliferation risks because the requirements for co-processing are significantly different from those needed for the aqueous technique. The key requirement is to obtain a process

in which all of the actinides are maintained in one stream, perhaps containing a small amount of fission products to increase the radioactivity of the material. Recent neutronics calculations indicate that decontamination factors from pyro-processes could leave enough fission products in the fuel to discourage diversion but not enough to upset economics significantly. Thus this technique might be an adequate nonproliferation measure and should be considered in detail.

3. Denatured Thorium Cycle

The denatured thorium cycle or the thorium-denatured U-233 cycle might reduce the risks of proliferation to a greater extent than would any of the modified plutonium cycles.^{7,8} If an actinide recycle option is included as part of the ^{233}U -Th breeder, then a comparable reduction in the long-term radioactive waste problem can be realized as well. One of the big advantages of the denatured thorium cycle from the safeguards point of view is that ^{233}U when diluted with ^{238}U or natural uranium will require the high barrier of isotope separation before weapons material can be produced. It is quite clear that this technology is not likely to be possessed by terrorists or criminals and would be very difficult for most nations to obtain. Another advantage is the signature produced from ^{232}U decay products. ^{232}U is always present in small quantities as a by-product of breeding ^{233}U from thorium. Its decay chain gives off a 2.6 MeV gamma which can be easily detected in small microgram quantities by remote sensing equipment. This attribute might increase the likelihood of recovering stolen or diverted material and would make it extremely difficult to divert in the first place, but this cycle is by no means proliferation proof.

It is difficult to compare the non-proliferation advantages and disadvantages of various fuel cycles without the establishment of a clear set of non-

proliferation criteria. One thing we can say is if the economics of this cycle are comparable to any of the modified plutonium cycles, the denatured thorium cycle might be slightly preferred because of the two reasons presented here. It can be argued, on the other hand, that there is no point in producing fresh fuel which is more proliferation-safe than the spent fuel. Since the safeguard chain is only as good as its weakest link, a fuel cycle producing links no weaker than those associated with the lightly enriched uranium fuel cycle is probably quite satisfactory from a nonproliferation viewpoint.

The denatured thorium- ^{233}U breeder reactors will require an initial inventory of fissile material. One likely source of this inventory is from LWR spent fuel. The use of this spent fuel as a source could pay for much of the storage, processing, and waste disposal.

If a co-processing method was selected for reprocessing the denatured thorium cycle the bonus of actinide transmutation along with a reduction in the risks from proliferations would accrue. For example, consider using a modified Super-Phenix (The French LMFBR) to breed ^{233}U . These fuel elements might be oxides, carbides, nitrides, or metallics, and the same reprocessing method may or may not be satisfactory for all fuel types. In addition, the form of the fuel used in the ^{233}U -burning LWRs (or other thermal reactors) could be similar or different in fuel type. A breeder hybrid reactor that uses uranium and plutonium in the core and thorium as the blanket could meet a denatured export criteria (if one is ever imposed) by producing denaturable ^{233}U .

The point is that if one reprocessing technology could handle all of these tasks, substantial savings in capital, operation, and maintenance costs would result. Other benefits stemming from consistent safeguards and environmental and safety standards may also accrue. The pyrophysical, pyrochemical,

and aqueous processes all have the potential for serving these fuel cycles. At present, because of the vastly different requirements of a reprocessing plant employed in a nonproliferation fuel cycle, it appears that high-temperature reprocessing technologies would most likely be superior if developed. However, the aqueous technology has the prime advantage of being "on the shelf."

Therefore, we conclude that a three-prong R&D approach consisting of generous funding for pyrochemical, pyrophysical, and aqueous development would be appropriate. Since there are several nonproliferation options that might benefit from this type of development program, careful consideration should be given to establishing a set of criteria for a nonproliferation reprocessing system pertinent to most of the nonproliferation fuel cycle alternatives.*

4. Fluid (Homogeneous) Fuel Cycle

Several fluid reactor concepts have been investigated as potential export reactors.^{9,10} These are capable of employing either thorium or uranium. If they could be developed, the best reactor fuel cycle from a waste management and proliferation viewpoint might be produced. This would be accomplished by adaptation of a kidney-type reprocessing system which would (a) remove only fission products from the reactor, (b) produce no purified actinide streams, and (c) maintain a breeding ratio of one. This fuel cycle would be much less subject to diversion or theft because no shipments of fissile material would be required either to or from the reactor once it has started up. This reactor, in principle, would consume all of the actinides that are inputted or generated within the core, thus reducing the long-term risks from waste management

*Note: An Extension Paper is in preparation which develops the methodology to evaluate specific reprocessing technologies. This new methodology will allow one to prepare a detailed outline of R&D needs and identify priorities.

without the costly separation, refabrication, and recycle as in the partitioning and transmutation option. However, the technology for the development of this concept may not be as close at hand as that for co-processing or the denatured thorium fuel cycles. (The molten salt breeder program, the closest to the concept, was recently abandoned.)

The reprocessing technology will be closely tied to the form of the fluid-fueled reactor, e.g., the fluoride volatility process might be readily adaptable to a UF_6 -He gaseous core reactor, while the aqueous method might be readily adaptable to a homogeneous uranium nitrate solution fueled reactor. For another example, take the homogeneous tin reactor, i.e., a liquid tin core fueled with ^{233}U -Th might be very adaptable to the "kidney" reprocessing concept. This kidney might operate by removing a small stream of core material and pressurizing it with nitrogen. Most of the fission products would form insoluble nitrides or intermetallic compounds and could be "skimmed off."

5. Tandem Fuel Cycle

The tandem fuel cycle utilizes some of the energy in the spent LWR fuel by consuming this energy in heavy water reactors.¹¹ This is one way of extending the various supplies of uranium without sacrificing the capital expenditures in present LWRs and without chemical reprocessing. Certainly we must recognize that going directly to a heavy water reactor system would probably produce the same benefit in extending the uranium supply. The tandem fuel cycle may require some mechanical reprocessing or partial chemical reprocessing to remove the zirconium cladding and restructure the fuel material. If so, this could be accomplished by a modification of the melt-refining method. The cladding could be stripped and the volatile fission products removed before re-cladding, but this would require reformation of fuel pellets. Other reprocess-

ing technologies that could be used for partial reprocessing as part of the tandem fuel cycle would be the high temperature hydride, liquid metal, and controlled halide. The relative difficulty of safeguarding fuel from the tandem cycle would be almost identical to that in the lightly enriched uranium open cycle. The need for a long-term energy option would not be satisfied by the tandem cycle but might be postponed for a short period. Remote fuel fabrication technology would be required and would add significantly to the expense.

6. Open Fuel Cycle

The present LWR fuel cycle is essentially an open fuel cycle; i.e., there is no reprocessing. No fuel cycle is completely diversion proof; however, the risks can be reduced to varying degrees by several technical alternatives. The lightly enriched uranium open cycle would be comparable in proliferation risks to that of the denatured thorium cycle. It would take fewer gaseous centrifuge units or other advanced isotope enrichment equipment to produce the same number of weapons from the denatured ^{233}U cycle.⁸ On the other hand a ^{235}U weapon would be more difficult to trace or recover. In addition, the spent fuel from the lightly enriched uranium open cycle contains about eight to ten times as much plutonium as the spent fuel from the denatured thorium cycle. The plutonium in the spent fuel is certainly a target that must be safeguarded. Therefore, safeguarding the open cycle may even be more difficult than the denatured thorium cycle if we neglect the fuel cycle center (perhaps multinational) where the plutonium is reprocessed and used for breeding ^{233}U or multinational fuel storage. However, there would be enough plutonium to make a significant amount of weapons in either case.

7. Plutonium Fuel Cycle

This cycle refers to three separate possibilities. The first is the separation of plutonium in LWR spent fuel and recycling this with the uranium as a mixed oxide fuel back into the LWRs.¹⁶ Second, the production of plutonium from ^{238}U in a breeder reactor such as the LMFBR would be used to start up additional LMFBRs or would be recycled in LWRs.¹⁸ This would extend our current uranium supplies by about a factor of sixty. To obtain this factor the efficiency of Pu separation becomes very important in the long term, i.e., one percent less in reprocessing may be too large. The plutonium oxides in both of these fuel cycles would be subject to diversion and theft in reprocessing, fuel fabrication, and in transportation. The plutonium dioxide in the fuel could be easily stolen and used for illicit purposes, including the manufacture of small nuclear explosives without chemical processing. A modification of this cycle would be to spike the plutonium with a gamma or neutron emitter that would ameliorate the safeguard problem slightly while substantially increasing the costs of fuel fabrication.¹⁷ This spiking approach penalizes the energy producers a great deal while penalizing the criminal only slightly. Simple chemical purification of this spiked material could render it suitable for use in weapons. Another modification would be to irradiate the mixed oxide fuel in accelerators or reactors prior to shipment from a secured area. Because this method does not require remote fuel fabrication the economic penalty would be minimized. The same hardware used for handling spent fuel could be used for this irradiated plutonium fuel. The only real economic penalty might be in slightly longer downtimes for reactor refueling. However, preventing diversion of weapons-grade material in reprocessing and fuel fabrication would require significant improvements in existing safeguards.

8. Accelerator-Breeder

Since the design configuration for the accelerator-breeder concept is still very much up in the air, determining reprocessing concepts at this stage might be premature. One configuration called the regenerative cycle has been suggested which might avoid reprocessing altogether.²¹ A spent fuel element would be placed in an accelerator-breeder to produce additional fissile fuel within that element. Next, the element would be reinserted into a reactor. The flux of neutrons in the accelerator-breeder occurs due to the high energy acceleration of protons impinging on a heavy metal target which produces spallation neutrons.²² It is the effect of these high energy neutrons which could lead to yet unclearly defined material problems. The technical feasibility of this concept will hinge on overcoming these radiation effects problems. And even if all technical problems are solved, this technique may not survive for economic reasons because the cost of neutrons is so high.

From a proliferation point of view, going from the best to the worst fuel cycle, we suggest the following order: (1) fluid reactor, (2) denatured thorium-²³³U, (3) open fuel cycle, (4) tandem fuel cycle, (5) regenerative cycle, (6) co-processing, (7) plutonium partitioning, and (8) plutonium. Fuel cycles (2) - (6) must be considered so close at this point as to be indistinguishable; likewise (7) and (8) are very close. It should be noted that by irradiating fresh plutonium fuel the danger of theft during transportation could be greatly reduced.

From a waste management point of view, assuming the long-term hazard is significant, the order suggested is: (1) homogeneous, (2) denatured thorium, (3) co-processing, (4) plutonium partitioning, (5) plutonium, (6) regenerative cycle, (7) tandem, and (8) open cycle. Fuel cycles (2) - (4) are so close as to be nearly indistinguishable.

Reprocessing

The methodologies for 32 reprocessing schemes are surveyed. Twelve of these are reviewed and discussed. (A more detailed paper on this subject is in preparation.) It is concluded from this survey that improvements in the purex technology will be required for advanced fission fuel cycle reprocessing; for example, in the LMFBR. Alternative reprocessing technologies, such as pyro-physical (includes fluoride volatility) and pyrochemical should be examined along with various modifications of the purex technology. The potential advantages of pyroprocesses, when applied to nonproliferation fuel cycles make R&D funding appropriate in these reprocessing technologies. For conventional LWR reprocessing, the purex technology is adequate. However, several improvements can be made that would reduce the quantity of waste to be solidified and reduce process and maintenance problems as well as the limits of error of material unaccounted for (LEMUF).

In addition modifications of the purex technology have been studied for application to the conventional scheme of uranium and plutonium separation. These include using uranium IV as a means of reducing plutonium. Uranium can be produced from the purified stream at the tail end of the purex process, thus avoiding the complications of shielding. Uranium IV can be produced from uranium VI by either photochemical or electrochemical techniques. Chemical reagents used to change oxidation states increase the quantity of material to be solidified by about 50 percent and interfere with process stream recycling. Most of these can be replaced by nonresidual reagents. These photochemical processes are much less sensitive to controlled solution conditions than other processes (because reactions can be initiated after ideal mixing). This tactic could reduce the LEMUF and make diversion less likely.

Mixed Oxide Recycle

However, the reprocessing of LWR fuel could easily be postponed until plutonium in the spent fuel needs to be utilized in a breeder or high converter of some sort. This would avoid the hazards of storing large quantities of separated plutonium in this interim period and might buy us the time we need to formulate adequate safeguards. The energy benefits of reprocessing without both plutonium and uranium recycle are negligible. The economic benefits from recycling plutonium in existing LWRs are marginal and probably not worth the risk. It has been shown from impact analysis that the biological risks to humans, i.e., the health effects from uranium and plutonium recycle, are only slightly greater than from the open cycle. The recycle of plutonium within the United States might be less risky than from international shipment of strategic nuclear materials, but still risky nevertheless. However, if plutonium were consumed in LWRs now, the eventual rapid expansion of the advanced fission fuel cycle, such as the co-processing or denatured thorium-²³³U breeder or even the plutonium cycle LMFBR, could be seriously curtailed. Until such time as these questions are resolved, the recycling of plutonium should probably proceed only on an experimental basis. However, if we postpone the decision to build reprocessing plants too long, we may end up eventually having to construct extra plants to get over the "hump" caused by rapid growth. This time frame depends on the very controversial uranium supply, and this controversy should be resolved before delaying decisions are implemented regarding the breeder program.

Breeders

Any ~~R&D~~ program can have failures, and advanced fuel cycle R&D is no exception. Therefore, aborting the LMFBR without a comparable alternative in hand should be avoided. However, the existing program can be restructured

while the technical and economic feasibility of co-processing, spiking, denaturing, fresh fuel irradiation, etc., is examined. We have time partly because of the success of the French breeder program (although that success could be interpreted by some as a justification for U.S. industrial competition), but mainly because the U.S. has a variety of energy alternatives, such as coal, oil, gas, uranium, and conservation. These can be implemented in the short term very effectively. Many other industrialized countries do not have uranium or fossil fuels, and they also cannot conserve because they already have their temperatures turned down and their automobiles miniaturized. However, any serious delay in our breeder program could eliminate the range of choices for our energy future. If we are concerned with helping the rest of the world meet their energy needs, it makes sense to maintain all our options.

Transnational Nuclear Policy

We propose that international safeguarded atomic fuel exchange (ISAFE) centers be set up (under IAEA or a comparable institution) for the purpose of storing all spent fuel shipped through international boundaries. These centers should be under direct international control, not simply international inspections. Eventually, if safe breeder technology is developed and implemented, then the breeders and reprocessing plants could be co-located at these centers. The fuel produced at the facility would then be shipped to foreign reactors and the spent fuel returned. The large quantities of plutonium spent fuel from existing LWRs and possibly CANDU reactors (HWRs) could be used as inventory in these breeder reactors. Many nations may have difficulty in selecting reactor sites and operating reactors. Therefore, a multinational organization that would construct and operate reactors for their countries might prove very worthwhile. This organization could then sell power to recoup its investment.

It is also suggested that an international environmental protective agency (IEPA) be established to cope with the global aspects of the radioactive waste problem from the international fuel cycle and, in particular, the ISAFE centers themselves. The IEPA could also help in site selection and regulation of reactors in emerging nations.

Environmental and biomedical impact analysis has shown radon-222 (which induces lung and other cancers) from uranium mill and mine tailings to be the largest single killer in the entire fuel cycle (by the year 2000 it is estimated that between 300 and 600 cancers would be induced). It is also noted that radon-222 emissions from phosphate mining probably produce more health effects than the radon from uranium mining. An international environmental protective agency could examine these and many other problems, e.g., the effect of chlorinated hydrocarbons on the protective ozone layer, toxic material in the ecosystem, ocean dumping, oil spills, etc.

It is concluded that, until adequate international institutions exist with the capabilities of safeguarding strategic nuclear material, serious thought should be given to the exportation of coal and slightly enriched uranium as an alternative to the plutonium economy, thus allowing time to develop alternative fission cycles that could ameliorate proliferation and waste problems. Coal has the advantages of lower capital cost and an economy of scale more conducive to small station sizes. Both traits are very desirable in developing countries. But coal like other fossil fuels produces CO_2 , and we don't yet know if large quantities of CO_2 can affect our climate. Coal also produces SO_x , NO_x , particulates, heavy metals, and other environmental damage. These pollutants from the burning of coal, as well as from other portions of the fuel

cycle (e.g., mining), add up to health impacts significantly larger than a comparable, properly operated nuclear system.

Even though safeguarding plutonium is probably feasible within the U.S. we would not be immune from nuclear terrorism resulting from nuclear material diverted abroad. This problem is truly transnational in character and needs to be addressed on that level. It is noted that presently no weapons have been produced from a pure power reactor.

We conclude that the debate over the establishment of a nuclear industry and breeder program should give way to minimizing the risks of the present nuclear industry and planning for a greater margin of safety and security for the public if the long-range fission option is required. The uncertainties in other long-range energy options make it prudent to proceed with the development of a suitable long-range fission alternative.

Most of the developed countries of the world are not as fortunate as the U.S. in having vast supplies of fossil fuels; therefore, they do not have as much time to reconsider the best alternative fission options. Since the costs and social impact of "ineffective safeguards" could be catastrophic to the entire planet, a multilateral fuel cycle development effort directed towards reducing these risks would be desirable. Considering the immediate needs of other nations, perhaps the program could be centered around an existing technology, e.g., modifying the French breeder (Super-Phenix). This type of cooperative effort might in itself create a healthier world climate in which the need for nuclear arms and nuclear terrorisms might be abated. However, to convince countries such as Brazil and Japan and the rest of the world to postpone their reprocessing until alternatives can be examined within the next 5 - 15 years will

require significant policy changes on the part of the U.S. and the rest of the world. Some of the U.S. options that might achieve this are listed below:

1. Increase our enrichment capacity in order to guarantee adequate nuclear fuel supplies abroad and/or contribute to international enrichment capabilities so other nations will not feel dependent on U.S. foreign policy whims.

2. Purchase spent fuel, giving credit for plutonium contained therein, until such time as ISAFE centers can perform this service.

3. Develop our coal resources for export.

4. Finance a large share of a multinational nonproliferation fuel cycle development program.

5. Work with other nations to establish an international council that will address the alternate fuel cycle question and attempt to come up with a plan to reduce risks from proliferation and the long-term hazards of radioactive wastes.

6. Work with other nations to establish an international institution capable of managing spent fuel in the near term and the entire fuel cycle in the long term. (This possibly could be done by strengthening IAEA.)

7. Work with other nations to establish an international environmental protective institution (possibly being created around some existing international environmental organizations such as the one in OECD) to address the transnational environmental consequences from multinational fuel cycle centers as well as a host of other environmental problems of a global nature.

8. Implement conservation policies that will convince the rest of the world that the U.S. is serious about assisting them in this world energy problem.

9. Develop retrievable storage systems for spent fuel elements, both surface and deep geologic.

10. Encourage exploration for energy resources.

11. Give support to soft technologies, e.g., solar heating, biomass conversion, etc., and work with other nations to transfer and finance these technologies in the developing world.

12. Work with other countries to set up an international financial institution to assist the developing world in meeting their energy deficits (perhaps through the World Bank).

II. INTRODUCTION

Two major controversies over nuclear power center around radioactive waste management and the safeguards issue. Both problems hinge on the fixing and controlling of the actinide elements. Public acceptability of policies to manage the actinides could indirectly determine the future of nuclear power and perhaps the economic welfare of the world.

The first critical facet of the actinide problem is the waste management issue. This problem may be ameliorated by actinide partitioning; i.e., by removing 99.9% of the actinides, secure storage requirements of high-level waste are reduced from hundreds of thousands of years to less than one thousand years.^{1,2} Disposal of radioactive waste in deep geologic formations appears more reasonable for 1000 years than for hundreds of thousands of years because geological forecasting is much more reliable on this time scale. Several methods of handling these separate actinides have been suggested. The separated transuranics can be (1) transmuted to stable or shorter-lived fission products by recycling them in reactors, (2) placed in super-secure storage, or (3) rocketed off the earth.

Many of the problems associated with waste management could be simplified if good separation methods for various elements and isotopes were available. Recently the economic and technical feasibility of removing actinides and long-lived fission products from high-level waste by standard chemical processes has been evaluated.^{1,2} In Section IV of this report some alternatives for improving the purex process to accomplish partitioning are discussed.

A second critical facet of the transuranic problem involves reducing the risk of nuclear weapons proliferation from the spread of the nuclear power in-

dustry. The October 28, 1976 nonproliferation policy statement by former President Ford illuminated the need to re-examine our fuel cycle options. As a part of this effort, 32 separation (reprocessing) technologies have been surveyed, and the role they will play in an overall assessment of waste management and proliferation has been examined. In some instances it is difficult to separate reactor concepts from the reprocessing concepts because they are so closely integrated as, for example, in the molten salt breeder reactor.

Any time that change in a major strategy is suggested, inertia against that change becomes more apparent. Therefore, if it becomes necessary to effect a transition from one fuel cycle plan to another, it might best be accomplished by a series of steps. The first step preserves as much present technology as possible yet still moves significantly in the direction we ultimately would like to go.

The possibility that many nations will undertake commercial reprocessing by themselves is highly likely without international agreements and multinational facilities. This would increase the potential for diversion of nuclear material. For the present and the immediate future a purex-type technology will probably be utilized for either national or international reprocessing. Therefore, an important task is to improve this technology to minimize the likelihood of diversion. This may be accomplished by reducing the quantity of waste, improving separation factors, reducing the limits of error of material unaccounted for (LEMUF), and developing more accurate analytical techniques to measure quantitatively the concentration of plutonium (or other fissile materials) at various points throughout the reprocessing facility.

In Section VII an illustration of how photochemical techniques might improve the purex process is given. This subject has been under review at BNL for the past two years.

Before delving into other specific types of separation techniques that could reduce or slow down the risk of proliferation, some potential fuel cycle modifications that might ameliorate the safeguards and proliferation problems by avoiding the circulation of weapons-grade plutonium (free of fission products) will be described. To be consistent with former President Ford's October 28, 1976 policy, let's call this a "more-proliferation-safe fuel cycle." Reducing the risk of international proliferation by one of several fuel cycle alternatives that avoids the circulation of fission-product-free plutonium across the boundaries of nations is perhaps feasible. For example, the once-through or throw-away cycle, the accelerator-breeder concept, the tandem fuel cycle, co-processing, and the denatured thorium fuel cycle can be constructed and implemented to minimize the latent risks from plutonium recycle and circulation.

None of these fuel cycle options is new, but nevertheless our current perception of the risk of latent proliferation and long-term waste hazards requires reassessment of these alternatives. The throw-away, the accelerator-breeder, and the tandem fuel cycles do not require chemical reprocessing, thus avoiding altogether the separation of weapons-grade plutonium but increasing the long-term waste hazard at the same time. Each of these methods has advantages and disadvantages; however, the primary advantage rests with the throw-away cycle. Neglecting waste disposal, it exists. Material problems will have to be overcome for the accelerator-breeder or the tandem fuel cycles.

Any one of these fuel cycles might serve satisfactorily in the short run. However, we must also plan for the scenario in which the growth of nuclear energy is such that the once-through or even the tandem fuel cycle cannot supply all the world's energy demands. In this case other fission alternatives might be required to meet demands if solar electric or fusion, the other ERDA long-term options (or some non-ERDA option), has not been commercialized by that time. To plan for this eventuality, we should look for ways to more safely breed (and/or convert) nuclear fuels without increasing the risk from radioactive waste or proliferation. These might include standard safeguard measures (see one of several books on the subject^{15,25-26}), spiking^{27,28,29}, and/or co-processing, irradiating plutonium fuel, the denatured thorium cycle, the plutonium cycle, or other fuel cycles.

III. WASTE MANAGEMENT

Present-day management of the actinides can be thought of in both humanistic and technological terms. Features of particular schemes can be related to present and future costs, perceived benefits, the time-scale of concern, and, in addition, the depth of responsibility felt for future generations. Most of the nuclear waste management schemes add an effective cost of only about 1% to the price of nuclear-powered electricity. Since the cost is so small in comparison to the time-scale of concern, perhaps the additional cost of partitioning nuclear waste and transmuting long-lived materials might be worth the additional reduction in future risks.

To illustrate this point let's consider dividing the wastes into long-lived and short-lived species. The first type contains the actinides, which have very long half-lives; that of plutonium-239, for example, is about 25,000 years. The radioactivity of type 1 materials (actinides and certain long-lived fission products) becomes dominant after about 500 years. The handling of the actinides can be thought of as a million-year problem.^{1,2} Thus, separation of this small volume of materials may offer many alternatives for disposal of the remaining larger volume of materials.

In type 2 materials, long-lived fission products such as ^{137}Cs , ^{85}Kr , and ^{90}Sr have half-lives approaching 30 years. After 700 years, only one ten-millionth of the radioactivity from these products would remain.

The taxonomy of options being considered has been recently discussed in some detail by Kubo and Rose.³⁰ From examination of this work, it is evident that a separation technique that successfully alters the long-term hazards of the bulk of the waste might have great value. This is because removing the ac-

tinides from the fission-product waste stream reduces the 1-million-year problem to a 700-year problem, except for a very small residue still in the million-year category. It may then be possible to treat the reduced million-year category by such techniques as neutron transmutation (e.g., recycling in fission reactors or possibly in fusion reactors) or extraterrestrial disposal thus eliminating this long-term hazard from the biosphere.

Since reprocessing of spent reactor fuel is a commercial venture, uranium and plutonium are the only actinides extracted from the spent fuel elements. The degree of recovery for solvent extraction approaches 99.5% which is considered moderate at best.² The use of photoexcitation, Talspeak solvents, ion exchange resin,² and other techniques offers the potential of achieving separations of 99.9 to 99.99% of the actinides, leaving a residue considered ultrapure. Under these extreme extraction conditions, the residue would approach nontoxicity in less than a thousand years, with a concomitant decrease in the cost of handling this waste [i.e., if depth requirements are relaxed for the actinide free waste] and the danger of its release into the biosphere. Such reduction could amount to three orders of magnitude in the time scale required for secure storage (to keep material out of the biosphere).

Possible waste management schemes discussed in recent years are engineered above-ground or shallow-burial storage, salt-mine disposal, granite-mine disposal, recycling of actinides, transmutation of fission products and/or actinides, glassification, melt in situ repositories, Antarctic rock, the continental ice sheet, the ocean bottom, and tectonic sinks. At present the lack of knowledge of the ocean bottom and tectonic sinks makes it unsafe to use them as repositories, given the long storage periods required for actinide materials.²⁹

Three separate categories of waste management techniques are geological burial or long-term storage, removal from the biosphere by rocket, and transmutation of actinides into fission products for subsequent storage. The use of burial techniques has been questioned because geology over the long time-scales involved is not well understood. The cost of rockets and the potential danger of failure make transportation into space somewhat questionable. One solution might be to transmute the long-lived isotopes to shorter-lived or stable isotopes and thus diminish the time-scale to one for which geological forecasting would be reliable. Transmutation or space disposal will be feasible, however, only if adequate separation schemes are devised.

IV. PARTITIONING

Several partitioning options have been considered in the past.^{1-3,31-35} One such option involves the separation of cesium and strontium, the principal heat sources in LWR spent fuel, from remaining wastes. This partitioning option has the advantage of reducing the heat in high level waste and therefore reducing cooling and storage value requirements. It has been argued that the remaining wastes would be cooler, and therefore the long-lived actinide fraction within that cooler waste would be less likely to perturb the not yet well understood geophysical or geochemical behavior of a particular disposal site. This would increase the reliability of the containment of the disposed waste; however, this strontium and cesium partitioning option was rejected after serious consideration for the following reasons:

1. By diluting or spreading out the unpartitioned high level waste, the same heat load on the geologic site can be obtained at a lower cost.
2. This method does not effectively reduce the long-term hazard associated with the waste, even if the strontium and cesium are transmuted.
3. The recycle of plutonium in a breeder economy or particularly in LWRs would produce a large percentage of the heat load from the actinides within the waste, thus requiring special cooling and handling equipment in any case.
4. There is no advantage to separating the strontium-cesium because reliable storage within its toxic lifetime is considered highly probable.

The partitioning option that has become a viable waste management alternative is the separation of the long-lived fraction from the short-lived fraction of high level wastes. Recognizing the importance of these long-lived waste separations, the U.S. Atomic Energy Commission initiated the waste par-

tioning program in 1974. Initial studies on the partitioning of actinides from high level wastes showed that the program needed to be greatly expanded and that secondary processing of the wastes from conventional reprocessing streams was not a particularly desirable way to remove actinides from fission products.

This partitioning method as a waste management alternative depends on separating and solidifying one stream from the reprocessing plant containing mainly the fission products, with an acceptably low content of long-lived radionuclides, and producing a second semi-pure concentrated stream containing the long-lived radionuclides. Oak Ridge National Laboratory,¹ PNWL,⁵ BNL,² and others³¹⁻³⁵ have been investigating the feasibility of accomplishing this partitioning task by modification of the existing purex process. Several process flow sheets have been developed for the recovery of uranium, neptunium, and plutonium with subsequent processing to separate americium and curium from the lanthanides. ORNL is currently undertaking a major three-year effort to:

1. Determine the technical feasibility of recovering very nearly all of the actinides by modification of purex technology (and possibly selected long-lived fission products) from virtually all waste streams in which they occur in the nuclear fuel cycle (neglecting mill and mine tailings).

2. Determine the technical feasibility and nuclear fuel cycle impacts of transmuting the recovered actinides (and long-lived fission products) in fission reactors.

3. Assess the incremental risks which would be incurred if partitioning and transmutation were to be implemented.

4. Assess the incremental benefits which would accrue from the destruction of the long-lived radionuclides.

This effort is a continuation of previous ORNL work depicted in the conceptual processing chart in Figure 1. First, suitable trivalent actinide-lanthanide feed solutions are separated from the exhaustively extracted high level waste (99.99% Pu free). Next, americium and curium are extracted by one of the following methods: (1) The cation exchange chromatography (CEC^{1,31}) process illustrated in Figure 2, which involves the selective absorption of actinides on a ion exchange resin and then elution of the purified species; (2) the Talspeak process^{1,4} illustrated in Figure 3, which is based on the preferential extraction of lanthanides from trivalent actinides by various solvents, e.g., bidentates; (3) a method being investigated at Euratom-Ispra that involves oxalate precipitation of crude concentrations of plutonium, americium, curium, and lanthanide fission products from high-level waste; (4) a technique studied by ORNL³¹ of coupling the oxalate precipitation with ion exchange; (5) a photochemically modified purex process capable of simplifying the flow sheet by avoiding residual solids in the waste examined at BNL;² and (6) inorganic ion exchangers being evaluated by Sandia.^{34,35} Major additional experimental testing is needed, however, to evaluate these and other methods for americium and curium separations.

The scientific feasibility of partitioning by any of these purex modification techniques is not in question; but, because of the added complexities to the reprocessing flow sheets introduced by partitioning, the technical feasibility on a commercial scale must be demonstrated before accurate cost and net benefits can be determined. However, any of the partitioning techniques mentioned above would probably at least double the size of a conventional reprocessing plant and therefore could easily double the cost of separation. The

Figure 1. Conceptual processing sequence for the removal of actinides from LWR reactor fuel.

Figure 2. Schematic flowsheet of cation exchange chromatographic process for recovery of americium and curium.

Figure 3. Conceptual flowsheet for solvent extraction processing of irradiated LWR fuel with waste partitioning and recycle of heavy-metal and nitric acid values.

development cost would probably run into the hundreds of millions and might take as long as 15 years to develop.³²

But the separation in itself accomplishes nothing, because the actinides must be kept out of the biosphere for extremely long times. This can be accomplished by removing them from the planet, isolating them in geological formations, or transmutating them. If transmutation is selected, these materials must be remotely fabricated as part of the reactor fuel elements or target elements. Because of the slow burn up (5 - 10% actinides fissioned per cycle), they will be continually separated and refabricated over many years, thus adding to the complexity. The benefit accrued from a decrease in long-term hazards, however, must be weighed against the potential short-term increase in occupational hazards and risks to the population from a potential accident. And even if this long-term benefit far outweighs the short-term increase, one has to ask how will this program, if implemented, affect the proliferation issue. It is not altogether clear that this particular partitioning and recycle scheme would be much different from the Pu recycle case. Even if we assume for the moment that safeguards would be satisfactory within the facility, the shipments (especially international) of purified plutonium and other actinides would be subject to diversion and theft.

It is apparent that partitioning and recycle by purex modification could perhaps reduce long-term health hazards but may increase occupational and short-term hazards without reducing the risk of proliferation and perhaps even increase proliferation risks since most of the higher actinides including neptunium are fissile and could be purified, thus subject to diversion. However, if these actinides could be co-precipitated, i.e., never actually purified but handled as a group, several advantages would accrue. First, the transmutation

of this actinide would reduce the long-term hazards of radioactive wastes. Second, diversion of even the smallest quantities could be detected by surveillance instruments. Since uranium is in a high concentration, the dilution of the actinides by nonfissile uranium would make chemical processing of this mixture necessary to produce a weapons-grade material. On the other hand, this chemical separation would not be all that difficult.

V. TRANSMUTATION

Under this part of the program, estimates of future actinide production by the nuclear power industry have been made, and estimates of actinide partitioning costs for the Oak Ridge partitioning scheme have been initiated.

A computer program has been written to estimate actinide production and accumulation in the high-level waste generated by an expanding nuclear economy over a period of up to a hundred years. Three options for treating high-level waste are considered: (1) no partitioning, i.e., a continuation of current practices; (2) partitioning of high-level waste and separate storage of the actinides; and (3) partitioning of high-level waste and recycling of the actinides in light-water reactors (LWR's). Each option includes two suboptions: recycling of Pu in LWR's or no recycling. It is assumed that actinide partitioning is performed in an add-on facility at the back of a reprocessing plant. Five actinide elements (17 isotopes) are considered: ^{234}U to ^{238}U , ^{237}Np , ^{239}Pu to ^{242}Pu , ^{241}Am to ^{243}Am , and ^{242}Cm to ^{245}Cm .²

The separation efficiencies for these isotopes in the reprocessing plant and in the actinide partitioning facility are treated as variable parameters. Data on actinide inventory in the spent fuel, assuming partitioning and recycling, were taken from BNWL-1900. Simplified semiempirical burnup curves, predicting for each actinide isotope its production as a function of time and recycle rate, were constructed. A more elaborate treatment of actinide production and burnup is being conducted by Croff at ORNL using ORIGEN.³³ The evolution of the nuclear power industry, and in particular that fraction represented by the LWR's, is an input data set specified by the user. The program output

includes the annual weight, or amount, of actinides handled by the fuel cycle industry under each of the three waste management options shown in Table 1.²

Table 1	
Total Weight of Actinides in Storage by the Year 2075	
Option (100-yr LWR)	Actinides in Storage (tons)
1. No partitioning, i.e., a continuation of current practices	1600
2. Partitioning of high-level waste and separate storage	1600
3. Partitioning and recycling	4

Initial results for option 1 (no plutonium or actinide recycling) were compared with similar results recently published by Ang and Pigford of UCLA.³⁶ Good agreement between the two sets of results was obtained for all the actinide elements treated at BNL. The effects of increased actinide production in the recycling of plutonium in LWR's and of actinide "ratcheting" (increase in higher actinide production due to recycle) in the recycling and transmutation of actinides were considered. This program is useful in that it delineates the magnitude of the actinide waste management problem for an evolving nuclear power economy. It is especially useful in indicating the reduction in actinide management problems expected as partitioning and transmutation methods of various efficiencies are developed. However, more detailed calculations and experimental verification of transmutation rates in various reactors will be required before a policy decision can be made on these options. It should also be noted that there will be more actinides remaining in the fuel cycle for option 3 than

1 and 2 at the end of the nuclear scenario, but that number will be small compared to the quantity in storage for options 1 and 2.

VI. PROLIFERATION

There are at least five separate paths by which proliferation can take place:

1. One may be the recipient (gift or purchase) of a completed weapon or the materials to make a weapon.
2. One may steal a weapon or the materials to produce one.
3. One may obtain isotope separation equipment for manufacturing weapons-grade material.
4. A production reactor and associated reprocessing plant may be constructed for this purpose.
5. One may obtain weapons-grade material from the nuclear power industry.

This study deals primarily with the latter and is concerned with modifying existing fuel cycles to ensure that the nuclear power industry is not the weakest link in the safeguard chain. There are several other classifications of proliferation that should be defined: First, there is vertical proliferation. This is the development of more sophisticated weaponry and delivery systems, commonly referred to as the "arms race," where one goes from small atomic weapons to thermonuclear weapons, to multi-warhead delivery systems. Then, we have horizontal proliferation of two types: governmental and nongovernmental. Governmental proliferation is simply the spread of nuclear weapons to additional countries; whereas nongovernmental proliferation involves the acquisition of nuclear weapons by terrorists, criminals, or other subgovernmental groups.

Recently many papers and books have appeared on the subject of proliferation.^{7,8,15,19,25,29,37,38} Several technical solutions to the societal problem of proliferation have been suggested. These methods include the following:

1. Denatured thorium cycle
2. Co-processing
3. Spiking plutonium
4. Low inventory reactors
5. Homogeneous reactors
6. Tandem cycle
7. Accelerator-breeder cycle
8. Open cycle

These fuel cycles are described, and their characteristics that help ameliorate the proliferation problem are examined.

For years the approach to solving problems within the nuclear fuel cycle was one of isolation; i.e., isolate a specific problem and focus on it. Therefore, the waste management problem was addressed entirely separate from the problem of proliferation and the reprocessing problem separate from fuel fabrication, etc. This approach has introduced certain complexities into the system; e.g., the complexities of partitioning, recycling, and transmutation might be greatly reduced along with the risk of proliferation if we coordinate our attack on these problems. If it is decided to prevent plutonium recycle either domestically or internationally, then one way to keep the long-range fission option alive would be to go back to step 1 in developing the nuclear fuel cycle and start again, this time designing and developing a system safer for proliferation and waste management.

1. Denatured Thorium-²³³U Cycle

The thorium-uranium breeder cycle may be constructed in such a fashion as to eliminate the need for international shipments of fission-product-free plutonium and at the same time reduce the transuranic inventory in the waste.^{7,8}

If the international shipment of plutonium is vetoed for political, or safety, or other reasons, then a breeding cycle based on the denatured thorium cycle might be one of several alternatives in replacing plutonium as an international recyclable fuel. In fact, if international safeguarded atomic fuel exchange (ISAFE) centers are set up at selected locations, the breeding of uranium-233 from thorium might be initiated by first consuming plutonium already produced by light water reactors and presently stored in various fuel storage facilities. The plutonium extracted from reprocessing plants could serve as the initial inventory for these breeders. Thorium could be placed in the breeder reactors as fertile material and converted to uranium-233 as illustrated in Figure 4.

Uranium-233 could be recovered and denatured within the confines of this facility. Denaturing or diluting could be accomplished with natural uranium or the tailings from a ^{235}U enrichment plant as depicted in Figure 4. Figure 4 also illustrates how the recycle of denatured ^{233}U could essentially replace plutonium recycle in the international fuel market. Fuel elements fabricated with denatured ^{233}U (if additional fuel elements are required, they could be fabricated out of slightly enriched ^{235}U) could be shipped internationally to various LWR or other types of thermal reactors.

In the event of theft, the production of weapons from these materials could not be accomplished without isotope separation technology. The possession of such technology would allow the development of weapons from natural uranium sources, thus precluding the need for these fuel elements. Spent fuel from uranium-233-fueled reactors would contain some plutonium, which would be returned to the ISAFE center's spent fuel storage facility as depicted by the dotted portion of Figure 4. These fuel elements would be reprocessed by some

INTERNATIONAL SAFEGUARDED ATOMIC FUEL EXCHANGE CENTER

Figure 4. A fuel cycle that avoids the shipment of mixed oxide fuel elements by: (1) breeding Uranium 233 from Thorium, (2) consuming the plutonium within the boundaries of the ISAFE Center, and (3) denaturing the fissile Uranium 233 with natural Uranium.

yet unspecified technology, and the plutonium fed into a ^{233}U breeder where it and perhaps the higher actinides would be consumed.

It has been pointed out that a reprocessing plant which must service such a breeder would have to make four cuts (separations): for example thorium, uranium, plutonium, and fission products, and would necessarily be somewhat more expensive than a typical three-cut facility. However, by co-processing the uranium, plutonium, and higher actinides for fuel fabrication and ultimate use in the $\text{Th}-^{233}\text{U}$ breeder, reprocessing costs could be reduced and safeguard problems ameliorated. Several high-temperature reprocessing techniques appear to be more suited for closing this cycle because of their adaptability to

- (1) Co-extract U, Pu, and other actinides
- (2) Enrich the concentration of U or Pu without purification of either
- (3) Produce decontamination factors adequate for neutron economy in either LMFBRs or LWRs but low enough to prevent diversion.

The reason these techniques were previously rejected is because they could not easily produce plutonium purified enough for "hands on" weapon or fuel fabrication. Now the requirements are reversed since the need to prevent proliferation is taking precedence over the expediency of working with purified plutonium. Previous advantages of using conventional materials on aqueous processes may no longer exist because aqueous reprocessing of thorium may require fluoride handling and therefore more expensive materials in plant construction.

The thorium fuel cycle just described can be thought of as one step in a transition to an advanced "nonproliferation fuel cycle." Actual reprocessing to take place within the ISAFE center could closely resemble conventional reprocessing, or it could be one of many advanced separation technologies, selection of which would certainly depend on the selection of the breeder concept

as well as on economic, safeguard, and environmental considerations. A comparative analysis and perhaps competitive development would be required to determine which breeder and which reprocessing technology are most adaptable to the framework of a thorium ISAFE center and how this concept compares to improved safeguards on the plutonium cycle and the coprecipitated modification of the plutonium cycle.

A few examples of breeder concepts with examples of corresponding separation technologies that might be applied to close those fuel cycles are listed below:

1. LMFBR

- (a) Modified thorex-purex aqueous process

- ~~(b) High temperature salt transport~~

- (c) Pyrometallurgical

2. Molten Salt Breeder

- (a) Salt bypass

- (b) "Kidney" reprocessing

3. Gaseous Core

- (a) Fluoride volatility

- (b) Kidney reprocessing

- (c) Aqueous reprocessing

4. Thermal Breeders

- (a) Thorex

- (b) Salt transport

- (c) Hydride

- (d) Pyrometallurgical

5. Fluid Bed
 - (a) Aqueous reprocessing
 - (b) "Kidney" reprocessing
6. Liquid Metal
 - (a) "Kidney" reprocessing
 - (b) Pyrometallurgical
7. Accelerator-breeder
 - (a) Regenerative cycle, no reprocessing
 - (b) Aqueous reprocessing
 - (c) Pyrometallurgical

A plutonium-thorium LMFBR is a leading candidate for development at present because this technology is much closer to commercialization than other Pu-Th breeders. And aqueous processing is the leading candidate for reprocessing simply because of the relative stage of development of this contender and the fact that most of the current administrators in office today have little experience in nonaqueous reprocessing. However, all of the alternative fission reactors and reprocessing technologies should be examined and evaluated.

2. Co-processing

Another approach to controlling the actinides is the co-processing technique as mentioned in Section III. This concept, developed by Pobereskin,⁶ modifies the fuel cycle by aggregation of all the actinides or just uranium and plutonium -- the first of which would no doubt make diversion somewhat more difficult. The second actually makes the material easier to handle, but additional chemical processing would be required (although not a complex step) to produce weapons. If the entire actinide group plus a few fission products are aggregated as a homogeneous mixture, the risk of theft of the recycled fuel

when shipped back to the reactor will be reduced. Both the long-term radioactive waste hazards from the actinides and the safeguards problem are ameliorated by this technique. In addition, it has a special appeal because it is adaptable to current reactor concepts (LMFBR-LWR) as well as to other existing fuel cycle technologies without completely overhauling the system.

The separation technologies for performing this reprocessing could possibly involve a modification of the purex process; however, it is clear that many other techniques, especially the high-temperature methods, would have some distinct advantages and should be considered. Advantages and disadvantages of particular technologies will be discussed in Section VII. Certainly the risk of proliferation from the shipment of recyclible fuel containing the aggregated actinides would be comparable to, or only slightly greater than, the risk for spent fuel shipments. This concept is similar to the well known spiking concept.²⁷

3. Spiking .

Several proposals²⁷⁻²⁹ for spiking plutonium with small quantities of higher actinides or fission products have been put forth. These would make "hands on" diversion of plutonium oxide fatal to the diverter. The spiked plutonium would therefore have to be fabricated into fuel using remote handling techniques, thus increasing the hazards and cost of fuel fabrication and transport. However, after plutonium has been recycled several times, it may have to be handled remotely in any case because of the build up of plutonium-240, 241, and 242. Chemical separation of a spiked material would not be as difficult as separating plutonium from spent fuel, but would be more difficult to handle than unspiked mixtures of uranium and plutonium. Recovery of stolen or diverted plutonium would certainly be easier if the material were spiked.³⁹

The level and type of spiking would be very important in determining the effectiveness of this concept.

An earlier BNL study²⁹ indicates that this spiking concept would be very expensive for the benefits gained. Another modified plutonium spiking concept would be to handle purified mixed oxides in secured reprocessing and fuel fabrication facilities and then irradiate the fresh fuel before shipment. This could prevent the likelihood of theft of the fresh fuel but would have little effect on the likelihood of diversion within the fuel cycle center. Because the hardware and techniques already exist for handling spent fuel, a minimum of capital cost and R&D would be required to implement this method. This irradiation technique could be coupled with other "nonproliferation" concepts to be effective against diversion.

4. Low Inventory Reactors

Additional risks of international proliferation arise from the very large fissile inventory in LMFBR and LWR reactors. One LMFBR reactor could be mined to produce an entire arsenal of nuclear weapons by a perhaps newly "initiated," irrational dictator. Of course, no one is advocating the export of LMFBRs, but a demand could eventually develop and it would be difficult to avoid for political and economic reasons. (Japan has already made preliminary arrangements to purchase a Super-Phenix from France.) Uncontaminated mixed-oxide fuel for LWR plants would be vulnerable to theft especially during shipment and prior to insertion into the core. However, even spent fuel can be used to make bomb material as India has proven.

To reduce this threat, several proposed low inventory reactors would be desirable, such as the gaseous core reactor.^{9,10} Those reactors that contain only enough fissile material for one small weapon have significant nonprolif-

eration advantages as export reactors: first, because developing nations need reactors much smaller than current LWRs and, secondly, because fuel shipments and inventories would be less subject to theft or diversion. But again no such reactor has yet been developed. If this reactor has a breeding ratio of one, then no fissile material need be shipped to it except for start-up. The input to the reactor would consist only of a fertile material such as thorium. Unlike the co-processing technique, the risk of theft from recyclable fuel would be eliminated entirely once the reactor is operating. It is the shipment of fission-product-free fissile material that is by far the most difficult to safeguard especially in the international arena.

All of the actinides contained in the homogeneous reactor could be eventually consumed, i.e., transmuted to fission products, therefore never leaving the facility, by incorporating an on-line reprocessing unit as in the molten salt reactor. To make on-line reprocessing an unlikely place for diversion, purified actinide streams might be avoided as much as possible. This type of reactor (e.g., gaseous core reactor) might be designed to shut itself down if any significant quantity of fissile materials is diverted.^{40,41} But this on-line reprocessing technology would have to be safeguarded to a greater extent than a conventional LWR because one might take over the reactor and separate weapons-grade material. The likelihood of diversion from within will depend on the technologies employed for various reactor concepts and which separation technologies are employed.

5. Fluid Fueled Reactor with "Kidney"

The weakest link in regard to the proliferation risk of the denatured thorium cycle involving the ISAFE center concept, as described earlier and illustrated in Figure 4, appears to be the spent fuel shipments from reactors

spread throughout the world. These shipments might be eliminated by the development of some type of homogeneous or fluid fueled reactor (e.g., molten salt,⁹ gaseous core,¹⁰ liquid metal, etc.), which consumes nearly all the actinides fed into it and has associated with it some sort of on-line separation system to remove fission products.

The separation device, depicted in Figure 5 and referred to as the "kidney," would process some of the fluid fuel and remove a portion of the fission products from each aliquot passing through the unit. The design of this type of device could avoid making it a likely point of diversion of nuclear materials. This would be accomplished by removing only a small percentage of the fission products per pass through the "kidney" as illustrated in Figure 5. Besides reducing the risk of weapons proliferation, this technique could have the advantage of more simply transmuting the actinides -- certainly simpler in principle than removing fuel, partitioning, purifying, refabricating, then transmuting each recycle at a rate of 5 - 10 percent, and repeating the procedure again and again. Materials that never leave the reactor certainly would be less subject to diversion. The time needed to develop such a fuel cycle is not calculable with present information. The problem or disadvantages of such a system have not as yet been completely calculated.

In considering all fuel cycle alternatives, one needs to separate the options which apply to the near- and mid-terms (the present to the year 2000) from those which apply to the long-term (beyond 2000). No breeder reactors are expected to make any significant impact on U. S. energy requirements before the end of this century, not even the LMFBR which is by far the closest to commercialization. It would probably take at least 20 to 25 years to implement any ²³³U breeding device into our national power system, not to mention

REACTOR THAT CONSUMES ALL OF THE ACTINIDES,
ONLY fp ARE REMOVED FOR PROCESSING AND
WASTE DISPOSAL

Figure 5.

internationally. The question is what can we do today with the limited near- and mid-term options available to get through the next 25 years.

6. Tandem Fuel Cycle

One way of theoretically recovering some of the energy left in the spent fuel is to reuse the fuel without chemical separation from the bulk of the fission products by inserting the LWR spent fuel into a heavy water reactor. Davies and Moss⁴² have estimated that an economic advantage equivalent to chemical reprocessing could be obtained, however, material problems regarding the cladding have yet been solved. If the material problems are solved, this technique would stretch our supplies a few decades at most (not a negligible benefit). But we still would have the long-term waste management and proliferation problems associated with spent fuel. However, the long-term option of an inexhaustible energy supply to feed a growing world in the 21st century would be absent.

7. Accelerator-Breeder

The accelerator-breeder can be implemented in several configurations.²¹ One configuration may avoid chemical reprocessing of spent fuel elements altogether and possibly enrichment as well. A spent fuel element or a fuel element made of natural uranium or thorium could be made fissile by placing it into an accelerator-breeder reactor powered by a high energy proton accelerator. The proton beam would collide with a heavy metal target producing spallation neutrons, which would breed either plutonium or uranium-233 within the fuel element. Even though this technique of regenerating spent fuel elements would avoid the opportunity for diversion present in a reprocessing plant or fabrication facility, the hijacking or theft of these fuel elements, containing chemically separable fissile material, could be a mechanism for proliferation.

However, if the Th-²³³U breeding cycle is employed, followed by denaturing, the proliferation risk may be ameliorated.

The primary problems associated with this technology appear to be in materials: first, in the spallation device itself, and secondly in the cladding of the fuel elements. All known cladding materials would be severely damaged by the high energy spallation neutrons (50 MeV). It is suggested that a recladding technique as simple as a graphite coating, which would be burned off and recoated after each cycle, might greatly improve the long-range potential of this concept. If required, the graphite could be coated with zirconium which could be selectively stripped from the graphite by pyrometallurgical, pyrochemical, or aqueous techniques.

The second accelerator-breeder configuration involves the production of fissile fuel by spallation of a target element which would later be reprocessed. This would increase the risks of diversion within the reprocessing and fabrication plants, but would improve the ability to denature the fissile material and thus strengthen the weakest link in the safeguard chain: transportation.

If adequate international control were placed on all breeding, reprocessing, and fuel fabrication facilities, e.g., if the ISAFE center concept were developed around an accelerator-breeder, this technology might become a satisfactory nonproliferation fuel cycle. It appears from first examination that the accelerator-breeder concept might be significantly more expensive than an LMFBR modified to the denatured thorium cycle. However, closer examination is required to be certain of these economics.

8. Open Cycle

The present LWRs and the CANDU heavy water reactors typify the uranium open cycle.^{12-15,43} There is no reprocessing in this once-through fueling

method, which is not a very efficient means of using our uranium reserves. The total energy equivalent of our uranium reserves, if utilized in this cycle, would be approximately the same as the world petroleum supply, while the closed LMFBR cycle would multiply this by approximately a factor of sixty. An impact analysis of the option compared to the LWR with plutonium and uranium recycle is presented in Section VIII.

The formulation of national as well as international energy policies should take these fuel cycle options into account in planning our long-term energy future. The various separation technologies available for adaptation to these alternative fission fuel cycles are described in Section VII. The history of each separation technology, the stage of development, the basic scientific and engineering principles, and the application of the technologies are discussed. This work attempts to gather all the pertinent information on reprocessing technologies that might be used in planning and policy analysis. This review can also serve as a data base and springboard for a detailed comparative analysis of separation technologies applied to advanced (nonproliferation) fuel cycle concepts.

Figure 6. Fission-yield for ^{235}U versus mass number.

Figure 7. Classification chart of spent reactor fuel reprocessing methods.

VII. SURVEY OF REPROCESSING TECHNOLOGIES

1. Introduction

The asymmetric fission of fissile material produces a spectrum of radio-nuclides as shown in a typical mass-yield curve (Figure 6).⁴⁴ These fission fragments, along with neutron capture and decay products, absorb neutrons and act as fission poisons. The fission products build up in the fuel matrix during reactor operation until the combined effects of poisoning and fuel depletion make replacement necessary.

The residual amount of fissile material in the spent fuel is made up of significant amounts of uranium-235 and plutonium-239. Removal of the fission products would make the fuel, or a blend of it, suitable for reuse. Hence, the reprocessing of the spent fuel offers an opportunity to conserve fuel.

A number of techniques for reprocessing spent nuclear fuel have been suggested. The only technique that survives actively is the aqueous liquid-liquid extraction process (purex), which was developed initially for military purposes. Nearly all the other techniques were developed with knowledge of the purex process. These techniques were usually directed at addressing a particular problem and different set of constraints than those for liquid-liquid extraction. For example, melt refining was specifically designed for short-cooled metallic breeder fuels, which were unsuitable for purex reprocessing. These different approaches to the separation of fission products gave significantly different performance characteristics. In view of the current issues on desirability to reprocess spent fuel from the standpoint of waste management, proliferation, economics, safety, and the potential need to handle reprocessing for advanced breeder fuels, a review of the state of knowledge of purex and non-purex reprocessing appears warranted.

2. Classification and Review

A. Classification

Several methods of classifying nuclear fuel reprocessing techniques have been proposed. Lawroski and Hyman⁴⁵ have proposed a method based on the phases which are contacted. This gives four classifications: 1) solid-liquid, 2) solid-gas, 3) liquid-liquid, and 4) liquid-gas. They have also suggested that processes could alternately be classified in terms of batch or continuous operations, or single stage or multi-stage contacting.

With continuous development in the area of spent fuel reprocessing since that time, two areas of active research can be recognized: 1) aqueous processes, and 2) pyroprocesses. The distinctive difference between these two areas is that aqueous processes are low temperature (room temperature) processes in which all fuel materials are converted to aqueous solutions before any attempt at separation. On the other hand pyroprocesses are high temperature processes, frequently involving either melting the feed materials, or dissolving them in molten salts and/or liquid metals, before carrying out the extraction or separation.

This basic division has been adhered to in this paper. Subdivisions of these two broad categories have been based on the principle involved in the actual separation step, regardless of the nature and amount of treatment required prior to separation. The "principle of separation" concept has been adopted in this classification since it was felt that identification of processes in this manner was least confusing and most meaningful.

As the name implies, in aqueous reprocessing techniques separation is effected after conversion of the materials of interest into aqueous solutions. This is generally achieved by acid dissolution (primarily HNO_3) which serves

as the salting agent as well. The chief separation principle employed is aqueous-organic solvent extraction. The purex process is an example of a process employing this principle. Besides solvent extraction, ion exchange, photochemical, and electrolytic separation also fall within this category. Ion exchange processes are of two types -- anion exchange and cation exchange. Photochemical separation "begins with the selective absorption of a photon resulting in the subsequently enhanced vulnerability of the excited species to oxidation-reduction, ionization, dissociation, or chemical reaction." The required photons could be produced either by conventional photon-emitting equipment coupled with appropriate filters, or by tunable lasers.

In pyroprocesses, the reactants and products are taken to elevated temperatures before the separation is effected. This is done either to exploit desirable properties (or property differences) that exist at elevated temperatures, or to liquefy the system and subsequently bring about a change in the chemical activities.

Pyroprocesses can be first divided into pyrophysical (commonly referred to as pyrometallurgical and used interchangeably in this paper) and pyrochemical techniques. Pyrometallurgical techniques could be defined as processes that use the difference in physical properties, such as distribution between immiscible phases, distillation, fractional crystallization, etc., to achieve separation of the desired elements/compounds. Pyrochemical techniques are based on the relative chemical stabilities of compounds of the elements of interest and which involve selective oxidation and/or reduction reactions.

Pyrophysical techniques proposed hitherto can be divided into processes utilizing one of the four following principles of separation:

1. Fractional distillation involving differences in vapor pressure
2. Fractional crystallization involving differences in melting points
3. Liquid-liquid partitioning which is based on relative solubilities
4. Liquid-solid extraction, in which case one component is converted to a solid compound that will precipitate out of the liquid melt

Pyrochemical techniques can be divided into processes which are based on one of the following separation principles:

1. Electrochemical (pyroelectrochemical)
2. Cyclic oxidation and reduction
3. Selective oxidation and/or reduction

Classification is done to order the basic principles. For example, the relative solubility of a solute in two solvents could be altered by changing its oxidation state, i.e., by selective oxidation. The problem arises when the distinction between pyrophysical and pyrochemical processes is unclear. In such cases the actual separation step determines the category into which the process falls. If the desired separation is achieved by differences in relative solubilities, the process is pyrophysical, even though a prior chemical process like selective oxidation had been carried out. It is clear that most aqueous separations also involve principles such as selective oxidation and relative solubilities, but as has been stated earlier the first basic distinction is between aqueous processes and pyroprocesses.

The classification described is presented as a chart in Figure 7.

B. Review of Spent Nuclear Fuel Reprocessing Techniques.

The Spent Nuclear Fuel Reprocessing Techniques reviewed by the authors is listed in Table 2. Features of each technique, viz., the type of fuel each

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Table 2. Summary of Spent Fuel Reprocessing Techniques Proposed to Date

No.	Classification Category	Name of Method	Reference	Input Fuels	Principle of Separation	First Major Ref. Found by Authors	Stage of Development	Ability to Extract Pu	Affiliation of Work	Remarks
1. Aqueous Processes										
1.1 Solvent Extraction										
1		BUTEX	49	UO ₂ oxide fuels	Solvent extraction	50, Apr. 1952	Lab studies	Yes	Union Carbide Nuclear Co.	Process for recovery and decontamination of Pu from irradiated U. Recovery of uranium is accomplished, but decontamination is not adequate to permit direct handling.
2		Chelation	51	UO ₂	Solvent extraction	51, Apr. 1949	Lab studies	Yes	UC Berkely Rad. Lab.	Pu recovery and decontamination from dissolver solutions and redox wastes.
3		HALEX	52	UO ₂ oxide fuels	Solvent extraction	53, Jan. 1956	Lab studies	Yes	Argonne	Identical to purex, except that CCl ₄ used as diluent for TBP.
4		Purex	54,55	Metallic, UO ₂	Solvent extraction	56, Sept. 1950	Commercial	Yes	Oak Ridge Natl. Lab.	Extensively employed commercially; bulk of R&D effort spent on this. Good separation of U & Pu. Good decontamination.
5		Redox	57	Metallic, UO ₂	Solvent extraction	58, Nov. 1949	Pilot plant	Yes	Oak Ridge Natl. Lab.	Superseded by purex.
6		Thorex	59	Th, ThO ₂	Solvent extraction	60, Jan. 1946	Pilot plant	—	Oak Ridge Natl. Lab.	Solvent extraction process for Th fuels; high decontamination factor achieved.
1.2 Ion Exchange										
7		Ion Exchange	61,62,63	Metallic U, UO ₂ , Pu, PuO ₂	Adsorption-Desorption	64, 1947	Commercial Possible	Yes	Oak Ridge Natl. Lab.	Alternate process in 1942-44 at Hanford for Pu production. Used primarily as tail-end to purex, etc. for further purification and concentration of 253U, 233U, Th, Pu, etc.
1.3 Photo-Chemistry										
8		Photochemical Induced Separation	65,66	Any type that fits purex	Selective oxidation or reduction	67, March 1975	Conceptual	Yes	Brookhaven	Still in conceptual stage, viewed as modification to purex tail end to permit almost complete separation of actinides.
1.4 Electrolysis										
9		Flurex	68,69	Metallic, oxides	Electrolytic (Aqueous)	70, 1955	Lab studies	Possible, with great modification	Hanford	Designed for U recovery from NH ₄ UF ₅ .
1.5 Precipitation										
		Bismuth Phosphate Precipitation	71	Metallic U, Pu	Precipitation	72, 1942	Military purpose plant	Yes	Hanford	Basically for separation of Pu from U & fission products. This has been expanded for recovery of other fission products individually.
2. Pyroprocesses										
2.1 Pyrophysical										
2.2 Fractional Distillation										
11		AEROX	73	UO ₂	Volatility	73, Feb. 1963	Lab studies	Not possible	Atomics International	Low decontamination, rapid recycle; remote refabrication needed, suitable only for UO ₂ fuels.
12		DEBOER	74	Metallic U U alloy	Volatility	74, May 1957	Lab studies	No	Knolls Atomic	Decontamination from all long-lived fission products except Zr by formation of UI ₄ . By selective condensation of UI ₄ , U & Zr can be separated slowly.
13		Fluoride Volatility	75,76	UO ₂ PuO ₂ , Alloys Metallic U, UC	Volatility	77, 1956	Pilot plant	PuF ₆ extraction possible. High purity.	Argonne	Along with chloride and bromide volatility, this method has been the subject of much research. More activity in France & USSR than in USA in recent years.
14		Molten Salt-Fluoride Volatility	78	Metallic U or U-Alloy	Volatility and sorption-desorption	78, Apr. 1971	Lab studies	Difficult	Oak Ridge Natl. Lab.	For recovering decontaminated U from U-Al fuels with Al cladding.
15		Nitrofluor	79	Alloys, UO ₂	Volatility	80, 1964	Lab studies	PuF ₆ extracted together with some F.P. Pu distillation possible.	Brookhaven	Non-aqueous fluoride volatility U, Pu separation and purification; kilogram amounts of fuel have been tested and studied.
16		Carbox	81	UC	Separation only of f.p. volatile at high temp.	82, Feb. 1965	Lab studies	Pu cannot be removed	Atomics International	Specially developed for UC fuels. Low decontamination recycling. Only volatile fission products removed by separation.

Table 2 (Cont'd)

No.	Classification Category	Name of Method	Reference	Input Fuels	Principle of Separation	First Major Ref. Found by Authors	Stage of Development	Ability to Extract Pu	Affiliation of Work	Remarks
	2.1.2 Fractional Crystallization									
17		Hermox	61,83	U, U Alloy, Th	Fractional Xtallization with solvent	84, Aug. 1957	Lab studies	No	Oak Ridge Natl. Lab.	Suitable for metallic uranium, low decontamination.
18		Hydride Separation	48	Metallic U	Sieving	85, March 1957	Lab studies	Pu-rich phase can be separated. Very low purity.	Ames, Oak Ridge	Poor separation, only lab studies. Kinetic rates expected to be very slow since processing will probably be in the solid state.
19		Pyrozone	86	Metallic U	Fractional Xtallization	87, June 1957	Lab studies	No	Argonne	Suitable for metallic fuels; Pu not separated. For U recycling only.
20		Zone Melting	88	Metallic U	Fractional Xtallization	89, 1952	Lab studies	Not possible	Argonne	Very slow; applicable only for the production of small quantities of high-purity metals for laboratory purposes.
	2.1.3 Liquid-Liquid Partitioning									
21		An extraction of Pu & fission products	90	Metallic U	Liquid-liquid partitioning	91, 1955	Lab studies	Low extraction ability for Pu contamination by other fission products.	Ames, Iowa	Low decontamination. Separation of Pu improves with addition of gold. Economic feasibility not expected for large scale separation.
22		Buffer Method	92	Metallic U U Alloys	Liquid-liquid partitioning	93, 1955	Lab studies	Doubtful	Brookhaven	Designed to separate U and other fission products. Possible to modify to remove Pu from fission products.
23		Fused Salt-Liquid Metal	94	²³³ U-Th	Liquid-liquid partitioning	95, 1961	Lab studies	—	Ames, Iowa	For reprocessing of ²³³ U-Th fuels, separating ²³³ U from Th.
24		Mg extraction of Pu from molten U	46,45	Metallic U or U alloy	Liquid-liquid partitioning	96, Aug. 1955	Lab studies	Process designed for Pu extraction. Purity of product low.	Atomics International	Suitable for metallic fuels, low decontamination for recycling. Only lab study reported.
25		Salt Transport Process	97	UO ₂ , PuO ₂	Liquid-liquid partitioning	98, Aug. 1969	Lab studies	Pu separable	Argonne	High decontamination developed for LMFBR but could be applied possibly to other fuel types with suitable modification.
	2.1.4 Liquid-Solid Extraction									
26		DAP	99	Metallic U, Pu U, Pu Alloys	Liquid-solid separation	100, Aug. 1960	Lab studies	Possible, but low purity	Dow Chemical Co.	Dow aluminum pyrometallurgical process. U is dissolved in molten Al and precipitated as UAl ₃ . Separate step required for Pu recovery. Applicability to oxide and carbide fuels doubtful.
27		Parlee-Anderson	44	Metallic, UO ₂ , UC	Liquid-solid separation	44, 1969	Lab studies	Possible if U-Pu separation step added	Stanford	Lab studies only, suitable for in-situ reprocessing of reactor fuels. Separate U-Pu from fission products.
	2.2 Pyrochemical									
	2.2.1 Electrochemical									
28		Electrorefining	101,102	Metallic U or mixed salt	Electrochemical (Pyro)	47, 1969	Lab studies	PuCl ₃ removed as vapor Pu stays in salt. Possibility of Pu extraction by adding another cell.	Kroll Atomic Power Laboratory	Only lab studies, though work not followed up in the US. Current research in USSR quite extensive. Not suitable for ceramic fuels and carbon coated fuels.
29		Molten Salt Electrolysis	82,103,104	UC	Electrolysis	105, March 1963	Lab studies		Atomic International	Application of molten salt to reprocessing of UC fuels.
30		Salt Cycle Process	106	UO ₂ , UO ₂ -PuO ₂	Electrolysis	107, Sept. 1961	Pilot plant	PuO ₂ crystals deposited, recovery = 99.5%	Battelle Memorial	Research began in 1956; lab and pilot plant studies have been done.
	2.2.2 Cyclic Oxidation Reduction									
31		Nitride-Carbide Cycle	108	UC	Oxidation-reduction	108, Aug. 1969	Lab studies	No	Atomics International	For processing of UC fuels only. Low decontamination pyrochemical method. High Pu losses.
	2.2.3 Selective Oxidation									
32		Melt Refining	109	Metallic U	Volatility and selective oxidation	110, 1955	Pilot plant	Pu not removed	Argonne Natl. Lab.	No U-Pu separation, decontamination by volatilization or oxidative-slugging. Experimental processing done at EBR-II for few years. Remote fabrication of fuel rods carried out successfully.

technique is capable of processing, the principle of separation and the references consulted are listed.

The fact that the various reprocessing techniques have been developed to different levels poses problems in a review of this nature. While on the one hand some processes are at the conceptual level, and are totally devoid of experimental verification, on the other hand some have been pilot-planted, and are available for scale-up; yet others are at the commercial scale. These differences are noted in the column "Stage of Development."

The ability to extract plutonium is directly linked to nuclear proliferation. In order to identify the military potential of each process, the "Ability to Extract Pu" column has been added. The history of each process, and the affiliation of the development work is included in so far as it is available.

The classic works by Manson Benedict,¹¹⁰ Thomas Pigford¹¹¹ and J.T. Long¹¹² were often consulted, and are included in the reference list.

3. Separation Processes

A. Aqueous Processes

The feed material is converted to aqueous solutions by dissolutions before separation is undertaken. The dissolving agent used has to be compatible with the separation process employed.

A review of the five aqueous processes follows.

(1) Solvent Extraction

The application of solvent extraction in nuclear-fuel reprocessing involves a sequence of transfers of one or more solutes from one liquid phase to another, essentially immiscible phase.

The purex process is probably the best developed of the various solvent extraction processes for nuclear-fuel reprocessing. Figure 8 is a flow-sheet of the purex process which utilizes solvent extraction to decontaminate, separate, and purify uranium and plutonium from dissolved irradiated slugs. A solution of 30 vol % TBP (tributyl phosphate) in a kerosene-type diluent is employed as the solvent, with nitric acid as the salting agent. A separation is accomplished because of the extractability of uranyl nitrate and plutonium (IV) nitrate, and the relative inextractability of fission products and plutonium (III) nitrate. Therefore, by changing the oxidation state of plutonium, it can be separated from uranium.

The heart of the purex process consists of two solvent extraction cycles for gross decontamination of each product: one cycle providing decontamination and partitioning of uranium and plutonium; the separate second cycle providing further decontamination of the separated products.

The solvents used with aqueous solutions are usually organics since it is necessary that the extracting solution be relatively insoluble in the leaching solution and also be capable of a clean separation; it is important that the extracting solution not form an emulsion. The extractant must be highly selective in its ability to react with or absorb the desired material. The specific requirements governing the choice of a suitable solvent are as follows:^{110,111,112}

1. Low extractability of fission products and relatively high extractability for uranium, plutonium, or thorium, according to the process involved
2. Low mutual solubility with the aqueous phase in contact
3. Good chemical stability in the system

4. Adequate radiation stability
5. Low vapor pressure and high flash point
6. Suitable density, viscosity, and interfacial tension in the sys-

tem

7. Easy purification for reuse
8. Low toxicity
9. Relatively noncorrosive
10. Available in needed quantities at reasonable cost

The solvent for the purex process is the organic complexing compound TBP, a relatively viscous liquid. To produce an organic phase of satisfactory viscosity, density, and surface tension, it is customary to dilute TBP with an appropriate inert hydrocarbon kerosene or dodecane. Carbon tetrachloride has also been used as a diluent, because of its nonflammable nature and its high density, in cases where it is desirable to operate with the organic as the heavy phase.

The solvent is brought into intimate countercurrent contact with the aqueous feed solution where the TBP extracts the uranium and plutonium into the organic phase, leaving the fission and corrosion products in the aqueous solution. The organic solution is selectively stripped of plutonium with dilute nitric acid after reducing the plutonium valence from +4 to +3. The uranium is then stripped in a third contactor. The plutonium is further purified by additional extraction cycles or by ion exchange.

The uranium and plutonium may be precipitated from their dilute nitric acid solutions and converted to oxides by thermal decomposition, or they may be directly converted to oxides by the sol-gel process, in which the nitric acid is removed from aqueous solutions of plutonium or uranium by extraction with an amine

or heavy alcohol. In the sol-gel process the uranium or plutonium, as the acid extraction proceeds, gradually forms into a colloidal suspension or sol. This can be handled like a true solution. Progressive removal of water by evaporation or by extraction with a hygroscopic solvent converts the sol to a plastic gel. Sols of plutonium and uranium can be combined and gelled together to form an oxide in which the two elements are homogeneously dispersed. When fired to about 1200°C, the gel approaches theoretical density and is suitable for fabrication into reactor elements.

(2) Ion Exchange

Ion exchange is merely the exchange of ions that occurs across the boundary between a solid particle and a liquid. When certain substances are dissolved in water, they become ionized. An equal number of positively charged ions and negatively charged ions are formed, and the solutions conduct electricity. Such substances are called electrolytes. Dissolved electrolytes, when in contact with ion-exchanger solids, will exchange stoichiometrically equivalent amounts of ions of the same sign. Ions from the solid migrate from sites on the surface and even from the interior of the solid. Materials that exchange cations with electrolytes are known as cation exchangers; those that exchange anions are called anion exchangers. Certain substances, capable of exchanging both cations and anions, are termed amphoteric ion exchangers.

The processing steps involved in ion exchange applied to nuclear-fuel reprocessing are:

1. Feed sorption or loading
2. Selective elution or scrubbing
3. Product elution or stripping
4. Resin reconditioning

These steps are carried out consecutively either by changing the solution flowing to a fixed bed or resin or by moving the resin through different solutions.

G. E. Boyd and J. Schubert⁶³ describe the first use of ion exchanges for separating plutonium from uranium and the fission products. Their schematic flow sheet is shown in Figure 9. The four steps in this flow sheet follow:

- a) A solution of uranyl nitrate was passed through a deep column or bed of cation exchanger until plutonium α -activity appeared in the effluent. Plutonium was taken up by the exchanger long after uranium break-through, and after the bed became saturated with uranyl ion.
- b) Uranium was removed from the exchanger by differential elution.
- c) The fission products were eluted with no loss of plutonium from the column. Eluants were employed which, by a combination of complex ion formation and cation exchange, desorbed the fission products but, because of the large affinity of the Pu(IV) cation for the exchanger, did not remove the latter.
- d) In the final step an eluant was used which quantitatively displaced the plutonium in a minimum volume. Some of the fission products accompanied the plutonium in this step, so that additional processing was necessary to effect a complete decontamination from β - and γ -activity.

The most important use of ion exchange in fuel reprocessing today is in the tail-end purification and concentration of plutonium following solvent extraction processing. Ion exchange can also be used for the separation and recovery of individual fission products. Various ion-exchange techniques have been developed for the recovery and concentration of specific constituents from solvent-extraction outlet streams. Both anion and cation exchangers are used for plutonium recovery and purification. In the thorex process, ion exchange was used to recover and concentrate the ²³³U product.¹¹³

Figure 9. Flowsheet of an ion-exchange process for the separation of plutonium.

Neptunium (IV) recovery by anion-exchangers¹¹⁴ gave decontamination factors of greater than 10,000 for uranium, plutonium, and common metallic contaminants; greater than 25,000 for fission products normally expected in the feed; and greater than 1,000 for thorium. Cesium, strontium, technetium, and promethium too are recoverable by the use of appropriate ion-exchangers.

Bobleter et al¹¹⁵ report the development of an ion-exchange apparatus for automatic separation of uranium fission products for analytical process control of the separation process of uranium (and plutonium) from the fission products in reprocessing plants. A sample is taken either from a process vessel, or a bypass, and first separated into the groups U, Zr, Nb, Mo, Te and Cs, Sr, Y, Ce. From these groups the individual elements are isolated by a step-wise elution with mineral acids.

(3) Photochemical Techniques

Use of photochemical techniques is possible because of the discrete energy level band structure of the nuclear waste materials. Because processes involving the selective excitation of one or more species to a higher quantum state have not been previously developed into an aqueous reprocessing scheme, emphasis has been placed on this technique. Excited species exhibit enhanced vulnerability to ionization, dissociation, or chemical reactivity, which allows them to be separated from the feed material by physical or chemical means. Such excitation is possible because the frequencies of the absorption maxima in the optical spectrum are generally unique for most ions in solution.

Use of an ion's entire spectrum is not required to carry out a given photochemical reaction. For a given actinide ion, the wavelength and monochromaticity of the excited laser or filtered lamp light required to produce the excited state, which undergoes the desired photochemical reaction, will be deter-

mined by the electronic absorption spectrum of the ground-to-excited state transition and the absorptions of any interfering solution components. One of the major advantages of a separation scheme based on photochemical reactions is that a component in a multicomponent mixture can be selectively excited and will react with a given component to the exclusion of all others.⁶⁵ The only limitation on this photoselectivity is that any other component absorbing at the wavelength of interest should not undergo interfering photochemistry.

The feasibility of photochemically fractionating the actinides in nuclear waste processing has been evaluated on a preliminary basis. The data indicate that there are several potentially useful photoredox reactions for these purposes. There is a lack of data on some photochemical parameters for the solution conditions existing in high level waste processing; however, there should be little or no question about the data relevant to processing most other nuclear waste streams. This evaluation deals mainly with the processing of spent fuel rods from nuclear power reactors.

The photochemistry of the actinides has been studied since the 1830's,¹¹⁶ with the bulk of the work being done on uranium ion systems. The photochemical behavior of the uranium systems serves as a basis on which the neptunium and plutonium systems can be compared and correlated. Although the literature on plutonium photochemistry is limited, data are sufficient to support the expectation that the photochemistry of plutonium and uranium will prove to be similar.

The photodecomposition of the nitrate ion occurs in aqueous nitric acid solutions with UV light. The nitrite ion photogenerated in some systems offers a potentially useful photochemical source of this reductant, which is required in portions of the purex process. This photogenerated nitrite ion could re-

place the addition of materials that leave a residue such as NaNO_2 in step 1 of the purex process. The effects of radiation-produced excited states and radiation-initiated thermochemistry on product yields were deemed to be negligible under most process conditions.

Since uranium is a major solution component, plutonium separation may be accomplished, as shown in Figure 10. In addition the oxidation states of neptunium and plutonium ions may be selectively altered directly by inputting light directly into the process stream and leave UO_2^{2+} unchanged. To avoid radiation effect problems, uranium photochemistry could be used to carry out the plutonium reduction in step 2 of the purex process as presented in Figure 10.

A conceptual photochemical process design of a backup system for the AGNS electrolytic reduction is described by Goldstein et al.⁶⁶ If the AGNS system fails after start up, very difficult remote maintenance techniques would be required to alleviate the problem. Therefore, a scheme that takes a portion of the purified U(VI) outside the shielded area and reduces it photochemically has several advantages.⁶⁶ This modification of the purex process flowsheet is depicted in Figure 11.

At present purifying plutonium in the series of steps illustrated in Figure 11 requires additives (NH_2OH , NaNO_2 , etc.) to oxidize the plutonium to a plus-four state. This oxidation can be done very economically and effectively by only adding UV light, thus reducing safety problems from explosive-forming additives as well as simplifying and improving this process.¹¹

(4) Electrolytic (Low Temperature Electrolysis)

In the electrodialytic process, known as the flurex process,⁶⁸ uranyl and nitrate ions of the feed solution are electrodialytically separated from each other, and uranyl ions are reduced at a mercury cathode to the tetravalent

Figure 10. Photochemical modified purex process.

Figure 11. Flow chart of purex process which has been modified with photochemical reduction of uranium to reduce Pu IV to Pu III.

state. The separation of uranyl and nitrate ions must be effected if reoxidation of U(IV) ions is not to occur.

With an arrangement as in Figure 12, in which the central or feed compartment is bounded by a cation (Permutit 3142) membrane and an anion (Permutit 3148) membrane, an applied potential transports uranyl ions into the cathode chamber. Here the uranyl ion is reduced to U(IV) and is precipitated as NH_4UF_5 . Water accompanies the negative and positive ions into the respective compartments, but 90% of the water is moved through the cation membrane. Hydrofluoric acid and ammonia ion fluoride are continuously added to the catholyte to maintain the concentration of these reagents.

The following reactions are thought to apply:

Under typical process conditions the membranes have electrical resistances of 5 to 40 ohms/cm² and maintain their electrical properties and permeability selectivity at temperatures up to 60°C.

(5) Precipitation Processes

A precipitation process was the first process used on a large scale for the recovery of plutonium.⁷² The bismuth phosphate process was the first precipitation process to recover plutonium from irradiated natural uranium and is depicted in Figure 13. This took place in a series of steps that utilized the difference in chemical properties of Pu(IV) and Pu(VI). In the (IV) state, plutonium was separated from uranium and most of the fission products because of the insolubility of the Pu(IV) phosphate, along with the ability of bismuth phosphate to carry insoluble plutonium phosphate. In the (VI) state, plutonium

Figure 12. Flowsheet for the electrodialytic preparation of NH_4UF_5 .

Figure 13. The Bismuth phosphate process.

in solution was further decontaminated by the precipitation of insoluble fission-product phosphates or fluorides. The decontamination steps could be repeated as often as necessary by carrying out a series of oxidation-reduction steps. After adequate decontamination the plutonium was concentrated by precipitation as Pu(III) or Pu(IV) fluoride, metathesized to plutonium hydroxide and isolated as the dioxide.

Uranium was present in the bismuth phosphate process as the uranyl ion (UO_2^{+2}) formed when the uranium metal was dissolved in nitric acid. The nitrate, chloride, sulfate, and fluoride of UO_2^{+2} are quite soluble, but uranyl phosphate is relatively insoluble in acid solution; it precipitates slowly from nitric acid solution when a phosphate ion is added. Sulphuric acid was added to the solution to prevent precipitation of uranium during the plutonium precipitation step. Uranyl and sulfate ions form a complex that keeps the uranium in solution when a phosphate ion is added to precipitate plutonium and bismuth. This process has become obsolete because of the development of the redox and purex processes.

B. Pyroprocesses

Pyroprocesses involve the application of heat and may utilize temperatures, one use of which is the distillation of plutonium from uranium. Some volatility separations, however, maybe effected with very mild applications of heat. Pyroprocesses conveniently subdivide into pyrophysical and pyrochemical processes, the former being primarily physical and the latter chemical in nature. Separation processes that depend on distillation or sublimation, volatilization, or liquid-metal extraction are primarily physical in nature. Others, such as slagging and fused-salt extraction, are primarily chemical in nature.

Pyrophysical Processes

Pyrophysical, often referred to as pyrometallurgical processes, employ differences in the physical properties of the materials concerned to achieve separation. These physical properties include melting and boiling points, densities, vapor pressures, solubilities, and the like.

(1) Fractional Distillation - Vapor Pressure Differences

Volatilization of uranium hexafluoride at moderate temperatures is an easy way of separating it from nonvolatile fluorides. Fractional distillation of the overhead vapors can effect additional concentrations of uranium. Table 3 shows the melting and boiling points of some of the fission-product fluorides and of uranium fluoride.

The great majority of the fission products form fluorides which volatilize only at high temperatures. Among those which contribute appreciable activity to irradiated reactor fuel, only the fluorides of tellurium, molybdenum, niobium, and ruthenium boil or sublime at low or moderate temperatures. Of the fluorides which volatilize more readily than uranium hexafluoride, only tellurium hexafluoride is a gas at ordinary temperatures so it can be readily separated. Any molybdenum hexafluoride that may be volatilized and condensed with uranium hexafluoride is not serious since the most important radioisotope, molybdenum-99, has a half-life of 67 hours and will decay almost completely in the course of a few weeks.

Figure 14 is a schematic flow sheet for the processing of $(U,Pu)O_2$ fuels by means of fluoride.¹¹⁸ The purpose of oxidation is to increase the surface of the fuel (UO_2 pellets) exposed to the reagent (fluorine) and facilitate heat transfer. This reaction is relatively exothermic.

Figure 14. Flowsheet for processing (U,Pu)O₂ fuels by means of fluoride volatility.

Prusakov et al.¹¹⁹ have used a dual temperature system to separate uranium and plutonium in the fluoridation step. Due to the small excess of fluorine and to the low filter temperature, the contamination of uranium hexafluoride by plutonium is very low. The addition of a sodium fluoride trap at 300°C elimi-

Table 3	
Boiling Points of Fluorides (120)	
Fluoride Boiling Pt (°C)	
TeF ₆	-35.5
IF ₇	4.5 (Sublimes)
MoF ₆	35
TcF ₆	55.3
UF ₆	56.2
NpF ₆	56.4
RnF ₆	62.2
PuF ₆	62.3
IF ₅	98
SbF ₅	149.5
NbF ₅	236
RnF ₅	250

nates trace amounts of plutonium from the uranium stream. During the second period plutonium is volatilized and condensed while fluorine is recycled. Two cold traps at 65°C collect about 99% of the plutonium hexafluoride. Sodium

fluoride sorption - desorption is utilized to separate maximum hexafluoride from fluorination off-gases, plutonium hexafluoride, and also from some fission products.

The final purification of uranium is performed by two distillation columns: one for low-boiling and one for high-boiling fission products. However, neptunium and technetium need special techniques for separation. As their sorbing points are too close to that of uranium hexafluoride to allow a sharp separation. Technetium can be separated by absorption on high specific surface magnesium fluoride. In addition to magnesium fluoride, bromine too may be used for the separation of neptunium.

Recycling of fluorine is necessary because of the quantity of contaminated waste which would be generated and the cost of the reagents. The reagent loop requires purification and a fission-product collection system.

(2) Fractional Crystallization

Fractional crystallization can be accomplished with or without a solvent. By "fractional crystallization without added solvent" is meant a separation whereby minor constituents are physically segregated by reason of their unequal distribution between a liquid metal and the solid phase in equilibrium with it.

The zone-melting process, developed by Pfann¹²¹, involves separation of solute elements by slowly moving a narrow melted zone through a bar of solid material. This method may be used to remove trace elements from a metal or material and to provide material of higher purity than would otherwise be obtainable. This method has been studied by several people for purification of commercial uranium, separation of fission products and plutonium from uranium, etc. However, it has been found to be applicable only to the production of small

quantities of high-purity metal for laboratory purposes. A major deterrent to the commercial application of this process is its slowness.

By the addition of a metallic solvent to the system, usually in large excess, the solubility relations, the nature of the phases formed, and the operating temperatures become items that can be varied by the experimenter to enhance the desired separability. One such method is the pyrozinc process,⁸⁶ which uses zinc as the solvent. It utilizes differences between the solubility of uranium and of other elements in zinc to achieve separation by fractional crystallization.

Figure 15, a portion of the U-Zn phase diagram, shows two, two-phase regions. One or the other of these phases is thought to contain the compound U_2Zn_{17} or UZn_{11} , depending on the temperature. This intermetallic compound exists in equilibrium with a solution of uranium in liquid zinc, the solubility decreasing with a decrease in temperature from about 13 w/o U at 910°C to approximately zero at 500°C. These facts are utilized in applying the pyrozinc process to the reprocessing of fuel elements, as indicated in Figure 16.

The first step is dissolution of sufficient spent reactor fuel in zinc at about 700°C to form what in practice should be a solution containing uranium in a concentration of the order of 0.3 w/o. The next step is to cool slowly to 500°C, crystallize the U_2Zn_{17} or UZn_{11} , and filter the crystals away from fission products. If fission products or other impurities coprecipitate appreciably with the U_2Zn_{17} or UZn_{11} , several recrystallization and filtration steps may be required. The third and final step is the recovery of pure uranium either by removal of the zinc into a liquid magnesium extractant or by distilling the zinc away from the uranium at 945°C.

Figure 15. Phase diagram of uranium-zinc system showing the regions of stability of U_2Zn_{17} and that is represented by the delta (δ) phase while the epsilon (ϵ) phase is UZn_{11} .

Figure 16. Flow diagram of the pyrozinc re-processing scheme.

(3) Liquid-Metal Extraction

The well known principles of liquid-liquid partitioning are equally applicable to liquid-metal partition studies. The technical difficulties of continuous multi-stage contacting of liquid metals make it desirable to employ systems with partition coefficients for the transferred material that are sufficiently large, so that a small number of batch contacts brings about the desired separation.

In theory, any pair of immiscible liquid metals might be examined for separation possibilities, e.g., the partition of uranium, plutonium, and fission products between unsaturated solutions in zinc and lead. The fused salt-liquid metal technique proposed by P. Chiotti⁹⁴ employs this principle of effectively transporting solutes from one liquid metal or alloy to a second liquid metal or alloy by bringing the two in contact with an appropriate fused salt. It is also possible to transport solutes between two liquid salt phases by bringing them in mutual contact with an appropriate alloy.

The process indicated in Figure 17 was designed for the reprocessing of Th-²³³U fuels from breeder reactors. It may be broken down into the following eight steps:

1. Dissolution of the thorium in magnesium at 800-900°C to form a Mg-42%Th eutectic solution (mp 582°C) and a finely divided uranium-rich (99+ without U) precipitate. At 585°C the solubility of uranium in the eutectic solution is only 40 ppm by weight.
2. Separation of the magnesium-rich and uranium-rich phases at 600-585°C.
3. Consolidation of the uranium-rich precipitate at 860-900°C by addition of chromium and formation of U-5.0 without Cr eutectic (mp 860°C).

Figure 17. Flowsheet of Th - ^{233}U fuel re-processing method.

Figure 18. Flow diagram of proposed Parlee-Anderson molten tin reprocessing scheme.

4. Purification of the uranium-rich phase by oxidation-reduction reactions in a KCl-LiCl/Zn system to separate the uranium from Cr, Mo, Nb, Zr, Ru, and other more noble metals. The purified uranium is separated as U_2Zn_{17} and the zinc removed by evaporation.

5. Treatment of the Mg-Th solution with KCl-LiCl-MgCl₂ salt to remove yttrium and rare earth metals.

6. Precipitation of thorium from the magnesium solution at 675°C as ThH₂ with hydrogen at 1 atm.

7. Separation of the ThH₂ precipitate and recovery of the thorium by heating under reduced pressure at 700°C to remove hydrogen and magnesium.

(4) Liquid-solid Extraction

This category covers separations based on metal systems in which one phase is a solid insoluble in another liquid metal. Clean separations of two liquid phases may require that the interface be accurately located -- an operation that is difficult to perform remotely. In the liquid-solid case, however, the solid may be suspended in the liquid and subsequently removed by filtration. Alternatively, it could either float or sink in the liquid, in which case it could be removed by skimming, or by draining off the supernatant liquid, respectively.

A separation employing these principles has been described by Anderson.⁴⁴ This process calls for dissolving the spent nuclear fuel in liquid tin, followed by selective nitriding of the uranium. The fission products would either remain dissolved in the liquid tin, or would form nitrides that float; whereas the UN and U₂N₃ precipitates would sink with the plutonium nitride. A schematic flow diagram of this process is depicted in Figure 18. The uranium is recovered as a nitride, which then needs to be either reduced to its metallic state, or converted to oxide, carbide, or halide for enrichment, as the case may be.

Pyrochemical Processes

In contrast to pyrometallurgical processes, pyrochemical processes exploit the relative chemical stabilities of the elements of interest, and/or their compounds, to achieve separation. These invariably involve selective oxidation and/or reductions reactions.

(5) Selective Oxidation

If the term oxidation is used in its broad sense as meaning an increase in the valence state of a metal, then most of the pyrometallurgical work is an example of oxidation. Selective oxidation as it is used in this section only includes reactions to form oxides, carbides, nitrides, sulfides, and halides. Only those methods where the actual separation is effected by this oxidation reaction will be discussed here.

One such well known method is melt refining,¹⁰⁸ which is a process of selective removal of fission products from highly irradiated metallic reactor fuels by a high-temperature oxidation step in which the ceramic crucible is the source of oxygen. Volatilization of fission-products contributes to the overall fission-product removal.

The irradiated, chopped fuel and a precalculated quantity of re-enrichment uranium is charged to a lime-stabilized zirconium oxide crucible. The crucible is placed in a sealed furnace, and the fuel alloy is melted and liquated under an inert argon atmosphere. During the run, the crucible is covered with a ceramic-fiber fume trap effective in trapping cesium and iodine. After purification, the fuel alloy is chill-cast into a graphite ingot mould.

Melt refining removes three classes of fission products from the fuel, and these represent about two-thirds of the total fission product yield. The chemically inert fission gases, xenon and krypton, are evolved on melting. Fission

products such as bromine, iodine, and cesium, which have high vapor pressures at 1400°C, are volatilized and trapped by the fume trap. Chemically reactive fission products, such as yttrium, strontium, barium, lanthanum, and the lanthanide elements, react with oxygen in the zirconia crucible to form oxides. These oxides remain with the crucible when the alloy is poured into the mold. One group of fission products, atomic number 40 through 46, is not removed by the melt refining process.

(6) Cyclic Oxidation-reduction

The low decontamination AIROX process is based on the oxidation of the fuel to U_3O_8 by oxygen and reconversion to UO_2 by reduction with hydrogen. The phase change during oxidation involves a 30% change in volume, with the result that the fuel is pulverized.

The first oxidation step is adapted to the decladding of UO_2 fuel rods. The cladding is punctured at 1-inch intervals along its length and the contained fuel exposed to oxygen at elevated temperatures (400°C). The increase in volume of the fuel material during oxidation causes the cladding to split and the U_3O_8 to fall free. The declad material is then reduced to UO_2 by hydrogen and blended with enriched UO_2 to restore nuclear reactivity.

By repeating the oxidation-reduction treatment three times, a material with excellent sintering properties is produced. Densities of pellets prepared from this material are consistently above 95% theoretical. Virtually all the xenon, krypton, iodine, cesium, and appreciable amounts of tellurium are removed, but little or no removal of plutonium or rare earths occurs. Ruthenium can be removed by one oxidation above 725°C.

(7) Pyroelectrolysis (Electrochemical)

The processing of fuel elements by electrorefining depends on electro-transport of uranium through a molten bath of alkali or alkaline earth halides, as described by L. W. Niedrach and A. C. Glamm.⁴⁷ In electrorefining, the metallic state of the uranium is momentarily lost as it passes through the electrolyte from anode to cathode. This is an advantage since ideally only those metals that are more electropositive than uranium will accompany it into the salt bath, but they will not readily accompany the uranium as it leave the bath during reduction at the cathode. A flow sheet which uses a manganese cathode and graphite anode is shown in Figure 19. Uranium fuel elements, if clad, would first go to a stripping station, and the cladding would be removed.

During electrolysis the uranium and active fission products would be anodically dissolved; the more noble fission products would accumulate in the bottom of the basket as an anodic sludge. The uranium would then migrate through the salt bath and be deposited upon the manganese cathode, while the active fission products accumulate in the bath. Alloying of the uranium and manganese causes the molten alloy formed to drip from the cathode to the bottom of the crucible.

The product alloy is to be removed from the cell periodically by tapping or other suitable means. The alloys, which in this case would be 5 - 10 w/o manganese, would then be transferred to a volatilization crucible and heated to about 1300°C to remove the manganese. The molten uranium then runs to a casting crucible and from there to molds in preparation for recycle.

Kazantsev et al.¹⁰² report on the "electrolytic production of alloys of uranium with nickel in molten sodium and potassium chlorides." Liquid alloys of uranium with nickel are produced by the electrolysis of molten chlorides of the

Figure 19. Flow diagram of proposed fused-salt electrolysis reprocessing method involving a dripping manganese cathode.

alkali and alkaline earth metals, containing oxygen-free chlorides of uranium, using metallic nickel as the cathode at temperatures of 800-900°C. The most uranium-rich alloys were found to form at the maximum current density and substantial cathode potentials, corresponding to the deposition of pure uranium.

A similar electrochemical process has been proposed by W. N. Hansen¹⁰⁴ for processing uranium carbide based on molten salt electrolysis. The uranium carbide is first dissolved at the anode of an electrolysis cell, and the uranium metal is formed as a dendritic mass at the cathode. The cathode deposit is then immersed in hot mercury. The uranium readily reacts to form a quasi-amalgam which separates from the occluded salt phase. The quasi-amalgam is heated in an atmosphere of propane. The uranium reacts readily to form stoichiometric UC, and the mercury is distilled away.

4. Process Development Activity

Figure 20 is a plot of the dates of the first major reference found for each new process. Between 1940 and 1950 five new processes were proposed. Between 1950 and 1960 fifteen additional processes were proposed; between 1960 and 1970 ten new processes were proposed which brought the total number to 30. However, the 1970's has produced only two new variants. Most of the conceptual work seems to have been completed in the fifties and sixties. In the 1970's the concentration seems to have been primarily on further development of a restricted few processes.

A notable feature of the various spent fuel reprocessing techniques studied is that the vast majority of them have been custom-designed for a particular type of fuel material or a specific purpose, or both. For instance, the AIROX method has been designed specifically for low decontamination pyrochemical reprocess-

Figure 20.

ing of UO_2 fuels. There does not appear to be an universal process capable of processing any type of feed material, giving out material in the exact desired form. Moreover, undue emphasis seems to be placed on the desired output, with very little attention, paid to the form of waste material generated by a given process.

The processes studied fall into one of the following categories:

- i) Processes designed for separation of uranium only
- ii) Processes designed for separation of plutonium only
- iii) Processes designed for separation of uranium and plutonium only
- iv) Processes designed for concentration of tail-end products
- v) Processes designed for low-decontamination fast recycle

Most of the pyroprocesses, with the exception of the halide volatility and molten salt extraction techniques, offer low decontamination of poisonous fission products and fast recycling time. This will save out-of-reactor inventory costs, but the penalty has to be paid in remote refabrication of the fuel elements. The melt refining process for EBR-II is one such process which was successfully run for several years.

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Several of the pyroprocesses have been designed for processing metallic uranium fuel, resulting in their being phased out with the advent of oxide and carbide fuels. However, it must be reiterated here that if the advantages offered by such methods were essential, reduction of oxidized fuels to their metallic states before reprocessing could easily be considered. The bismuth phosphate process was developed primarily for the separation of plutonium from spent reactor fuels. This technique has since been applied to the isolation of other fission products as well. Ion exchange technology, too, has a long history within spent reactor fuels reprocessing. It was originally the alternate

to the bismuth phosphate process for the separation of plutonium. It has since been developed into a tail-end process for isolation and concentration of aqueous solvent-extraction products, i.e., concentration of plutonium or isolation of individual fission products.

Though several processes appear at first sight to offer reclamation of only uranium (and plutonium) by the addition of suitable tail end processes, the extraction of other elements could also be possible. The classic example is the ion-exchange-solvent extraction combination which has enabled not only a 99.9% recovery of U and Pu, but also the recovery and isolation of several other fission products. Research on halide volatility techniques, particularly fluoride volatility techniques, is progressing in this direction. Several other apparently less desirable processes too could probably achieve separation of the fission products if sufficient effort were devoted to it.

Several processes, with the change in reactor fuel types that have taken place, are only of historical interest today. But some others have been the subject of continuous study and development. Solvent extraction methods, especially the purex process which is the only one employed on a commercial scale, have been researched widely. Other processes, such as halide volatility and molten salt, have also been the subject of intense research and several pilot plant studies.

A survey of recent USSR literature shows a surge of interest in electrochemical methods; there are several reports of studies of molten salts for use in electrochemical separations.¹¹⁸ In this survey of available literature up to December 31, 1975, no comprehensive evaluation of the various methods was found. The best comparisons came from qualitative statements at the beginning of various publications highlighting the advantages of the method being proposed.

This review has identified the many approaches that have been suggested for reprocessing spent nuclear fuel. These techniques that were suggested in the shadow of the liquid-liquid extraction process offer different characteristics. Considering advanced fuel cycle materials and new regulatory constraints, some of these processes may now offer a significant attractiveness not apparent earlier.

VIII. ENVIRONMENTAL IMPACTS

A methodology has been developed for estimating the number of health effects resulting from atmospheric releases of radioactive materials during normal operation of nuclear power reactors and related fuel cycle facilities. This new methodology can be used to coordinate environmental and energy policy and to assess technology. This improved technique has been extended to the existing Brookhaven National Laboratory's reference energy system which traditionally measure environmental impacts solely in terms of tons of emissions. The energy and environmental policy assessment methodology can be applied to any energy system contained in these models including various advanced energy technologies. At present this application to the nuclear system has been demonstrated.

This procedure consists of applying the BNL Energy System Network Simulator (ESNS) program to project radiological emission levels coupled to state-of-the-art methods to estimate population dose and resultant cancer occurrences from these emissions. This tripartite methodology for integrating energy, economy, and the environment is depicted in Figure 21. A means of measuring cost in dollars, as well as cost in population dose and health impacts of various environmental and energy policy strategies, is now available:

1. By simulating emissions produced under different environmental controls in the ESNS model.
2. By modeling the mechanisms for distribution of the emissions throughout the biosphere in the RABGAD model (Radiological and Biological Generic Atmospheric Dose).
3. By using a dose response function to obtain the number of health impacts for a given population exposure.

Figure 21. Computerized integrated energy, economy and environmental system.

To demonstrate this new procedure, several alternate LWR fuel cycle scenarios were analyzed. This work was partially supported by the Savannah River Laboratory (SRL) and was used to aid SRL in preparing an environmental impact statement on reprocessing. These scenarios covering the years 1976-2000 included:

Case 1: No reprocessing of spent nuclear fuel (hence no recycle of either uranium or plutonium)

Case 2: Reprocessing and recycle initiated in the early 1980's with one year cooling of the spent fuel before reprocessing

Case 3: The same as Case 2 except with five years' cooling

Total population dose commitments for exposures of total body and bone (which was judged to be the critical body organ), as well as the associated excess cancers, were estimated for the years 1976-2000.

Major Steps in Analysis

The five major steps taken in carrying out this demonstration study are discussed below.

1. Expansion of ESNS Data Base

In order that light water reactors fueled only with uranium (i.e., no recycled plutonium) could be included in the ESNS calculations, it was necessary to develop a data base on radiological emissions from such reactors. This was accomplished separately for PWR and BWR operations, as the nuclear power growth scenarios used in the study included separate information on the installed capacity for each type of reactor. Radiological emission data for PWR and BWR operations with recycled plutonium were also developed, but these have not as yet been included in the ESNS data base; instead, the ESNS calculations involving

U + Pu reactors were performed using the existing data base entries for a generic light water reactor fueled with mixed oxides.

The major source of information for radiological emissions from operating reactors was the "Final Generic Environmental Statement on the Use of Recycled Plutonium in Mixed Oxide Fuel in Light Water Cooled Reactors (GESMO)," issued in August 1976 by the U.S. Nuclear Regulatory Commission.¹⁸ Releases of radioactivity by nuclide, as given in GESMO, were carefully composited into the categories used in the current ESNS data base in order to avoid a restructuring of the ESNS files at this time.

A network of detailed energy flows through the nuclear fuel cycle was added to the existing BNL ESNS model.⁴³ This detailed network is illustrated in Figure 22. The efficiency costs and emissions data were obtained under a separate contract with Teknekron, Inc., and results were obtained using this data and data obtained as part of the CONAES (Committee on Nuclear and Alternative Energy Systems) study being conducted by the National Academy of Science. A large discrepancy was noted between the radon emissions from mining and milling of these two data bases. After careful study and collaboration with W. Grimes of Oak Ridge National Laboratory, the CONAES data were deemed correct and should supersede the old data.¹²²

2. ESNS Calculations

Output from these runs included the yearly radiological emissions from each step in the nuclear fuel cycle, given for the broad categories defined in the ESNS data base. Savannah River data included electrical demands in GWs (from PWR and BWR) and load factors for years 1976-2000 as shown in Table 4.¹²³ This data was converted to BTUs/yr as required by the ESNS model.

URANIUM:

Figure 22. LWR fuel cycle.

Table 4. Conversion of GWe to BTU

Year	GW	<u>PWR</u>	<u>Equiv.</u>	GW	<u>BWR</u>	<u>Equiv.</u>
		<u>Load Factor</u>	<u>BTU</u>		<u>Load Factor</u>	<u>BTU</u>
1976	26	.692	5.381×10^{14}	17	.735	3.737×10^{14}
1977	33	.697	6.880×10^{14}	18	.736	3.962×10^{14}
1978	39	.712	8.305×10^{14}	18	.750	4.038×10^{14}
1979	43	.727	9.350×10^{14}	19	.737	4.188×10^{14}
1980	48	.724	1.039×10^{15}	22	.716	4.711×10^{14}
1981	54	.722	1.166×10^{15}	26	.712	5.537×10^{14}
1982	66	.705	1.392×10^{15}	34	.691	7.027×10^{14}
1983	77	.714	1.644×10^{15}	40	.712	8.518×10^{14}
1984	88	.719	1.892×10^{15}	47	.713	1.002×10^{15}
1985	103	.714	2.200×10^{15}	53	.722	1.145×10^{15}
1986	117	.720	2.520×10^{15}	62	.714	1.324×10^{15}
1987	131	.723	2.833×10^{15}	70	.721	1.510×10^{15}
1988	146	.724	3.162×10^{15}	76	.730	1.659×10^{15}
1989	163	.724	3.530×10^{15}	82	.732	1.795×10^{15}
1990	178	.729	3.881×10^{15}	90	.728	1.960×10^{15}
1991	195	.728	4.246×10^{15}	98	.730	2.140×10^{15}
1992	213	.729	4.644×10^{15}	107	.729	2.333×10^{15}
1993	230	.732	5.036×10^{15}	115	.733	2.521×10^{15}
1994	248	.732	5.430×10^{15}	124	.732	2.715×10^{15}
1995	266	.733	5.832×10^{15}	133	.733	2.916×10^{15}
1996	283	.735	6.221×10^{15}	141	.736	3.104×10^{15}
1997	298	.737	6.569×10^{15}	149	.737	3.285×10^{15}
1998	312	.739	6.896×10^{15}	156	.739	3.448×10^{15}
1999	325	.740	7.193×10^{15}	163	.739	3.603×10^{15}
2000	338	.740	7.481×10^{15}	169	.741	3.746×10^{15}

End use demands were required to drive the model. Based on values given for future reference energy systems in the Sourcebook for Energy Assessment (BNL 50483),⁹⁹ a set of end use demands for the years 1976-2000 was interpolated.

3. Distribution of Emissions by Nuclide

In order to carry out the dose calculations, it was necessary that the emissions given in the various ESNS categories be allocated among the specific nuclides released to the atmosphere. This required careful scrutiny of the data base entries for each step in the fuel cycle, with appropriate assumptions made on the nuclide makeup of categorized emissions. Then this information was used to develop a computer linkage between the Brookhaven ESNS model and the RABGAD model.

4. Calculation of Population Dose Commitments

Doses were calculated using the RABGAD program, which was developed by Teknekron for the U.S. Nuclear Regulatory Commission to estimate exposures from atmospheric radiological releases from generic sites.¹⁸ RABGAD was employed in the development of dose information contained in GESMO, and assumptions made for this study were entirely consistent with those used for GESMO (which was corroborated by checking our RABGAD results with the emissions and doses given by GESMO).

The calculated doses represent the dose which would be received by a population from one year's operation of a plant or facility and take into account the activities (breathing, eating, etc.) of the population during that year. However, the doses include a future integration over the next 50 years of life from any isotope taken into the body that will continue to reside in

the body even after exposure has stopped. Thus, the calculated doses are 50-year commitment doses which result from a year's exposure time.

Doses to the total body and various organs were determined, taking account of all exposure pathways (food, inhalation, direct contact with releases, etc.). Assumptions of population density, meteorological factors, food intake, etc., were made that were consistent with those used for the various fuel cycle facilities in the GESMO analyses.

Detailed information on the methodology and assumptions implicit in RABGAD is provided in Chapter IV, Section J, Appendix A of GESMO and therefore will not be repeated here.¹⁸

5. Health Effects

Because of its widespread use in standards setting, and since no other correlation has achieved universal acceptance for quantitative estimates of health effects at low levels of exposure, the linear, non-threshold dose-response relationship was adopted for use in this study.¹²⁵ Information used to relate the number and types of health effects to population exposure was taken from the report, "Approaches to Population Protection in the Case of Nuclear Accidents," issued by the U.S. Environmental Protection Agency in November 1974.¹²⁶ This reference assumes that approximately 50 percent of radiation-caused cancers are fatal.

In determining the number of excess cancers (i.e., those in addition to the number that result from normal background radiation levels, it was assumed that the critical body organ (the one receiving the highest dose) is bone tissue. This judgment was made based on the dose commitment results presented in GESMO. Total 50-year population dose commitments for exposure of total body and bone, as well as associated excess cancers, are given in Table 7 for each

case on a cumulative basis for the period 1976-2000. Note that the excess cancers shown for the years 1976-2000 do not necessarily occur during that time frame, but instead result from the operation of the nuclear fuel cycle during those years. Excess cancers can occur as much as 50 years hence. Approximately one-half of the excess cancers are generally assumed to be fatal.

Note that short-term, one-year storage of spent fuel followed by reprocessing and recycle leads to a 73 percent increase in the estimated number of excess cancers using the old Teknekron data compared with the no-recycle option. The increase of from 475 to 630 excess cancers was estimated from the corrected radon-222 emissions for the no-recycle option. Note that short-term, one-year storage of spent fuel followed by reprocessing leads to a 51 percent increase in the estimated number of fatalities using this data from the CONAES study.

If the unsmear model¹²⁷ for lung cancer is adopted the number of lung cancers would increase dramatically and could change the qualitative nature of this result possibly equalizing the health impacts from both options. Pohl¹²⁸ has calculated that nearly 400 lung cancers would result from radon emissions by the year 2000. If this unsmear model is correct for low lung doses, then nearly 8 percent of all lung cancers may be induced from natural background radon and its daughters.¹²⁸ However, the far-reaching significance of this result is not associated with the inequality of environmental impacts of the open and closed cycles, but with waste management policies and strategies focused on uranium mill and mine tailings as well as on high level waste. The estimates of radon-222 emissions from phosphate mining indicate that this industry may cause an equal or even slightly larger number of excess cancers than the radon emission from the uranium industry. These estimates, of course, all depend on the validity of the linear hypothesis presented in the BEIR report

(Biological Effects of Ionizing Radiation).¹²⁵ This linear relationship is generally considered conservative, representing an upper limit of health effects. It is possible that the effects of radiation from natural isotopes could be somewhat less than that from other radionuclides because humans may have built up some natural immunity to these disease mechanisms. However, there is no evidence for this at the present time. Since it appears about half of the short term health effects are caused by radon and from the fact that radon emissions will continue from these sources (at some reduced level) for 80,000 or more, the entire waste management policy including radiation standards and criteria for mining and milling (phosphate, uranium, copper, etc.) should be subject to review. Therefore, the need to question the linear relationship and establish its validity or a replacement is very apparent.

Epidemiological data on lung cancers in the high radon-emitting regions of this country indicate that there are only two-thirds as many lung cancers in these regions as in the eastern part of the U.S. where the emissions of radon are much lower. However, there could be other air-quality factors distorting these figures. If the linear hypothesis is correct a substantial number of cancers would be caused by natural background radiation.

The contribution of uranium mill tailings to the natural background has been shown to be relatively small, i.e., 0.06 and 0.19 percent for earth covers of 8 and 2 ft. respectively.¹²² Thus, the total insult from man-enhanced radon emissions may be very small in comparison to those from nature. About 1 percent of the population die every year, and about 1 percent of those deaths (thus .01 percent of the population) is induced from U.S. energy systems in some form.¹²⁹ This is very small compared to the 2.5 percent of population deaths that are attributed to auto crashes¹²⁹ and is, no doubt, much smaller than the

TABLE 7
SUMMARY OF RESULTS (Cumulative Over 1976-2000)

Case	Reprocessing Start	Fuel Cooling	Person-rem		Data		
					Smear		Unsmear
			Total Body	Bone	Teknekron Excess Cancers	W. Grimes Excess Cancers	W. Grimes Excess Cancers
1	Never	--	1.1×10^6	3.7×10^6	475	630	~1000
2	1983	1 year	1.8×10^6	9.2×10^6	820	949	--
3	1983	5 years	1.6×10^6	7.4×10^6	709	854	--

number of deaths attributed to cigarettes. Yet no one can question that the benefits from our energy technologies surpass those from cigarettes.

Grimes has shown that there are several methods of curbing the rate of emissions of radon-222 from uranium tailings piles.¹³⁰ In the present calculation the level of emissions assumed is from a control strategy using an 8-foot-thick layer of earth placed over the tailings pile.¹²² 100 feet of earth would virtually eliminate the emissions, and a 1-inch layer of asphalt would do the same.¹²² How long the asphalt would last, or even the 100 feet of earth, would certainly depend on meteorological conditions, ice ages, and other geophysical and meteorological phenomena.

The long-term effects of radon from such piles would be somewhat more difficult to manage than the long-term effects from high level waste because of the tremendous bulk of the tailings material compared to the highly concentrated form of high level waste. The real culprit in these tailings piles is thorium-230, the parent of radon-222, which has a half life of 80,000 years. Comey¹³¹ and Pohl¹³² have estimated that a very large number of fatalities could occur from a one-year operation of the nuclear fuel cycle by integrating radon-induced lung cancers over 80,000 years, even though the number per year is very small compared to those cancers induced by background. If no society exists or a cure for lung cancer is developed 100 or a 1000 years from now, health impacts could, of course, be greatly reduced.

The problem of isolating this material for extended periods can increase the costs of not only uranium, but also phosphate fertilizer and thus food. Many other mill tailings, including coal and copper, contain thorium-230. And even natural gas contains radon. Other methods of handling radon-emitting tailings piles have been considered, such as mixing the material with cement and

pumping it back into the mine, separating the thorium-230, or placing these tailings piles in the trenches of deep-surface mines. To know whether we should really endure this added expense of handling mill tailings associated not only with the uranium industry but with many other industries, a higher level of scientific confidence in the results will be required. A high priority R&D program to determine dose response curves (including lung cancer model) and health effects from these sources would certainly appear to be in order at this time.

IX. SAFEGUARDS

This section deals with various fuel cycles and reprocessing technologies and how they affect safeguards on an international level; i.e., the potential for these technologies to reduce the safeguards requirements by a corresponding reduction in the risks of governmental and nongovernmental proliferation. It is assumed throughout that either government or nongovernment groups are capable of diverting, hijacking, or siezing strategic nuclear materials, and therefore little distinction is given to these groups in this brief summary. For a detailed approach, see one or several of the following references (15, 25, 26, 37, or 38).

Safeguards is not a well defined term but refers to a whole set of systems for protecting nuclear materials that includes auditing and accounting, automatic instrument checks and inspections, barriers, alarms and surveillance, watchmen, guards and other security forces, personnel clearances and procedures, and by engineering process design of the nuclear facility.

The U.S. safeguards system is set up to (1) prevent theft of nuclear materials, (2) detect theft, (3) recover stolen material, and (4) respond to threats of violence involving nuclear materials. It is, of course, well recognized that, because of the possibly dire consequences of responding to such threats, emphasis should be placed on prevention. At present, the International Atomic Energy Agency's safeguarding procedures are limited; i.e., they are designed primarily to deter governmental diversion by the risk of detection and resulting disclosure to the international community. The Agency's safeguards are based on a materials accounting system, supplemented by containment and surveillance techniques.

The inaccuracy in analytical and reprocessing technology greatly increases vulnerability to diversion because of what is known as "materials unaccounted for" (MUF). Because of the limits of error (LEMUF), i.e., the limits of accuracy in measurement of these processes, it is impossible to determine whether a country has actually diverted enough material to produce one or even several weapons. However, it should be noted that no country that has signed a nonproliferation treaty has, to our knowledge, developed weapons or diverted material for this purpose. And of course reprocessing is still in its infancy. Fear of inadequate international safeguards, coupled with increased terrorist activity in the world, has led several nations--U.S., Canada, France, Great Britain, Japan, Soviet Union, and West Germany--who are the major exporters of nuclear technology to reach an agreement to strengthen international safeguards last year. But, there is still a great deal lacking.

The international spread of nuclear technology raises two important policy questions about transnational aspects of waste management and safeguards. The planet we live on is becoming smaller and more interdependent politically and environmentally. The effects of illicit nuclear explosions or dispersal of plutonium may cause damage without regard to national boundaries, just as improperly managed or dumped radioactive waste of one nation could harm or damage the entire world. Therefore the risks as well as the cost must be considered when developing safeguard measures for a given fuel cycle.

The economic advantages of one fuel cycle might be offset by extra safeguard costs, and, conversely, the safeguard advantages of a particular fuel cycle might be offset by the economic disadvantages inherent within that fuel cycle. As there is no perfect solution to any social problem, there is none to the nuclear proliferation issue. Policy makers will have to decide what is

an acceptable risk and then will have to look at what type fuel cycle meets the acceptability criteria. Accurate cost estimates of these fuel cycle systems must be derived and must include detailed costs of safeguarding requirements on a national as well as a transnational basis.*

Taylor has estimated that the cost of conventional safeguarding within the U.S. would be small compared to the cost of nuclear power.¹⁵ However, if as a part of the safeguard requirements only expensive, nonproliferating reactors are allowed (either for export or domestic use), this could greatly increase these cost estimates. The extent of those cost increases needs to be determined precisely; however, that task is beyond the scope of the present program.

There are probably several nuclear fuel cycle options that would meet most risk acceptability criteria. And only a clear definition of a nuclear power proliferation policy with specific criteria could lead to the selection of an alternative fuel cycle for development. What the necessary safeguard requirements would be could then be determined. Certainly the energy demand growth, coupled with the availability of non-nuclear options, will have a great effect on what steps we should take in this regard. If an impending need for breeder reactors develops in the international marketplace, as it is expected to do near the year 2000, the most logical choice of a reactor and its fuel cycle concept might be decided by the criteria of speed rather than safety and security; i.e., by the length of time required to enter the marketplace.

Since the LMFBR reactor and purex reprocessing technology are by far the closest to commercial development, modification of this technology to a more

*Note: ERDA is currently embarking on such a program.

proliferation-safe fuel cycle might make international safeguard problems more manageable as well as more realistically obtainable. The spiking of plutonium, the co-processing of the actinides, the irradiation of fresh fuel, or the de-natured thorium cycle could be adapted to the LMFBR concept. Thus, the scrutiny of these alternatives with regard to the cost effectiveness of safeguards should be performed.

The shipment of spent reactor fuel must be handled and safeguarded in all of these options. Although the plutonium content of the ^{233}U -Th cycle spent fuel would be about one-half to one-tenth that of the other cycles, it would still need to be safeguarded. It has been estimated by the mid 1980's that nearly 40 countries will have enough chemically separable plutonium in their power reactors' spent fuel for at least a few weapons. Nearly half of these countries are presently planning to reprocess this material. If plutonium re-cycle follows, many more governments will have access to this material in the form of mixed oxide fuel inventories for their LWRs.

In order to build its own nuclear weapon, a country or terrorist group must have only several kilograms of highly enriched ^{235}U , less of plutonium, more of neptunium and other actinides. Fission-explosive materials, obtained other than by diversion or theft, require at present a considerably long lead time. However, the production of weapons-grade material from unirradiated mixed-oxide fuel elements would be considerably simpler; in fact it is so easy that it has caused worldwide concern about the risk of proliferation in a plutonium economy. Therefore, stiff safeguard measures will be required to prevent theft of plutonium if the plutonium fuel cycle, currently contemplated, is implemented. Transnational shipments of unirradiated mixed-oxide fuels would be especially vulnerable. Governments safeguarding these materials during shipments prob-

ably could adequately manage by employing an armed nuclear security force and other measures used in the U.S., as the record of safeguarding nuclear weapons indicates. The problem of accomplishing this on an international scale, possibly in unstable nations, is very difficult. Thus, the key transnational policy issue arises because, in order to adequately safeguard free-circulating plutonium dioxide fuel while in shipment between nations, the sovereignty of each nation must be sacrificed to some extent. We believe convincing the vast majority of nations to conform to this type of safeguards system is politically infeasible.

At present there are no adequate safeguards on the shipments of nuclear materials, either over or on the high seas, and these shipments may be subject to hijacking or diversion. Absence of any transnational institutions with real power along with a needed international police force makes preventing theft of strategic nuclear materials even more difficult. Certainly, confining these shipments to special cargo vessels or special aircraft which would carry nothing other than these nuclear materials would help. In fact, it may not be possible to obtain the safeguards needed for transnational shipments of plutonium without the creation of an international organization with authority and an arm to enforce it.

The likelihood of theft might be reduced if the characteristics of the fuel were such that it could be easily located and recovered. The denatured thorium cycle contains small quantities of ^{232}U , which has a decay chain that produces an easily identifiable gamma ray allowing it to be spotted by remote sensing equipment and therefore simplifying recovery. The high energy gamma source would require massive shielding to avoid detection, thus inhibiting rapid unnoticeable or untraceable movement. The spiking and coprocessing

options would also aid in the recovery in a similar fashion. If nuclear material is stolen or diverted and then eventually recovered, being able to trace where the material originated would also be a very important parameter in preventing a reoccurrence. One safeguard measure would be a doping technique that produces specific signatures similar to serial numbers. Thus a specific signature could be assigned to every fuel shipment. This traceability might greatly inhibit a prospective diverter.

Another safeguard measure would involve the construction of special vessels for nuclear material transport which house tamper-proof containers equipped with telemetry packages in constant real time contact with satellite surveillance systems. The ship and the containers would have separate instrumentation capable of reading the radiation signatures and radiation levels from the nuclear fuel. Movement of either the containers or the fuel itself would be sensed by the surveillance system. If the ship or aircraft were drawn off course by the hijackers, this too would be sensed by the satellite surveillance system. This technique, coupled with other standard alarms and barriers, might give a recovery team enough time to act in the event of hijacking.

There would be an additional vulnerability to hijacking once the containers were offloaded from the ships and transported overland to their destination, especially if this transit were through undeveloped or unstable territory. Surveillance could be interrupted, e.g., inside tunnels, and recovery might be hampered by national sovereignty. Even with these and other safeguard measures free circulating plutonium may be an unacceptable risk. Therefore to reduce the risk to a politically acceptable level, one might suggest a fuel cycle in which;

- (1) The fuel could not be made into weapons without isotope enrichment

(2) The spent fuel container could not house enough chemically separable plutonium or other actinide to produce a weapon

(3) Both the fuel and spent fuel contain signatures detectable by remote sensing equipment

(4) Each batch from each reprocessing plant would be doped with an analytically detectable signature similar to a serial number, thus allowing the diverted material to be traced to its point of origin.

One question that must be asked when trying to determine the effectiveness of safeguarding strategic nuclear materials is who are we safeguarding this material from, i.e., from other governments or from nongovernmental organizations, such as criminals and terrorists. Some of the fuel cycle options could be very effective safeguard measures from one form of proliferation but not the other. Implementation of the advanced "nonproliferation" fuel cycle concepts discussed earlier is not expected to be feasible much before the turn of the century. One has to look at near-term and long-term safeguard measures.

Probably the greatest danger from theft of nuclear material is not from industrial or small developing countries. As Epstein has said, "Unless effective steps are taken to reduce governmental proliferation, the world will inevitably be faced with a more fearsome and much less manageable threat of nongovernmental proliferation."²⁶ At present, international safeguards will not adequately protect fissionable material from terrorists, and certainly the plutonium mixed-oxide fuel would be considered as holding a bright red carrot in front of their noses.

Of course, both the nonproliferation treaty and the international safeguards systems can be criticized for their inadequacy in preventing governmental proliferation by diversion or theft. One reason is because legitimate diversion

of this material is allowed for either military propulsion purposes or research on military propulsion, both of which are not subject to international safeguards. Certainly, once this material has been diverted and stockpiled for this purpose, it would be very difficult to determine just what actual use this material was being put to. Thus, there are loopholes that exist in the present international structure of preventing either governmental or nongovernmental proliferation. For the near-term we can try to close the existing loopholes without creating new ones.

For the long term, either mixed or spiked plutonium, denatured ^{233}U , homogeneous reactors or another nonproliferating technology will have to be considered. Therefore, the ISAFE centers, as discussed in Section VI, could be an important long-term option. If plutonium is not managed by some international authority, the uncertainty as to who may or may not be diverting nuclear material could force many countries to hedge against this uncertainty by going nuclear. Their anxieties could easily be aroused by the ineffectiveness of existing safeguards. If a nation could be assured that their neighbors were not diverting fissile material from their nuclear power industry, then it might be able to rest somewhat easier and, it is hoped, avoid the nuclear weapon option, thus correspondingly reducing the risk of nongovernmental proliferation.²⁶

The most difficult step in the fuel cycle to safeguard is the one from the reprocessing plant to the fuel fabrication plant. The handling of the small, easily concealed plutonium dioxide powder and pellets must be extremely well guarded. Shipment of plutonium dioxide to the reprocessing plant would be the weakest line in the chain. Co-location of reprocessing and fuel fabri-

cation facilities would help alleviate this problem.¹⁵* While the economies of scale and the number of suitable sites do not favor nuclear power parks, they do favor the co-location of reprocessing and fuel fabrication. Certainly spiking or co-processing would help even within an ISAFE center where humans must handle plutonium in some form or another.

Therefore, reprocessing technology, e.g., co-processing of plutonium within one of these ISAFE centers, could have a significant effect on proliferation risks as well as long-term risks from actinide wastes. Using similar arguments, one can show several safeguard advantages for high-temperature separation methods over aqueous methods. These differences, however, might be small, and the selection of reprocessing might be made on other grounds, such as environmental or economic.

*Note: On the other hand, some studies (e.g., The Nuclear Energy Center's Survey-1975, NUREG-001) have concluded that co-location would not be needed in the U.S. but we do have to deal with the rest of the world and leading, by example can be useful in establishing credibility.

X. TRANSNATIONAL NUCLEAR POLICY

The primary proliferation policy questions we must address relate to the plutonium economy and the international shipment of plutonium.^{133,134,135} These questions should be addressed on an international basis since they will affect the world community and only significant international agreement is likely to be effective in establishing a world policy to deal with them. These energy plans ought to take into account the level of R&D effort that will be required and what costs are expected in the development of these long-term options. Therefore, we must look hard at the available energy options for the next century when we make decisions for the near- and mid-term. Developing any kind of breeder capability will take a very intense R&D effort. This effort must be initiated in the near-term if we are to have available that option at the end of the century. The absence of an inexhaustible energy source might do more to encourage proliferation than a nuclear power industry based on plutonium, e.g., a U.S. policy of denial on breeders, reprocessing, etc. leading to counterproductive indigenous, unsafeguarded civilian development abroad. If the basic needs of many of the industrial and developing nations are not satisfied because of an extremely short supply of energy, competition for the remaining materials could create serious tensions. Since they may consider their energy supply essential to their national survival, they might therefore turn to military solutions and develop deliverable arsenals of not only nuclear, but also chemical and biological weapons. If nuclear energy is to be employed as a remedy, vaccinating the world from energy starvation, then above all we should be assured that this remedy will not spread a comparable disease such as nuclear proliferation.

Although some of the various fuel cycle concepts presented in this paper would reduce the risk of proliferation substantially, they would not eliminate it completely. Nothing will! A country could shut down a power reactor and mine the core to produce weapons materials. Such action is unlikely because of pressure within the international community, even though at present IAEA is powerless to act and there are no stringent sanctions placed on any country violating the NPT or other safeguard agreements.¹³⁶ What was our response to India -- hardly even a protest. It is not all that difficult for industrialized nations, such as Japan, Germany, Israel, etc., to construct a facility to produce weapons-grade fissile material, but it is very expensive. The construction of such a large facility for this purpose would make it fairly evident that this government was indeed in the nuclear bomb business, but it is unclear if we could prevent this action if a country believes its national survival is at stake. Dedicated facilities could, in fact, be hidden, but the cost of hiding them would be so high that this is considered unlikely. Once a weapon was test exploded, the "cat would be out of the bag."¹³⁴

These weapons facilities could take the shape of plutonium-producing graphite or heavy water moderated reactors using natural uranium much as they did in the U.S. during World War II; or with the advent of centrifuge and possible laser enrichment technology, they might be employed to produce weapons-grade ²³⁵U. In this paper when we refer to reducing the risk of proliferation, we are referring to the (latent) risk of proliferation caused by the spread of nuclear electric generating facilities, not the dedicated facility which is not directly related to the nuclear power industry. However, the importance of satisfying world energy demands to maintain world stability should not be overlooked or underestimated in formulating a transnational nuclear energy policy.

Before any policy decision is made to commit this nation to any radical change in our long-range nuclear fission program, there are many questions, technical and political, that must be answered -- not to mention the economics! And we ought to start trying to answer some of them now. Before addressing these issues directly, it is useful to review some of our past transnational nuclear policies.

There has been a history of proposals regarding the international management of strategic nuclear materials dating back to the post-war era and the Bernard Baruch plan which called for international ownership of all strategic nuclear facilities and materials.¹³⁴ The Acheson-Lilienthal report (1946) proposed to denature plutonium to make it suitable only for peaceful purposes. This was to be accomplished by leaving the fuel in the reactor for long periods of time to produce large quantities of plutonium-240. The initial report was predicated on the belief that this high quantity of plutonium-240 would interpose the high barrier of isotopic separation between the use of plutonium for civilian and military ends.¹³⁴ This report encompassed much of the international control mechanisms put forth in the Bernard Baruch plan which were at the time thought to give a 2-3 year warning of any country attempting to develop plutonium weapons. The Baruch plan would have given sovereign states control only of the denatured, so-called "safe," plutonium. They would have obtained all of their fissile material in denatured form from spent fuel in plants owned by an international authority. This authority was to have a monopoly over all activities handling weapons-grade materials. This plan rejected the present concept of international inspections rather than international ownership as being inadequate. It could never work because plutonium-240 is fissile and could by itself, be made critical under certain conditions.¹³⁶ Therefore, it is logi-

cal to assume isotope separation would not be required to construct weapons from any mixture of plutonium-239 and 244.^{138,139} However, the plan could be revived using the denatured thorium cycle. Here natural uranium is indeed capable of rendering uranium-233 safe or nonfissile with the high barrier of isotope separation.

The Barnard Baruch plan at that time was rejected by the Soviet Union, and it is certainly possible that they would reject a similar plan put forth by the U.S. today. It is considered extremely important to the international acceptability of any nonproliferation fuel cycle concept that it be formulated in an international form or amphitheatre where the French, the Soviets, and the other nuclear suppliers, as well as representatives from the third world, would all have a part. National chauvinism, instinctive in most humanity, as well as widespread anti-Americanism sentiment make it highly unlikely that we could impose such a fuel cycle concept without true international authorship and sponsorship.

It is clear that nuclear energy has an important role to play in the future world energy mix. However, the positive contributions as well as the projections for market penetration have been "puffed up" out of proportion. Recent estimates by several independent organizations indicate that, without extensive conversion of uranium-238 into plutonium and the separation of plutonium from spent fuel, the U.S. would indeed have enough natural uranium and coal at reasonable prices to last us well into the 21st century.^{134,135} By that time we should be able to make an intelligent transition to the use of one of our possible inexhaustible resources, such as a safe and economic breeder or a safe form of fusion or some form of solar electric energy. There are contrary claims coming from the nuclear industry regarding the use of plutonium; however, this

industry has suffered chronically from premature commitments based on exaggerated energy demand. In 1970 predictions as high as 300 GW electric of nuclear capacity were projected for the U.S. by 1985.¹³⁴ Today, on the basis of actual construction or orders, FEA expects 145 GW electric or less.¹³⁴ This, coupled with the expected reduced growth rate in energy demand out to the year 2000, would indicate that a more than adequate supply of uranium exists for our needs.

Other nations, such as France, are not as fortunate as the U.S. They must continue to develop their lifeblood in energy, and they must do it as soon as possible. Since we no longer hold a monopoly on exportable nuclear technology, only an international agreement to modify the plutonium fuel cycle for export would be effective in reducing proliferation. Maybe we or, better yet, an international organization such as IAEA should begin to examine the possibilities of modifying the Super-Phenix. This might be accomplished by an international development effort.

A politically feasible way to assure that nonproliferating reactor fuels are exported is to get all the nuclear-supplying nations to agree on an exportable alternative before its commercial implementation. This may be accomplished by putting the exportable nuclear industry under international control. Even though there are other paths to proliferation which cannot be regulated, the acceptability of nuclear power to the public and therefore its rapid expansion in the last decade of this century may well hinge on the development of a "nonproliferating" breeder concept.

The circulation of plutonium in the international marketplace should be avoided until such time as international institutions exist which are capable of safeguarding strategic nuclear materials. It is unlikely that all nations would agree to recognize an international police force or that all nations

would even sign an agreement to return hijacked strategic nuclear materials. Therefore, the political realities call for the nuclear-supplying nations to examine the possibilities of implementing a fuel cycle which might ameliorate this safeguard problem.* Regional International Safeguarded Atomic Fuel Exchange (ISAFE) centers could play a role in this endeavor. We recommend that several ISAFE centers be formed now. These would initially function as spent fuel depots and possible enrichment facilities for LWR fuel. When reprocessing is required to produce plutonium to start up the breeder economy, the facilities would be located within ISAFE centers and would service all exported reactors and possibly other reactors.

In addition, the U.S. has the potential to export coal until adequate international safeguard measures can be instituted, or until such time as a non-proliferating export fuel cycle can be implemented. Many third world countries, including Ireland, have a large need for small electric generating units which can be made more cost effective with coal than nuclear. The U.S. is presently the only stable nation with a large export capability that third world nations could rely on for long-term fuel commitments.

It has been argued that over-estimates are safer than under-estimates; however, over-inflated estimates of energy demand could be harmful in developing a transnational nuclear policy. Exaggerated estimates in the past have severely damaged both national policy and the profitability of the nuclear industry. It appears that many U.S. reactor manufacturers have suffered large cumulative losses because of reduced reactor demand.

It is this collapse of expectations in the domestic market that has unfortunately led to aggressive selling campaigns abroad, including the underdeveloped

*Note: It is significant to note that President Carter has done just that.

world where nuclear power is only marginally economic. This is because of the high capital intensity and the economies of scale mentioned earlier. It has been these sales abroad, not only by the U.S. but by the French and Germans, which have led us to suspect that these reactors were bought only partially for the power they would produce. Digging into our government stockpiles of uranium, worth \$8 billion at current prices, and developing our coal resources for export would give us some time to add enrichment capacity in order to meet world demands.¹³⁵ If we direct our policies toward an open fuel cycle, it is necessary to start planning for relatively long-term storage for spent fuel elements, either in surface storage sites or in some sort of retrievable facility. Probably both would be required.

ERDA's most recent plans call for a decision on the commercial breeder by 1986.¹³⁶ If this decision is positive, the first commercial breeder is expected to be in operation about ten years later. By conservation and proper utilization of our resources, we should be able to postpone commercialization of a breeder until the next century. This may give us time to form an organization capable of implementing adequate safeguard measures and managing spent fuel. And, it is hoped, the world will then have time to develop a more proliferation-safe fuel cycle. Certainly, sacrifice on our part as well as the rest of the world will be necessary.

The International Atomic Energy Agency is chartered with the promotion of nuclear energy for peaceful purposes. This charter is in conflict with the regulatory and environmental protection roles that need to be implemented. This system is in some way analogous to the United States Atomic Energy Commission before Congress saw fit to remedy the matter by creating the U.S. Environmental Protective Agency and the Nuclear Regulatory Commission. The U.S. presently

has three agencies involved in nuclear waste management issues. However, we recommend that only a second agency be created on the international scale. This would be an international environmental protective agency. Its function would be, among other things, to deal with the transnational aspects of radioactive emissions from ISAFE centers as well as various portions of the nuclear fuel cycle. For example, radon-222, computed to be the single largest killer in the nuclear fuel cycle at present, is produced in larger quantities from phosphate mining than from uranium mining and milling. Radon-222 is distributed worldwide and is of transnational concern, just as much as the radioactive wastes from reprocessing plants would be. This agency could deal with all of the transnational environmental issues including depletion of the ozone layer, long-term effects from ocean dumping, oil spills, etc.

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Table 2. Summary of Spent Fuel Reprocessing Techniques Proposed to Date

No.	Classification Category	Name of Method	Reference	Input Fuels	Principle of Separation	First Major Ref. Found by Authors	Stage of Development	Ability to Extract Pu	Affiliation of Work	Remarks
1. Aqueous Processes										
1.1. Solvent Extraction										
1		BUTEX	49	UO ₂ oxide fuels	Solvent extraction	50, Apr. 1952	Lab studies	Yes	Union Carbide Nuclear Co.	Process for recovery and decontamination of Pu from irradiated U. Recovery of uranium is accomplished, but decontamination is not adequate to permit direct handling.
2		Chelation	51	UO ₂	Solvent extraction	51, Apr. 1949	Lab studies	Yes	UC Berkely Rad. Lab.	Pu recovery and decontamination from dissolver solutions and redox wastes.
3		HALEX	52	UO _x oxide fuels	Solvent extraction	53, Jan. 1956	Lab studies	Yes	Argonne	Identical to purex, except that CCl ₄ used as diluent for TBP.
4		Purex	54,55	Metallic, UO ₂	Solvent extraction	56, Sept. 1950	Commercial	Yes	Oak Ridge Natl. Lab.	Extensively employed commercially; bulk of R&D effort spent on this. Good separation of U & Pu. Good decontamination.
5		Redox	57	Metallic, UO ₂	Solvent extraction	58, Nov. 1949	Pilot plant	Yes	Oak Ridge Natl. Lab.	Superseded by purex.
6		Thorex	59	Th, ThO ₂	Solvent extraction	60, Jan. 1946	Pilot plant	—	Oak Ridge Natl. Lab.	Solvent extraction process for Th fuels; high decontamination factor achieved.
1.2 Ion Exchange										
7		Ion Exchange	61,62,63	Metallic U, UO ₂ , Pu, PuO ₂	Adsorption-Desorption	64, 1947	Commercial Possible	Yes	Oak Ridge Natl. Lab	Alternate process in 1942-44 at Hanford for Pu production. Used primarily as tail-end to purex, etc. for further purification and concentration of 253U, 233U, Th, Pu, etc.
1.3 Photo-Chemistry										
8		Photochemical Induced Separation	65,66	Any type that fits purex	Selective oxidation or reduction	67, March 1975	Conceptual	Yes	Brookhaven	Still in conceptual stage, viewed as modification to purex tail end to permit almost complete separation of actinides.
1.4 Electrolysis										
9		Flurex	68,69	Metallic, oxides	Electrolytic (Aqueous)	70, 1955	Lab studies	Possible, with great modification	Hanford	Designed for U recovery from NH ₄ UF ₅ .
1.5 Precipitation										
10		Bismuth Phosphate Precipitation	71	Metallic U, Pu	Precipitation	72, 1942	Military purpose plant	Yes	Hanford	Basically for separation of Pu from U & fission products. This has been expanded for recovery of other fission products individually.
2. Pyroprocesses										
2.1 Pyrophysical										
2.2 Fractional Distillation										
11		AEROX	73	UO ₂	Volatility	73, Feb. 1963	Lab studies	Not possible	Atomics International	Low decontamination, rapid recycle; remote refabrication needed, suitable only for UO ₂ fuels.
12		DEBOER	74	Metallic U U alloy	Volatility	74, May 1957	Lab studies	No	Knolls Atomic	Decontamination from all long-lived fission products except Zr by formation of UI ₄ . By selective condensation of UI ₄ , U & Zr can be separated slowly.
13		Fluoride Volatility	75,76	UO ₂ PuO ₂ , Alloys Metallic U, UC	Volatility	77, 1956	Pilot plant	PuF ₆ extraction possible. High purity.	Argonne	Along with chloride and bromide volatility, this method has been the subject of much research. More activity in France & USSR than in USA in recent years.
14		Molten Salt-Fluoride Volatility	78	Metallic U or U-Alloy	Volatility and sorbtion-desorbtion	78, Apr. 1971	Lab studies	Difficult	Oak Ridge Natl. Lab.	For recovering decontaminated U from U-Al fuels with Al cladding.
15		Nitrofluor	79	Alloys, UO ₂	Volatility	80, 1964	Lab studies	PuF ₆ extracted together with some F.P. Pu distillation possible.	Brookhaven	Non-aqueous fluoride volatility U, Pu separation and purification; kilogram amounts of fuel have been tested and studied.
16		Carbox	81	UC	Separation only of f.p. volatile at high temp.	82, Feb. 1965	Lab studies	Pu cannot be removed	Atomics International	Specially developed for UC fuels. Low decontamination recycling. Only volatile fission products removed by separation.

Table 2 (Cont'd)

No.	Classification Category	Name of Method	Reference	Input Fuels	Principle of Separation	First Major Ref. Found by Authors	Stage of Development	Ability to Extract Pu	Affiliation of Work	Remarks
	2.1.2 Fractional Crystallization									
17		Hermex	61,83	U, U Alloy, Th	Fractional Xtallization with solvent	84, Aug. 1957	Lab studies	No	Oak Ridge Natl. Lab.	Suitable for metallic uranium, low decontamination.
18		Hydride Separation	48	Metallic U	Sieving	85, March 1957	Lab studies	Pu-rich phase can be separated. Very low purity.	Ames, Oak Ridge	Poor separation, only lab studies. Kinetic rates expected to be very slow since processing will probably be in the solid state.
19		Pyrozine	86	Metallic U	Fractional Xtallization	87, June 1957	Lab studies	No	Argonne	Suitable for metallic fuels; Pu not separated. For U recycling only.
20		Zone Melting	88	Metallic U	Fractional Xtallization	89, 1952	Lab studies	Not possible	Argonne	Very slow; applicable only for the production of small quantities of high-purity metals for laboratory purposes.
	2.1.3 Liquid-Liquid Partitioning									
21		An extraction of Pu & fission products	90	Metallic U	Liquid-liquid partitioning	91, 1955	Lab studies	Low extraction ability for Pu contamination by other fission products.	Ames, Iowa	Low decontamination. Separation of Pu improves with addition of gold. Economic feasibility not expected for large scale separation.
22		Buffer Method	92	Metallic U U Alloys	Liquid-liquid partitioning	93, 1955	Lab studies	Doubtful	Brookhaven	Designed to separate U and other fission products. Possible to modify to remove Pu from fission products.
23		Fused Salt-Liquid Metal	94	²³³ U-Th	Liquid-liquid partitioning	95, 1961	Lab studies	—	Ames, Iowa	For reprocessing of ²³³ U-Th fuels, separating ²³³ U from Th.
24		Mg extraction of Pu from molten U	46,45	Metallic U or U alloy	Liquid-liquid partitioning	96, Aug. 1955	Lab studies	Process designed for Pu extraction. Purity of product low.	Atomics International	Suitable for metallic fuels, low decontamination for recycling. Only lab study reported.
25		Salt Transport Process	97	UO ₂ , PuO ₂	Liquid-liquid partitionings	98, Aug. 1969	Lab studies	Pu separable	Argonne	High decontamination developed for LMFBF but could be applied possibly to other fuel types with suitable modification.
	2.1.4 Liquid-Solid Extraction									
26		DAP	99	Metallic U, Pu U, Pu Alloys	Liquid-solid separation	100, Aug. 1960	Lab studies	Possible, but low purity	Dow Chemical Co.	Dow aluminum pyrometallurgical process. U is dissolved in molten Al and precipitated as UAl ₃ . Separate step required for Pu recovery. Applicability to oxide and carbide fuels doubtful.
27		Parlee-Anderson	44	Metallic, NO ₂ , UC	Liquid-solid separation	44, 1969	Lab studies	Possible if U-Pu separation step added	Stanford	Lab studies only, suitable for in-situ reprocessing of reactor fuels. Separate U-Pu from fission products.
	2.2 Pyrochemical									
	2.2.1 Electrochemical									
28		Electrorefining	101,102	Metallic U or mixed salt	Electrochemical (Pyro)	47, 1969	Lab studies	PuCl ₃ removed as vapor	Knolls Atomic Power Laboratory	Only lab studies, though work not followed up in the US. Current research in USSR quite extensive. Not suitable for ceramic fuels and carbon coated fuels.
29		Molten Salt Electrolysis	82,103,104	UC	Electrolysis	105, March 1969	Lab studies	Pu stays in salt. Possibility of Pu extraction by adding another cell.	Atomic International	Application of molten salt to reprocessing of UC fuels.
30		Salt Cycle Process	106	UO ₂ , UO ₂ -PuO ₂	Electrolysis	107, Sept. 1961	Pilot plant	PuO ₂ crystals deposited, recovery = 99.5%	Battelle Memorial	Research began in 1956; lab and pilot plant studies have been done.
	2.2.2 Cyclic Oxidation Reduction									
31		Nitride-Carbide Cycle	108	UC	Oxidation-reduction	108, Aug. 1969	Lab studies	No	Atomics International	For processing of UC fuels only. Low decontamination pyrochemical method. High Pu losses.
	2.2.3 Selective Oxidation									
32		Melt Refining	109	Metallic U	Volatility and selective oxidation	110, 1959	Pilot plant	Pu not removed	Argonne Natl. Lab.	No U-Pu separation, decontamination by volatilization or oxidative-slugging. Experimental processing done at EBR-II for few years. Remote fabrication if fuel rods carried out successfully.