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LONG-TERM WASTE MANAGEMENT METHODS

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ABSTRACT

Battelle-Northwest has been conducting extensive evaluations of permanent or ultimate methods for storage and/or disposal of high-level radioactive wastes. Included are use of extraterrestrial transport, transmutation, cavities deep in the earth, the sea bed including use of tectonic subsidence areas, and various combinations of geological and engineered structures.

A large number of factors can influence the selection of new methods for the long-term management of radioactive wastes. Because of the sketchy knowledge of these factors at this time, the initial studies have focused on feasibility, timing, and safety.

This paper reports the methodology being used to study these subjects. Specific information is presented on the potential usefulness of transmutation, extraterrestrial disposal and polar ice for management of nuclear wastes.

The requirements of each option for modification in methods of reprocessing or subsequent waste treatment are discussed.

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Introduction

A study of alternative methods of storing or disposing of radioactive wastes from fuel reprocessing operations is being performed by Battelle, Pacific Northwest Laboratories, for the USAEC's Division of Waste Management and Transportation. The general objective of the study is to provide information pertinent to assessing the feasibility of various permanent approaches to the problem. The information includes identification of various candidate methods and assessment of the technical status, development requirements, development costs, development schedules and factors influencing public acceptability and overall feasibility.

The concepts under consideration can be categorized in two general classes: those that would ~~remove~~ *from the biosphere* or eliminate the radionuclides, and those that store or dispose of the nuclides on earth. Figure 1 shows the concepts being studied.

*where does
transmutation fit?
How about 1) store on earth
2) ~~store~~ *store* of earth
3) ~~Rid~~ *Rid* completely*

The current decision or logic diagram for the long-term waste management studies is shown in Figure 2. Note that the alternatives under study fall in two generically different categories: those requiring an additional partitioning ~~or segregation~~ step and those that do not.

Even though comprehensive conclusions cannot be reported at this time, we are prepared to discuss the methodology employed in the studies and some preliminary indications from specific studies. ~~A first example will be.~~

Extraterrestrial Disposal

The basic feasibility of nuclear waste disposal off ^{the} earth involves identification of a system that provides adequate safety, ~~a favorable energy balance,~~ an effective contribution to reduction of long-term waste inventory, and reasonable cost.

The subject of energy balance can be quickly resolved. The chemical energy equivalent of the 10,000 MWD_e produced by one ton of nuclear fuel would lift 1,125,000 kg or about 10,000 times the total weight of fission products and actinides in the waste from the tonne of fuel. More efficient vehicles such as the space shuttle and tugs should reduce energy requirements *further*.

Some of the parameters of the systems that must be investigated *concerning safety, cost, and waste inventory reduction* include the following:

1. Composition and form of the waste.
2. Method of waste encapsulation.
3. The shielding and heat removal requirements.
4. The vehicle or combination of vehicles.
5. The mission profile and destination.
6. Reentry or abort survival and recovery.

→ In cooperation with the Lewis Research Center of the National Aeronautics and Space Administration, we are examining these subjects in detail.

Both safety and payloads are established by the specific design of waste encapsulation. Early in the study, it was determined that payload ^{cost considerations} (i.e., the waste, criticality control materials, shields, and structural materials in the waste package) ~~cost considerations resulted in a prohibitive penalty~~ ^{ed} for materials requiring substantial shielding for gamma emissions or Bremstrahlung produced by energetic betas. In such systems the waste-to-payload ratio may be below one percent ^{and are therefore clearly unfavorable}. Studies have ~~thereby~~ been restricted to payloads of a partitioned fraction of the waste consisting of *all the* transuranics contaminated by 0.1 to 1 percent of the fission products.

A The shielding requirements for these levels of fission product contamination do not significantly increase shield weights above that necessitated by neutron attenuation requirements. Waste-to-payload ratios increase to five to eight percent.

Safety ~~in the~~ use of isotopic generators for space missions has been extensively investigated by the United States Atomic

Energy Commission. It is reasonable to expect that the design principles and fuel form types developed for such applications may be adapted to waste disposal.

For example, Figure 3 shows a design for a Pu-238 heat generator for space use. In the design by General Electric Company, Valley Forge, Pennsylvania, multiple spheres of oxide fuel are each encapsulated in a carbon impact shell enclosed in a graphite ablation container and clad in iridium for containment. The waste-to-payload ratio in this concept approximates 50 percent.

An approximation of the gross-to-net weight ratio for a specific concept to handle wastes was obtained by the preliminary designs illustrated in Figure 4. In the capsule design shown, the actinide oxide particles are first spherodized, then provided with a tungsten shell which incorporates a void volume to accommodate helium buildup. The tungsten is coated with molybdisulfide for oxidation resistance. These microcontainers are assembled in a matrix of lithium hydroxide ^(for neutron absorption) and a metal ~~for neutron absorption and~~ improved thermal conductivity, respectively. ^{(for} Boron is indicated as a neutron poison but may not be required. The consolidated particle matrix is encapsulated in lithium hydride for additional neutron shielding and impact resistance. (Additional thermal calculations particularly during aborted reentry may change the specific design, but it is believed the payload parameters will remain at values approximated by this design.)

Very preliminary calculations indicate that exposure from this type of system will be in the range of $1R/hr$ at 1 meter and will provide sufficient integrity to withstand most of the failure modes contemplated.

The transuranic content of this design is about 7.7 percent for a gross-to-net ratio of about 13.

Many factors have to be included in an assessment of mission profile and destination. These include the location of abort ^{and recovery} impact areas ~~and recovery~~, international agreements on "potentially harmful interference with activities in the peaceful exploration and use of outer space" and fundamental technical questions about long-term stability (and chance of earth intercept) of either high earth or solar orbits.

Payload capabilities with parameters of transport vehicle and destination are shown in Figure 5. Mission energy requirements are presented as impulse velocity change (ΔV) beyond a reference circular earth orbit of 185.2 km (100 nautical miles) ^{and a velocity of} ~~The~~ 7,793 meters/sec ~~velocity for a 185.2 km circular orbit is the~~ ^{relative} actual orbital speed referred to earth-centered nonrotating coordinates. All launches are considered to occur at the Kennedy Space Center in a southeasterly direction.

For the sake of preliminary economic evaluation, it was assumed ~~The mission destination would be either~~ that ~~energy requirements and payloads were equivalent to either~~ earth escape or high earth orbit, i.e., a ΔV of 3,300 meters/sec. (Note ~~here~~ that this would not impart the sun and the difficulty of such a mission from ΔV required.) ~~Energy requirements and payloads are nearly equivalent for these destinations~~

With this criteria, a comparison of payload capabilities costs are shown in Table 1. Minimum costs would appear to be in the range of \$1,500 per kilogram for a shuttle-tug combination. This cost is the basis for ^{the Figure 6, which} ~~the following cost calculations.~~

not ^{This graph} Figure 6 relates the weight of waste per tonne of fuel and percentage of waste in the payload to a unit cost per kilogram of original fissile and fertile material (fuel) and the cost of electricity. At 5% and 50% waste-to-package ratios the transport ~~costs~~ for a kilogram of waste ~~costs~~ would be in the range of \$30,000 to \$3,000.

Disposal of the transuranics from a LWR-Pu cycle would involve about 6 kg of waste per metric tonne. The previously described package design had a 7.7% waste-to-payload ratio after provision for encapsulation, ablation shield and safety devices. It would result in transport costs of \$116 per kg of fuel or 0.50 mills per ^{PW-hr} ~~kwh~~ for 6 kilograms of transuranics. Designs or process modifications (e.g. complete elimination of the gamma shield) which would increase the waste-to-payload ratio to 25% ^{PW-hr} would reduce transport costs to \$35 per kg of fuel or 0.15 mills per ~~kwh~~.

Appropriate perspective on these costs requires consideration of the expense in managing the other wastes not transported to space, the cost of alternative waste management or power production schemes and the significant and extraordinary nontechnical factors involved in waste disposal by this method.

These considerations are outside the possible scope of this paper, but will be explored in depth, ~~in the reference study.~~

In summary, ~~it would appear that disposal or storage (in parking orbits) of selected fractions of nuclear wastes is~~
~~feasible although costly.~~ ^{of pure disposal} The costs, although high, may be reasonable compared to alternatives, or in the context of removing very long-lived radionuclides from man's biosphere.

Transmutation

Transmutation as a waste management scheme is the concept of inducing nuclear reactions in the radioactive isotopes to produce a stable isotope or one that is less hazardous. There are several important criteria to be applied in the evaluation of the technical feasibility, however. These are:

- ① Overall energy balance. If the source of power for the transmutation scheme is based on a fission reactor economy ^{waste generated in developing countries must be less} then the power used for transmutation must produce significantly less waste than that destroyed.
- ② The specific transmutation rate must be significantly faster than the natural radioactive decay rate.
- ③ The transmutation process itself must produce significantly less waste than that destroyed.
- ④ The transmutation process must be able to transmute a quantity of waste which would be a significant fraction of the waste inventory.

Using the above criteria, early studies of transmutation concepts clearly indicated that no known ^{and available} device could beneficially transmute the gross high level waste from the reprocessing of light water reactor fuels. ^{us} Then, the following discussion will be concerned with feasibility of transmutation of selected fractions of high level wastes.

On an energy balance basis, transmutation of either fission products or actinides by nuclear reactor ^{low} induced ^{directly} by accelerator sources of electrons, protons, or photons all appear to fall far short of achieving a positive energy balance for either fission products or actinides.

Similar energy-^{to} neutron balances indicate that an LWR operating under current conditions cannot destroy its own unseparated fission products. One tonne of LWR fuel exposed to 33,000 MWD/T undergoes 9×10^{25} fissions. Under the best of conditions an excess of

perhaps one neutron per fission ~~on~~ ^{or} 9×10^{25} neutrons might be made available ~~for~~ ^{for} transmutation.

The total fission products, exclusive of actinides, are about 300 gram moles per tonne. Transmutation at one neutron per atom would require 2×10^{26} neutrons or 2 fold greater than the excess available.

Of equal concern is the rate of transmutation. The rate of transmutation required for a practical device may be determined by rather broad considerations. The U.S. National Academy of Sciences, in reviewing the Savannah River Bedrock Storage Concept, ⁽¹⁾ implied that storage times of 1000 to 1200 years would probably reduce cesium-137 and strontium-90 concentrations to tolerable values. The 1000 years is about 30 half-lives. If the same residual quantities of strontium and cesium were to be achieved in say 10 years, the transmutation rates would have to be 100-fold greater than the natural decay rates or an effective half-life of 0.3 years.

For the fission product nuclei with small transmutation cross-sections such as ^{90}Sr and ^{137}Cs , the transmutation rate in commercial thermal power reactors is much lower than their natural radioactive decay rate. ~~The~~ ^{the} transmutation ^{rates} of this type of ~~nuclei~~ ^{nuclei} in fast breeder reactors can be approximately equal to ~~their~~ ^{the nuclei's} radioactive decay rates. ~~The~~ ^{however} transmutation of any fission product nucleus ~~however~~ ^{is} in a fission reactor of any type means that it was obtained at the expense of approximately one fission event. A fission event results in a fraction, f , of undesirable radioactive fission product nuclei. This fraction, f , has been estimated by Keilholtz and Battle ⁽²⁾ of ORNL to be 0.23 for a LMFBR. Thus, if all the other criteria could be satisfied, the inventory of such fission product nuclei could only be reduced by a factor of about four at best by transmutation in a fission reactor.

(1) An Evaluation of the Concept of Storing Radioactive Waste in Bedrock Below the Savannah River Plant Site, ISBN0-309-02035-2, Spring 1972.

(2) G. W. Keilholtz and G. C. Battle, Jr., ORNL-NSIC-37 (March 1969).

The Canadian Intense Neutron Generator, ING, is the most intense accelerator source of thermal neutrons that has been seriously proposed. An expanded ING with an order of magnitude larger neutron source was proposed as a source of neutrons for fission product transmutation by Gregory and Steinberg of ~~BNW~~ ^{Brookhaven Natl. Lab. (3)}. Analysis of the capabilities of the ING, however, indicates that it is questionable that a breakeven energy balance could be obtained as shown by Davidenko. ⁽⁴⁾ In addition, the production of undesirable radioactive elements in the ING target could be appreciable although definitive information on this aspect seems not ^{to} be available. Estimates, ~~however~~, range from minimum values of f of about 0.1 to values ~~much larger than this~~.

thus The likelihood of transmutation processes for low cross section fission product nuclei ~~at the present state of evaluation~~ seems to depend upon the achievement of the ~~CTR~~ ^{Controlled Thermonuclear Reactor (CTR)}.

However ^{likelihood} The possibility of transmutation process for the actinides, ~~however~~, appears to be much different. The effective radioactive decay half-lives for almost all of the actinide isotopes are the order of 10,000 years when the radioactive daughters are considered (as opposed to about 30 years for ^{strontium cesium} ~~Sr. or Cs.~~). In addition, the transmutation cross sections are, generally, an order of magnitude or more larger than the low cross section fission product nuclei except for 14 MeV neutron processes.

A more fundamental difference is that transmutation processes for the actinides ultimately result in fission. Thus, the actinide isotope is ultimately converted to about 1/4 of troublesome fission product nuclei and fission energy. The transmutation of actinides thus depends on the assumption that the ~~f.p.~~ ^{fission products} are less hazardous than the actinides. In this respect an atom of curium, for example,

(3) M. V. Gregory and M. Steinberg, BNL-11915 (1967).

(4) V. A. Davidenko, Sov. At. Energy, 29, No. 3, 866 (Sept. 1970).

represents a potential for energy and for ultimate fission product which differs only in detail from an atom of ^{239}Pu . Thus, in a fission reactor economy, the transmutation of actinides in power reactors appears quite favorable from the point of view of technical feasibility. Preliminary examinations of the neutronic behavior of ^{Plutonium} ^{239}Pu and higher actinides of americum and curium indicates:

- The effect of recycle of higher actinides has a small impact on the neutron economy of a commercial thermal power reactor.
- The effective half-lives of the actinides would be reduced to 10^{10} years under repeated recycle.
- The available fission energy of the actinides is ^{used} ~~utilized~~ as an energy resource.

The proposed continued recycle of plutonium and actinides in power reactors would lead to an inventory of these isotopes which would in time become ^a constant ^{amount either} in-reactor and in-reprocessing directly proportional to the fission energy economy.

Calculations by Claiborne⁽⁵⁾ studying the effect of actinide recycling have indicated the necessity of the separations process maintaining low losses. In the reference work, 15 recycles of the actinides were made through a PWR reactor with losses of either 0.1% or 0.5% at each pass. The cumulative loss of actinides to waste was thus in the range of 0.5 to 2.5%. Accordingly, the long-term (10^6 year) hazard of the waste was ^{only} reduced by a factor of 200 (i.e., $1/0.005$) or 40 (i.e., $1/0.025$).

An alternate to a very highly efficient separations scheme ^{might} ~~would~~ be a Phoenix or poison fuel which would remain in the reactor until end of life.

(5) H. C. Claiborne, High Level Radioactive Waste Disposal by Transmutation, ANS Transactions, Volume 15, No. 1.

The use of transplutonium nuclides as separate reactor fuel elements might constitute a burnable poison with the interesting property that the reactivity of the elements would change from negative to positive near the end of fuel life.

All in all, it would appear that the significant advantages evident in an actinide recycle scheme warrant further thought and work. Similarly the advent of commercial controlled thermonuclear reactors and their usefulness in transmuting fission reactor wastes should be anticipated.

Polar Ice

The polar regions, comprised of the Arctic ice cap and the continent of Antarctica, together form about one-twelfth of the earth's surface. The remoteness of these areas, coupled with the major heat sink represented by climatic conditions and the major ice deposits are potential attributes for a repository for nuclear wastes.

In contrast to transmutation with its problem of basic technical feasibility, or extraterrestrial storage/disposal ^{with its} problems of cost, serious consideration of disposal of nuclear wastes in polar ice must conclusively demonstrate removal from man's biosphere for periods of time ranging from 500,000 to 1,000,000 years for unpartitioned nuclear waste. The evidence required for such proof includes prediction of future glacial effects---including anticipated "ice ages"---and detailed knowledge of the hydrology and meteorology pertinent to a specific disposal site.

~~Thus, in contrast to the methodology employed to evaluate transmutation and extraterrestrial disposal,~~ studies of disposal in polar ice have to be concerned at the outset with the geology, hydrology, and glaciology of the region, waste placement logistics and legal-political considerations.

For our study, we have confined most of our attention to Antarctica. Unlike the Arctic, ^{which is} a frozen sea bordered by land masses, Antarctica is a continental land mass surrounded by ocean. An ice cap, with an average thickness of two miles, covers the continent. However, some very high mountains in the Queen Maud range of the Transantarctica Mountains jut upward through the mantle of ice and snow. The size of the Antarctic ice sheet is

To illustrate this point, it should be noted that enormous. ^{and near the "hole of inaccessibility"} If all the ice in this sheet could be transformed to water, the world sea level would raise some 76 meters.

Zeller and Turner⁽⁴⁾ have suggested that the best location for a repository would be in a substantial thickness of ice and near the tops of ridges in the underlying bedrock. The specific area suggested is in the Polar Subglacial Basin near 80° longitude and 80° latitude ^{and near the "hole of inaccessibility"} (Figure 7) This is an area with a land elevation of over 10,000 feet above sea level and ice thicknesses of ~~3000~~ to 8000 feet. The 80°/80° site is 1250 kilometers from the east coast and Indian Ocean.

Much work needs to be done on the complex geology of Antarctica. There is ample proof that glacial conditions have been the exception and not the rule in Antarctica. Although the present glaciation is severe, it appears to have passed its maximum; some evidence indicates that in the past the ice sheet was some hundreds of feet above its present level. Ice as old as 500,000 years has been found.

~~Movement of typical glaciers~~ ^{move a} is at ^{about} the rate of 10 meters per year. If the wastes moved at the ^{average} rate, they could reach the ocean in 100,000 to 200,000 years ^{from the site noted}. This is probably too short a time to allow adequate decay of the transuranics.

It is also recognized that the presence of fluid water between the bed rock and ice (or flowing at the earth's surface) could be more important than glaciation rates. ~~Undesirable movement~~ ^{movement} of such water could result in rapid transport of radionuclides.

Of similar concern are projections that indicate the next ice age may arrive in 10,000 to 50,000 years. According to the

(4) Radioactive Waste Disposal, E. J. Zeller and D. F. Saunders, April 1972.

theories of A. D. Wilson^() the ice deposits produced by such an occurrence might approach instability. If the Antarctica ice sheet were to surge, the ice (and possibly nuclear wastes) would be transferred to the sea.

The subject of hydrology, ice age effects, and the actual glaciation rate, to be expected in subglacial basins is being examined in concert with the World Data Center for Glaciology, USGS, Tacoma, Washington. Until further data is available on this subject, our studies are assuming that polar ice could only be used as a repository for nuclear wastes that have radiological hazards for periods of 50,000 to 100,000 years. In practical terms this would limit waste disposal to ~~the~~ fission products and require alternate disposal of the transuranics.

Two disposal concepts are being explored. The first envisions simply depositing the canister in shallow holes drilled in the surface and allowing them to melt through the ice until they reach the earth below. For the sizes and heat generation rates under consideration, the canisters would sink at rates of a meter per day. Total sinking time would be in the range of 2^{to}₇ years.

The total accumulated heat load in the year 2000 if all fission products from nuclear power were so deposited would be about 575 MW. This would melt 120,000 tonnes of ice per day. This energy is considered very small in contrast to that exchanged to the Antarctic by natural phenomena (i.e., solar energy, precipitation and winds), but the actual effects are being further evaluated.

() The Climatic Effects of Large-scale Surges of Ice Sheets, A. T. Wilson, Canadian Journal of Earth Sciences, Vol. 6, 1969.

Page 1

ADVANCED WASTE MANAGEMENT METHODS PROGRAM

CONCEPTS UNDER STUDY

ELIMINATION

TRANSMUTATION

ACCELERATOR

FISSION REACTOR

CONTROLLED THERMONUCLEAR REACTOR

EXTRATERRESTRIAL

SOLAR IMPACT

ORBITING

SOLAR ESCAPE TO DEEP SPACE

ACCELERATED PARTICLE

DISPOSAL/STORAGE IN EARTH

GEOLOGIC FORMATIONS

MINED VOID

NUCLEAR CAVITY

DEEP HOLE (10 MI)

DRILLED HOLE MATRIX

ENGINEERED GEOLOGIC STORAGE

SEA BED

SEDIMENTATION BURIAL

DEEP TRENCH (TECTONIC SUBSIDENCE)

CONSOLIDATED SEDIMENTS

POLAR ICE

ICE BURIAL

ICE SURFACE FACILITY

PNL ADVANCED WASTE MANAGEMENT METHODS PROGRAM SIMPLIFIED PROGRAM LOGIC DIAGRAM

Fig 2

TRANSMUTATION

ISOTOPIC FUEL FORM

~~HELL-PAN DESIGN~~

Figure 3

GRAPHITE IMPACT
SHELL -.460 THK.

FUEL - 1.465 DIA.
82.5% DENSITY

WOUND T-50 IMPREG.
CRUSH-UP -.74 THK.

POCO ABLATOR
.400 THK.

IRIDIUM SPHERE
.020 THK

PYROCARB 406
RETAINER -.200 THK.

GRAPHITE
CLOTH
COMPLIANCE
PADS

PRD & VENT TUBE

16.880 OVERALL

2.554
FUEL SPH.
DIA.

2.172
(TYP.)

7.190 O.D.
CLAD @68°F

7.356 O.D.
EMISSIONITY
SLEEVE
@68°F

IRIDIUM CLAD
.010 THK

.050 THK. EMISSIONITY
SLEEVE - T-50 GRAPHITE

TIE BOLTS &
GRAPHITE SPHERE LOCKS

REF. HELI-PAK DESIGN - 4 x 6 MATRIX

1/2" Dimension unit?

Figure 3

~~PRELIMINARY DESIGN OF NUCLEAR WASTE~~ ^{PACKAGE} ~~ACTINIDES~~

MATRIX
MATERIAL
(LiH, Cu, Al)

ACTINIDE OXIDES (SPHERES) (~0.1 IN.)

VOID FOR He (ALPHA DECAY)

TUNGSTEN LAYER (~0.005 IN.)

MOLY DISULFIED PROTECTIVE LAYER (~0.001 IN.)

TUNGSTEN, γ , SHIELD

LiH, η , SHIELD

SS, IMPACT PROTECTION

INSULATION (SWITCH)

GRAPHITE HEAT SHIELD

^{Silicide}
TEFLON HEAT SHIELD

FRAGMENTATION BARRIER

FIG 4

ΔV ABOVE 185.2 KM (100 N.MI) CIRCULAR ORBIT, 10^3 FT/SEC

SHUTTLE LAUNCHED STAGES

FIG 20

Fig 5

Table 1

LAUNCH VEHICLE COMPARISON EARTH ORBIT OR ESCAPE

<u>VEHICLES</u>	PAYLOAD, KG	\$ / KG
SATURN V / CENTAUR	49,000	4100
SHUTTLE (1) AND REUSABLE TUG (1)	8000	1380
SHUTTLES (2) AND REUSABLE TUG (1)	14,000	1490
SHUTTLES (2) AND REUSABLE TUGS (2)	17,000	1300
SHUTTLE (1) AND EXPENDABLE TUG (1)	11,000	2300

LOUGHEE & SONS CO.
MADE IN U.S.A.

July 6

EXTRATERRESTRIAL TRANSPORT COSTS

Figure 4. Tentative Antarctic Repository (dashed lines)
(Map adapted from Ref. 7)