

AN OVERALL ASSESSMENT OF ACTINIDE PARTITIONING AND  
TRANSMUTATION FOR WASTE MANAGEMENT PURPOSES\*

J. O. Blomeke, A. G. Croff, B. C. Finney  
Oak Ridge National Laboratory  
Oak Ridge, Tennessee 37830 U.S.A.

and  
D. W. Tedder  
Georgia Institute of Technology  
Atlanta, Georgia 30332 U.S.A.


**MASTER**

For Presentation at the Second Technical Meeting  
on the Nuclear Transmutation of Actinides,  
Ispra (Varese) Italy, April 21-24, 1980

**DISCLAIMER**

This book was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

*By acceptance of this article, the  
publisher or recipient acknowledges  
the U.S. Government's right to  
retain a nonexclusive, royalty-free  
license in and to any copyright  
covering the article.*

  
DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

\* Research sponsored by the Division of Waste Products, Office of Nuclear Waste Management, U.S. Department of Energy, under contract W-7405-eng-26 with the Union Carbide Corporation.

## **DISCLAIMER**

**This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.**

## **DISCLAIMER**

**Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.**



AN OVERALL ASSESSMENT OF ACTINIDE PARTITIONING AND  
TRANSMUTATION FOR WASTE MANAGEMENT PURPOSES\*

J. O. Blomeke, A. G. Croff, B. C. Finney  
Oak Ridge National Laboratory  
Oak Ridge, Tennessee 37830 U.S.A.

and

D. W. Tedder  
Georgia Institute of Technology  
Atlanta, Georgia 30332 U.S.A.

ABSTRACT

A program to establish the technical feasibility and incentives for partitioning (i.e., recovering) actinides from fuel cycle wastes and then transmuting them in power reactors to shorter-lived or stable nuclides has recently been concluded at the Oak Ridge National Laboratory. The feasibility was established by experimentally investigating the reduction that can be practicably achieved in the actinide content of the wastes sent to a geologic repository, and the incentives for implementing this concept were defined by determining the incremental costs, risks, and benefits. Eight U.S. Department of Energy laboratories and three private companies participated in the program over its 3-year duration.

A reference fuel cycle was chosen based on a self-generated plutonium recycle PWR, and chemical flowsheets based on solvent extraction and ion-exchange techniques were generated that have the potential to reduce actinides in fuel fabrication and reprocessing plant wastes to less than 0.25% of those in the spent fuel. Waste treatment facilities utilizing these flowsheets were designed conceptually, and their costs were estimated. Finally, the short-term (contemporary) risks from fuel cycle operations and long-term (future) risks from deep geologic disposal of the wastes were estimated for cases with and without partitioning and transmutation. We concluded that, while both actinide partitioning from wastes and transmutation in power reactors appear to be feasible using currently identified and studied technology, implementation of this concept cannot be justified because of the small long-term benefits and substantially increased costs of the concept.

1. INTRODUCTION

This paper summarizes the results of an overall assessment of the feasibility and incentives for operating the nuclear fuel cycle so that the most troublesome long-lived constituents of radioactive wastes are partitioned and transmuted [1]. Partitioning, when conducted for waste management purposes, is defined as treatment designed to reduce the levels of chemical elements having undesirable, long-lived isotopes in radioactive wastes to a greater extent than that dictated by normal economic considerations.

Transmutation is defined as a process whereby long-lived nuclides are converted to shorter-lived or stable nuclides by bombardment with subatomic particles, such as neutrons from nuclear power reactors. Partitioning and

---

\* Research sponsored by the Division of Waste Products, Office of Nuclear Waste Management, U.S. Department of Energy, under contract W-7405-eng-26 with the Union Carbide Corporation.

transmutation (P-T), when taken together, form a waste management concept which would be capable of reducing the amounts of certain long-lived, radioactive species normally present in radioactive wastes and converting them to shorter-lived or less toxic species.

Although we define P-T as a waste management option, it is clear that, when all of the impacts of P-T on the nuclear fuel cycle are considered, P-T is really a new fuel cycle option which would affect most fuel cycle operations to varying degrees. For example, it would:

1. require the installation of additional waste processing steps in reprocessing and mixed-oxide (MOX) fuel fabrication plants,
2. alter the volume and composition of radioactive wastes being sent to a waste repository,
3. require requalification of reactor fuels and possible new fuel fabrication techniques,
4. alter the neutronic behavior of nuclear reactors because of the presence of recycled actinides in the fuel, and
5. necessitate new shipping cask designs because of the increased amounts of neutron emitters present in nuclear materials containing recycled actinides.

Thus, P-T constitutes an overall fuel cycle concept that is significantly different from those involving the recycle of only the principal fissile and fertile values in spent fuels.

It is necessary for the fuel cycle being examined to be "closed" before the implementation of P-T can be evaluated or even considered. That is, provisions must exist for reprocessing spent fuel to recover the principal fissile and fertile values and fabricating them into fresh fuel. However, it should be noted that this project represents the evaluation of an advanced waste management alternative and should not be construed as advocating or implying the eventual implementation of these technologies.

### 1.1 Background

Studies have been made of various selected aspects of P-T since the mid-1960s. The most common type of study involved actinide transmutation calculations followed by calculation of the radiotoxicity of the high-level waste with and without transmutation of the actinides. The conclusion reached by most of these studies was that the radiotoxicity, and therefore the risk, due to high-level wastes in a repository could be reduced by factors of 100 to 200 for waste decay times greater than 1000 years [2]. These studies generally ignored partitioning, many of the more realistic aspects of transmutation, and other fuel cycle impacts of P-T.

Limited studies of partitioning processes and technology were conducted during 1973-1975 at the Oak Ridge National Laboratory (ORNL) [3]. The principal results of this work were (1) evaluation and incorporation of the experimental work into reprocessing plant flowsheets for partitioning actinides from the waste streams, and (2) recommendations concerning the approaches that should be used in future partitioning studies.

Only one realistic study has been made of the long-term benefits of removing the actinides from high-level waste [4]. The previously mentioned studies of long-term benefits that were based on the radiotoxicity index are not realistic because this index assumes that the wastes are ingested directly with no change in their composition, whereas in reality, any natural migration of these materials from the geosphere to the biosphere would not only retard their release but also would result in changes in the elemental and isotopic mixtures that might become available for ingestion. Reference [4], which was based on a pathway analysis, concluded that "... for the situations investigated the incentives for a special effort to remove any elements, including the transuranics, from high-level waste are vanishingly

small... ." However, since this study did not consider and compare the near-term risks and costs of removing the actinides from high-level waste to the calculated benefits, it is difficult to say conclusively that the incentives for actinide removal are "vanishingly small."

No studies have been made of the other varied, but important, impacts of P-T on the nuclear fuel cycle. Examples of these impacts are the effects of the highly neutron-active transplutonium isotopes on fuel fabrication, transportation, and handling; the effect of neptunium, americium, and curium on the in-reactor behavior, fabricability, and cladding compatibility of reactor fuels; and the disposition of actinides produced prior to the implementation of P-T. Finally, and most importantly, there has been no comprehensive study in which all short- and long-term advantages and disadvantages were included. This is necessary if the incentives are to be realistically evaluated.

In 1976, ORNL initiated a broadly based program consisting of both experimental and computational activities that were required to develop a meaningful evaluation of the P-T concept. Other organizations that participated in the program were Argonne National Laboratory, Brookhaven National Laboratory, Mound Laboratory, Savannah River Laboratory, Sandia Laboratory, Rocky Flats Plant, Idaho National Engineering Laboratory, the Ralph M. Parsons Company, Science Applications, Inc., and Los Alamos Technical Associates. During the first 2 years of the program, ORNL and the first seven organizations listed above conducted studies on specific aspects of P-T. The results of this work were used to develop fuel cycle material and facility descriptions which were, in turn, used as a basis for risk and cost analyses by the last three organizations. The final part of the program consisted of an overall assessment of the feasibility and incentives for P-T, and specification of the research, development, and demonstration (RD&D) requirements that would be needed for implementation. It is anticipated that the results of this program will be used by the U.S. Department of Energy (USDOE) as the technical backup for a policy decision on the future emphasis to be placed on P-T in the U.S. nuclear program.

## 1.2 Scope and Ground Rules

The procedures and detailed results of the initial studies and of the risk and cost analyses conducted in the latter part of this program are summarized in Refs. [1,5-10]. Details of these studies and analyses will not be given here except as required to support the feasibility, incentives, or RD&D assessments.

The basic ground rules used to guide the P-T program were as follows:

1. Only conventional chemical processes with a reasonably high assurance of near-term success and availability were considered.
2. The reprocessing flowsheets were based on coprocessing of uranium and plutonium in order that the resulting facilities be consistent with the U.S.A.'s nonproliferation objectives.
3. LWRs were taken as the primary transmutation devices, but FBRs were examined as potential alternatives.
4. The principal objective of the program was to obtain a meaningful and defensible analysis of the feasibility, incentives, and not the development of either the technology or an optimum P-T fuel cycle.

A qualitative evaluation of the potential impact of these approximations and assumptions is given in Sect. 1.4.



### 1.3 General Approach

The general approach used in this program was to perform an "incremental cost-risk/benefit analysis" of the P-T concept. To do this, two closed LWR fuel cycles were defined. These fuel cycles are the same in all respects except that the "reference" fuel cycle involved the use of a coprocessing flowsheet for recovery and recycle of uranium and plutonium, whereas the "P-T" fuel cycle used additional partitioning processes for the maximal recovery of all actinides in refabrication and reprocessing plant wastes. The reference and P-T fuel cycles are depicted jointly in Fig. 1. The reference fuel cycle includes everything to the left of the dashed, vertical line. The fuel charged to the PWR is assumed to be comprised of 33% MOX fuel and 67% enriched-uranium fuel. After an exposure of 33 GWd/MTIHM (3 years in the reactor), the fuel is discharged, allowed to decay for 1.5 years, and then transported to the fuel reprocessing plant (FRP). The fuel is reprocessed using a coprocessing Purex flowsheet adapted to recover and purify uranium and plutonium with only a partial separation of the uranium from the plutonium. After an additional 0.5-year decay, the recovered uranium and plutonium, together with a substantial fraction of the neptunium, is sent to the colocated MOX fuel fabrication plant (FFP) where 33% of the fuel for the next reactor loading is fabricated. Some additional enriched uranium may be required at this point to maintain the reactivity of

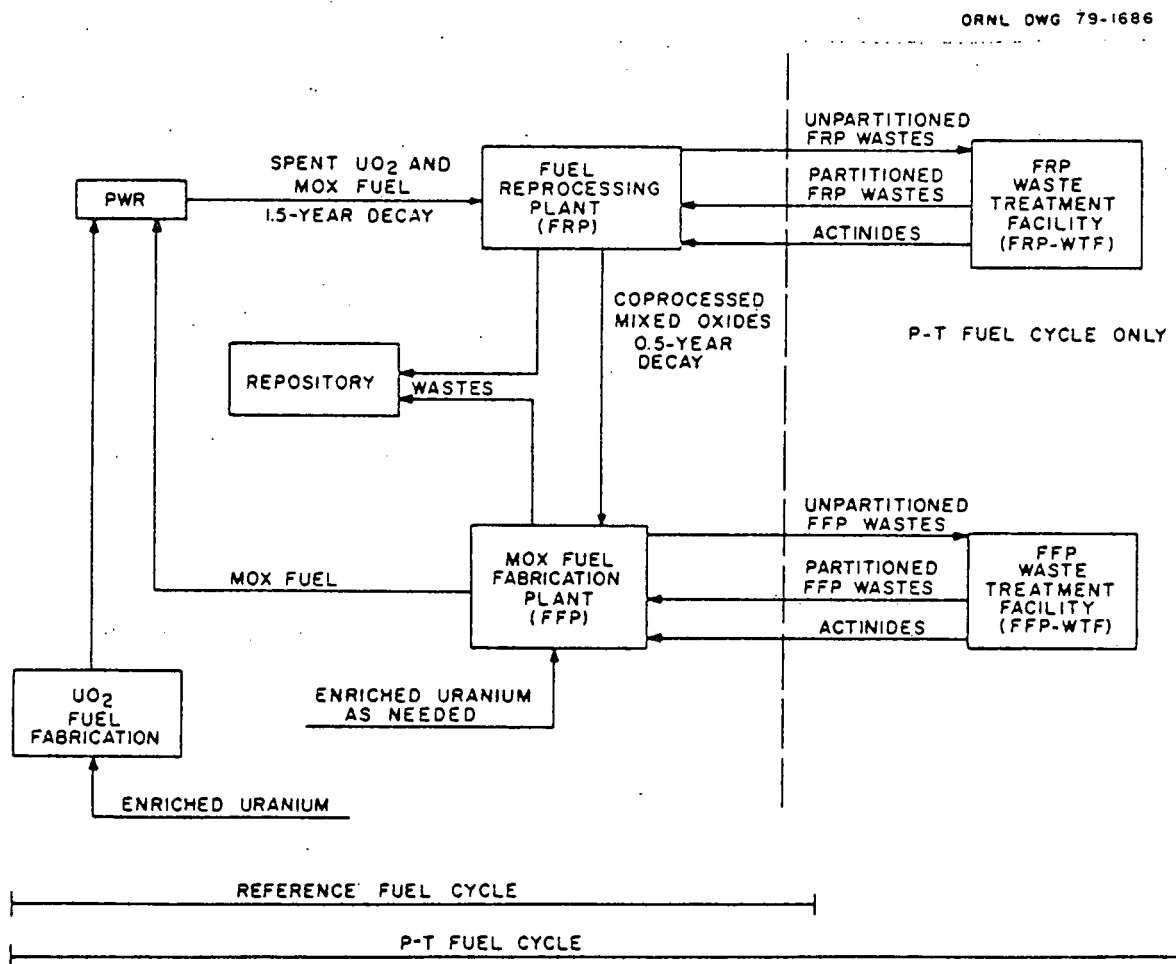


Fig. 1. Schematic diagram of the reference and P-T fuel cycles.

the MOX fraction since the fissile plutonium content decreases as it is recycled. The wastes sent to the repository include about 4% each of the uranium and plutonium, 25% of the neptunium, virtually all of the fission products, americium, curium,  $^{14}\text{C}$ , and all fuel assembly structural materials. The P-T cycle is similar to the reference cycle except that the waste treatment facilities (WTFs) (to the right of the dashed line) are brought into play. These facilities process the actinide-bearing wastes from the fuel reprocessing and MOX fuel fabrication plants to reduce their actinide contents. The streams returning to the reprocessing and fabrication plants are the actinide-depleted (partitioned) wastes and the actinides that were recovered by partitioning in the WTFs. The actinide-depleted wastes are conditioned for disposal, and the recovered actinides are mixed with the uranium and plutonium recovered in the FRP and fabricated into fresh MOX fuel. In the P-T cycle, the total amount of unrecovered actinides is about 0.25% of that in the spent fuel.

The fuel cycle operations where P-T is expected to have a significant impact were characterized according to three criteria: risks, costs, and benefits. Risk implies the short-term routine or accidental, radiological, or nonradiological impact of each facility. This risk was developed by using effluent source terms for routine operation and/or probabilistic accident frequencies and consequences with conventional meteorological and biological pathway models. The third criterion, which denotes only the monetary cost, is generated by developing conceptual plant designs and applying standard costing techniques. The benefits refer to the reduction in the long-term probabilistic dose from a waste repository. These benefits are determined using computer codes that calculate the probabilistic accident consequences for the repository and then use conventional meteorological and biological models to determine the radiation dose to man.

The "incremental" risks, costs, and benefits of the operations in the two fuel cycles that are attributable to P-T were calculated by examining separately each of the fuel cycle operations depicted in Fig. 1. The advantage of using an incremental analysis is that those operations not affected by P-T will be identical in the two cycles (e.g.,  $\text{UO}_2$  fabrication) and need not be considered at all.

The only fuel cycle operations expected to be significantly affected by P-T, and thus have nonzero incremental changes in the risks, benefits, or costs, are (1) the FRP and its waste treatment facility, (2) the MOX fuel fabrication plant and its waste treatment facility, (3) the waste repository, (4) the transmutation reactor, and (5) the transportation of fuels and wastes. By defining the scopes of the fuel reprocessing and MOX fuel fabrication plants appropriately, it was possible to neglect the differences in these two facilities in the reference and P-T cycles and consider only their respective waste treatment facilities.

The final step in the incentives analysis was to compare the risks, costs, and benefits. This was done, first, by comparing them individually to other "reference" values such as natural background for the risks and benefits and the cost of electricity for the "no P-T" (reference) case. Next, the risks, costs, and benefits were combined to yield the cost of reducing the long-term risk from the waste repository by 1 man-rem (i.e., dollars per man-rem for the P-T cycle). This value was then compared to the \$1000/man-rem guideline used in the United States to determine whether additional waste treatment should be supplied for reactor effluents. The sensitivity of the incentives for determination to uncertainties in data and the effects of major assumptions was also examined.



## 1.4 Major Assumptions

Four major assumptions were made in this assessment of P-T. The first was that there is no process capable of partitioning the actinides that would have a very low cost and impact (i.e., a "magical" process). We believe that, even though partitioning processes superior to those described herein might be developed, they would not reduce the costs and impacts of P-T by more than a factor of 2.

The second major assumption was that certain technological aspects of P-T are feasible even though they have no firm experimental basis. Two important examples are fuel performance and the operation of partitioning facilities closely coupled to reprocessing and refabrication plants. Fuels containing high concentrations of neptunium, americium, and curium were assumed to have acceptable irradiation behavior based only on the acceptability of uranium-plutonium fuels. All of the processes included in the WTFs have been tested experimentally to some extent. However, there have been no tests of an integrated flowsheet, with its many recycle streams and possible impurities. Until such integrated tests are performed, the effect of recycling these streams on the operability and performance of the facility is uncertain.

The third major assumption was that a probabilistic risk analysis is an acceptable measure of the actual risks of operating fuel cycle facilities and waste repositories. Although this approach has been widely used, it has never been formally accepted by the responsible government agencies. Finally, the fourth major assumption made in this analysis is that the impact of P-T on fuel cycle facilities other than reprocessing and refabrication plants is negligible and, therefore, that these facilities need not be considered. In reality, there would probably be second- or third-order effects of a detectable, but probably insignificant, magnitude on the other facilities. The accuracy of this approximation can be verified only by performing a detailed conceptual design of all fuel cycle facilities, an undertaking that was far beyond the scope of this program.

## 2. PARTITIONING

Conceptual partitioning flowsheets were developed for both the fuel reprocessing and the MOX fuel fabrication plants [5].

### 2.1 Actinide Partitioning Flowsheets [5].

Partitioning of the wastes involves two generic steps: separation of the actinides from the waste, and recovery of the actinides. The separation step is accomplished by either leaching or breaking down strong organic-actinide chemical compounds that interfere with their recovery. Recovery is effected by using the following techniques:

1. The tetravalent and hexavalent actinides (uranium and plutonium) and neptunium are recovered by TBP extraction, stripped from the solvent, and returned to the parent facility.
2. The trivalent actinides (americium and curium) and lanthanides are coextracted from the waste using a bidentate compound, dihexyl-N,N-diethylcarbamylmethylene phosphonate (CMP). The residual waste is returned to the parent facility for solidification before disposal.
3. The trivalent actinides are separated from the lanthanides using CEC. The lanthanide fraction is returned to the parent facility and mixed with the treated HLLW prior to solidification. The recovered actinides (americium and curium) are returned to the parent facility for conversion to the oxides

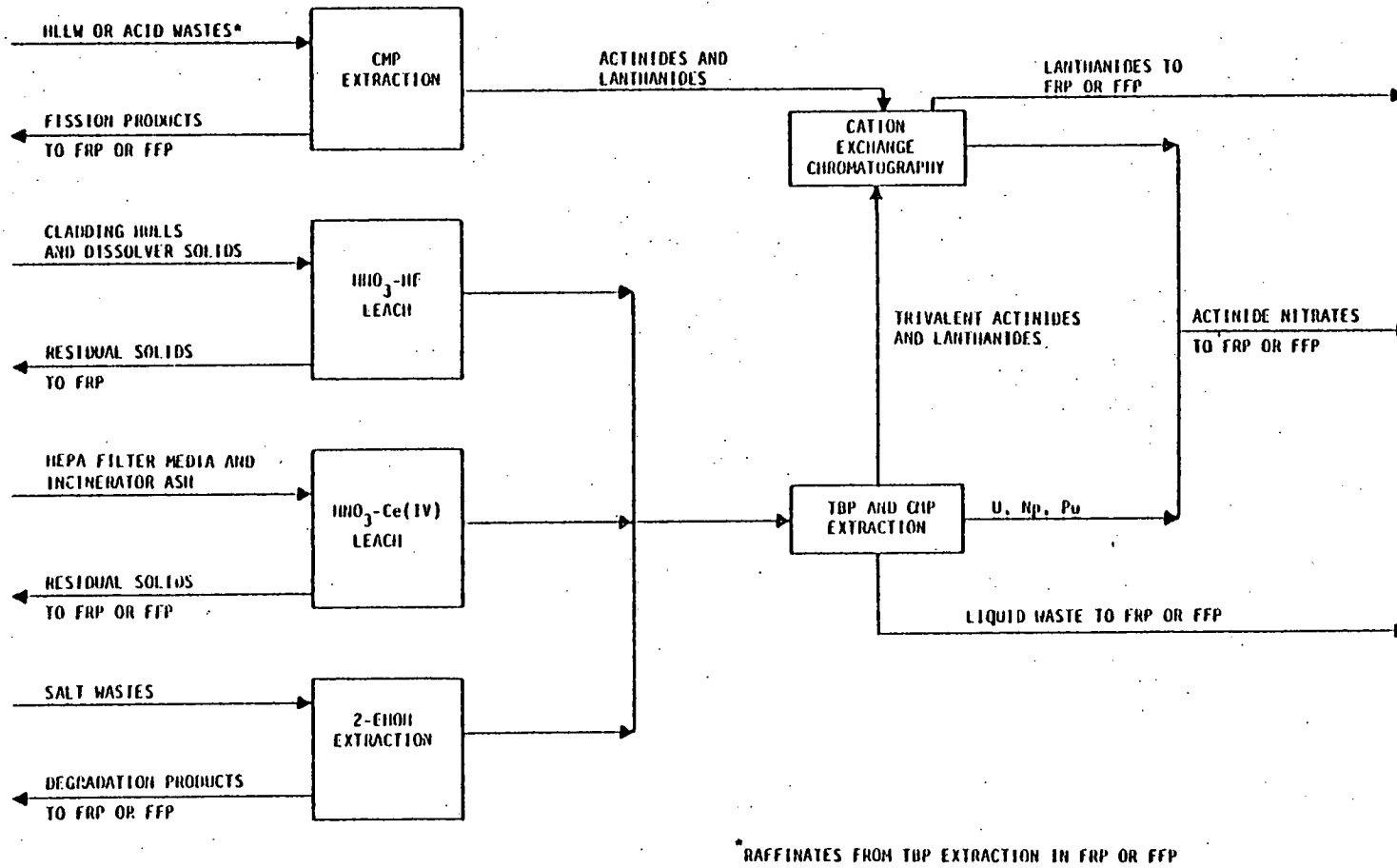


Fig. 2. Generic actinide partitioning flowsheet

This sequence is depicted schematically in the generic flowsheet shown in Fig. 2.

The high-level liquid waste (HLLW), which is the raffinate from a TBP first-cycle solvent extraction in the FRP, is fed directly to the CMP extraction and CEC processes. The cladding hulls and dissolver solids, which have been previously leached with nitric acid, are subjected to a final leaching with  $\text{HNO}_3$ -HF for removal of additional actinides. The HEPA filter and incinerator ash wastes from both the FRP and the FFP contain actinides that are also largely insoluble in nitric acid. Consequently, these are leached with  $\text{HNO}_3$ -Ce(IV) solution. The Ce(IV) is produced by the electrolytic oxidation of Ce(III); and when the leaching is complete, oxalic acid is added to the system to convert Ce(IV) to Ce(III) to reduce equipment corrosion rates. The leachate is first contacted with TBP for the recovery of uranium, neptunium, and plutonium and the trivalent actinides are then recovered by extraction with CMP followed by CEC. The salt wastes, principally  $\text{Na}_2\text{CO}_3$  solutions from solvent cleanup, contain a variety of actinides, some of which are bound in non-strippable organic phosphoric complexes. These complexes are destroyed by extraction of the phosphoric acids with 2-EHOH, and the actinides are then recovered by TBP and CMP extraction followed by CEC.

All of the actinide-depleted wastes are returned to the parent facility for final treatment (i.e., vitrification or concretion) and packaging for disposal. In general, the increases in waste volumes are held to a minimum because the use of chemicals that can be recovered and recycled is emphasized. Only the concreted wastes increase significantly in volume (~50%), and this is attributable to the reagents used in solvent cleanup and CEC.

The total actinides reporting to the fuel cycle wastes for both the reference case and the P-T case are summarized in Table 1. As is evident, the total amount of unrecovered actinides has been decreased over the reference cycle by factors of 16 for uranium and plutonium, 100 for neptunium, and 400 for the transplutonium actinides. Only 25% of the neptunium reports to wastes in the reference cycle because it follows the uranium and plutonium in the FRP's coprocessing flowsheet.

Table 1. Actinides in wastes of reference and P-T cycles as a percentage of facility feed

Element	Fuel Reprocessing Plant		MOX Fuel Fabrication Plant	
	Reference cycle	P-T cycle	Reference cycle	P-T cycle
Uranium	2.0	0.1	2.0	0.15
Neptunium	21.4	0.1	4.5	0.15
Plutonium	2.0	0.1	2.0	0.15
Americium <sup>a</sup>	100.0	0.1	20.5	0.15
Curium <sup>a</sup>	100.0	0.1	20.5	0.15

<sup>a</sup>The same values are assumed for berkelium and californium.

## 2.2 Partitioning of Technetium and Iodine

While procedures for partitioning technetium from the HLLW and dissolver solids in which it occurs were not identified in these studies, we believe that acceptable techniques could be devised within the framework of the

actinide partitioning flowsheets that have been developed. Iodine partitioning is, in effect, already incorporated in the reference FRP flowsheets. Its removal from FRP streams by techniques such as caustic scrubbing, mercuric nitrate scrubbing, zeolite adsorption, charcoal adsorption, and the Iodex process has been demonstrated.

### 2.3 Feasibility of Actinide Partitioning

Six identifiable processes used in the partitioning facilities are described in Sect. 2.1: Purex (TBP) extraction, CMP extraction, cation exchange chromatography, 2-EHOH extraction,  $\text{HNO}_3$ -HF leaching, and  $\text{Ce(IV)}-\text{HNO}_3$  leaching. Based on the engineering evaluations and experimental studies conducted as a part of this program, we believe that all of these processes are feasible and, consequently, that partitioning per se is feasible. Both TBP extraction and  $\text{HNO}_3$ -HF leaching are well demonstrated technologies that have been widely used within the nuclear program. CEC has also been demonstrated, although less extensively than has the Purex process. The CMP extraction process has been tested under radioactive conditions on small samples of discharged LWR fuel and on Idaho Chemical Processing Plant waste. Finally, both the 2-EHOH extraction and the  $\text{Ce(IV)}-\text{HNO}_3$  leach processes have been demonstrated with tracer levels of actinides on a laboratory scale.

The principal caveat concerning the partitioning flowsheets developed in the course of this program is that they are very highly integrated (i.e., considerable internal recycle of plant streams is required) to prevent actinides from reporting to the wastes. As a result, it is possible that one or more chemical species having a detrimental effect on plant performance could be formed and continue to build up within these streams. Additionally, it should be noted that the CEC process, while judged to be feasible, has some serious operational drawbacks resulting from radiation damage to the resins by  $^{242,244}\text{Cm}$ . This results in a possible safety hazard, due to the production of explosive degradation products, and in increased cost and operational complexity.

### 2.4 Advanced Partitioning Processes

Taken in their entirety, these partitioning flowsheets are believed to constitute a near-optimal partitioning technology since (1) the reagents are relatively radiation-stable, (2) the wide variety of wastes to be treated will require multiple processes in any case, and (3) the processes generally involve the use of reagents that permit the use of relatively small and reliable equipment and result in only modest increases in final waste volumes. The exception is the CEC process, where considerable improvement is desirable. A second possible advancement of partitioning technology would be the integration of partitioning directly into the parent facilities (i.e., the FRP or FFP), thus allowing overall optimization. Based on the cost attributable to CEC and our engineering judgment, we believe that the combination of an improved actinide-lanthanide separation process and an optimized, integrated reprocessing-partitioning flowsheet might reduce the cost of partitioning by as much as a factor of 2.

## 3. TRANSMUTATION [6]

### 3.1 Actinide Transmutation

Much of the available information on actinide transmutation is quantitatively useless because of flaws in the calculational assumptions or



in the methodology. The two principal problems in this regard are that (1) many of the calculations allow unfissioned actinides to be removed from the system during recycle and thus are not "closed"; and (2) incorrect criteria are used to determine the enrichment of the fuel, which results in substantial errors in the composition and transmutation rate of the recycled actinides.

The primary transmutation device considered in this study was a PWR fueled with self-generated plutonium. Recycle of the actinides to this reactor is possible by any of several methods: (1) dispersing them homogeneously in enriched uranium fuel, (2) dispersing them only in fuel that is enriched with plutonium, (3) concentrating them in target rods in a normal fuel assembly, and (4) concentrating the actinides in an assembly composed totally of target rods. Based on considerations of transmutation rate, fabrication cost, and reactor operational considerations, it appears that the second option is the preferred mode.

Based on the transmutation studies conducted as a part of this program as well as the results of other studies, the following statements concerning actinide transmutation in PWRs can be made:

1. Actinide transmutation rates range from 5 to 7% per full-power year (2.5 to 3.5% per calendar year).
2. The  $^{235}\text{U}$  enrichment penalty resulting from actinide recycle varies from cycle to cycle but declines from a penalty, initially, to zero at about the fifth recycle. Thereafter, the recycled actinides result in a decrease in enrichment (i.e., benefit).
3. Recycle of the actinides results in increased amounts of  $^{238}\text{Pu}$  and transplutonium actinides in the fuel, particularly the troublesome  $^{242}\text{Cm}$ ,  $^{244}\text{Cm}$  and  $^{252}\text{Cf}$  nuclides.
4. The presence of the actinides in the PWR may cause significant power peaking which may, in turn, require more extensive enrichment grading within fuel assemblies.

Finally, and most importantly, subject to the acceptability of the actinide recycle fuels (see Sect. 4.2), transmutation of the actinides appears to be feasible and the impacts on the reactor do not appear to be unduly large. However, operational changes would probably be required as a result of the radiation and neutronic characteristics of the P-T fuels.

### 3.2 Technetium and Iodine Transmutation

Only brief, survey calculations have been performed concerning the transmutation of technetium and iodine. If present as the metal (melting point about  $2400^\circ\text{K}$ ),  $^{99}\text{Tc}$  can be transmuted at the rate of about 11% per full-power year. The products of the irradiation are, for the most part, stable isotopes of ruthenium. Thus, the transmutation of  $^{99}\text{Tc}$  appears to be feasible subject to the identification of an acceptable fuel form and to the determination of the impact of small amounts of long-lived  $^{98}\text{Tc}$  that would be formed by irradiation of the  $^{99}\text{Tc}$ .

Assuming that iodine is present as sodium iodide, its transmutation rate is about 3% per full-power year, which is relatively low. In addition, most iodine compounds are volatile or unstable at reactor operating temperatures; iodine is very corrosive toward virtually all metals; and the transmutation product is xenon gas, which could cause fuel rod pressurization problems. Thus, it would appear that the transmutation of iodine in power reactors should be considered to be marginally feasible at best.

### 3.3 Impact of Alternative Transmutation Devices

The use of thermal power reactors other than PWRs has been investigated to some extent. In general, all of them were found to perform similarly to the PWR when operating with their own self-generated actinides.

The use of high-flux thermal reactors as transmutation devices would offer higher transmutation rates. However, these reactors are very expensive to operate since they (1) typically require highly enriched uranium cores, (2) usually operate at temperatures near the normal boiling point of water and thus do not produce usable power, and (3) are generally small due to heat removal considerations. Thus, high-flux thermal reactors should not be considered for this application.

The use of LMFBRs as a transmutation device is the most widely studied actinide transmutation option. The following statements can be made about transmutation in LMFBRs:

1. The transmutation rate ranges from 5 to 11% per full-power year, depending on the actinide composition and the reactor design.
2. Actinide recycle has no significant impact on the fissile requirements, breeding performance, or power peaking in the LMFBR.
3. LMFBRs produce lower amounts of the principal neutron-active nuclides, particularly  $^{252}\text{Cf}$ .
4. Actinide transmutation is feasible in LMFBRs, subject to the fabrication of acceptable P-T fuels.

Based on the available information, it would appear that given a choice between an existing LMFBR and an existing PWR as a transmutation device, the LMFBR has a significant advantage. However, transmutation could be implemented using only thermal reactors, provided adequate supplies of fissile material remain available.

The use of projected fusion reactors as transmutation devices has also been extensively studied. Recent calculations indicate that, while actinide transmutation in the fusion reactor blanket is feasible, the transmutation rates will probably not exceed those in an LMFBR due to material radiation damage and heat transfer limitations.

The use of accelerator-driven spallation neutron transmutation devices would appear to be an extremely inefficient method for producing neutrons in light of the energy losses in, first, making electricity from heat and, then, in converting the electricity to highly energized, charged particles. Furthermore, these devices are still limited by the same materials damage and heat transfer considerations as LMFBRs. Thus, it is likely that spallation reactors would be at a substantial cost disadvantage with respect to LMFBRs and LWRs with no overriding benefits to compensate for this.

Nuclear explosives have also been proposed as a means for transmutation. However, the large number of explosions required, their social and political unacceptability, the fact that the residuals are irretrievably entombed wherever the explosion occurs, and probable future restrictions on nuclear explosives make this option very unattractive.

#### 4. OTHER IMPACTS OF P-T [7]

Four other impacts of P-T on the fuel cycle that do not fall within either the partitioning or transmutation areas are:

1. the effect of P-T fuels on fresh and spent fuel shipping cask design,
2. the impacts of P-T on fuel fabrication and the suitability of fuels containing recycle actinides.
3. the impact of P-T on the disposition of fuel and/or waste inventories that will accrue before partitioning could be implemented, and
4. the relationship of P-T to current U.S. nuclear policy.

#### 4.1 Transportation Impacts

The design of a shipping cask for both fresh and spent P-T fuels is markedly different from that for ordinary MOX fuels because of the very high neutron activity caused by  $^{242}\text{Pu}$ ,  $^{244}\text{Pu}$  and  $^{252}\text{Cf}$  present in the P-T fuels. A shipping cask that appears to meet all applicable requirements was conceptually designed as a part of the ORNL P-T program. In contrast to ordinary casks, which are typically comprised of lead, steel, or uranium, this P-T cask would be comprised of major amounts of boron carbide, copper, and lithium hydride with stainless steel being used for structural integrity. These unconventional materials are necessary to reduce the external dose rate from neutrons to acceptable levels while maintaining the accident resistance required of the casks. Another important aspect of the cask is that its payload is only two-thirds of that for existing spent fuel casks because of weight and size limitations.

#### 4.2 Fabrication Impacts

The presence of the neutron emitters in the P-T fuels also affects the design philosophy of the MOX fuel fabrication plant. In the reference case, it was assumed that the fuel was fabricated remotely but that the plant could be decontaminated for contact maintenance. However, in the P-T MOX fuel plant, the fuel is considerably more radioactive due principally to the neutrons. Increased shielding thickness must be provided; in addition, it is unlikely that the plant could be sufficiently decontaminated to permit contact maintenance. Thus, the P-T MOX fuel fabrication plant was assumed to be both remotely operated and remotely maintained. This results in a substantial cost penalty (see Sect. 5.1.1) in the fabrication process and raises additional feasibility questions since a facility of this type has never been built or operated.

A second impact of P-T on fabrication is the higher concentrations of neptunium, americium, and curium in the fuel. The resulting effect is to question the suitability of the fuels with respect to their irradiation behavior (cracking, swelling, etc.) and their compatibility with the cladding. In the recycle modes where these actinides are relatively dilute, this will probably not be a problem. However, if target rods or assemblies with their higher actinide concentrations are used, significant difficulties may be manifested.

#### 4.3 Impacts on Existing Inventories

The possible implementation of P-T at some time in the future leads to considerations of the disposition of current spent fuel and/or waste inventories. If too much of this material is committed to a repository, the overall effect of P-T will be small because of the large amount of actinides already in the repository. On the other hand, holding this material on the surface is both more expensive and more risky. However, the decisive argument would appear to be that, since the actinides are shown to have a very small impact on the risk from the repository, the quantity that is committed to a repository is not of overriding importance.

#### 4.4 Policy Implications

The implementation of P-T would conflict with current United States policy that defers both reprocessing and the recycle of plutonium. Specifically, P-T would require that the spent fuel be reprocessed and that all actinides, including plutonium, be recycled. In fact, the processing and actinide recycle required by P-T would be considerably greater than that for a standard uranium-plutonium fuel cycle.

A second policy conflict would occur if LMFBRs were to be used as transmutation devices since the use of LMFBRs has also been deferred. Implemen-

tation of P-T would require the eventual use of LMFBRs because of long-term limitations in the supply of fissile materials for thermal reactors.

## 5. ANALYSIS OF THE INCENTIVES FOR P-T

### 5.1 Costs, Risks, and Benefits of P-T

The cost of P-T is taken to be the increase in the cost of nuclear electricity that would be realized by implementation of P-T. The risks include the increases in short-term (contemporary) radiological and nonradiological risks to the public resulting from the additional steps in the processing, the larger quantities of actinides in the fuel cycle, and the greater number of fresh and spent fuel shipments required. The benefits of P-T are derived from the reduction in long-term risk from the repository that contains actinide-depleted wastes.

#### 5.1.1 Costs

The incremental cost of implementing P-T was estimated to be 1.58 mills/kWh(e) [7,8]. This cost is attributed to various fuel cycle functions, as follows: reprocessing waste treatment facility, 0.62 mill/kWh(e) [39%]; MOX fuel fabrication waste treatment facility, 0.46 mill/kWh(e) [29%]; P-T MOX fuel fabrication penalty, 0.30 mill/kWh(e) [19%]; transportation, 0.16 mill/kWh(e) [10%]; waste management, 0.03 mill/kWh(e) [2%]; and fissile carrying charges, 0.01 mill/kWh(e) [1%].

The two waste treatment facilities are the principal partitioning facilities in the P-T fuel cycle. The MOX fabrication penalty results from increased shielding thickness and remote maintenance requirements. The incremental transportation and waste management costs stem from the larger waste volume and the smaller capacity of the cask needed to carry the fresh and spent P-T fuel. The fissile cost is due to the interest charges on the larger amount of plutonium in the P-T cycle.

The 1.58 mills/kWh(e) cost is equivalent to (1) \$419 per kilogram of heavy metal charged to the reactor, (2) 6.2% increase in the cost of nuclear-generated electricity, and (3) \$11.1 million per GW(e)-y.

#### 5.1.2 Risks

The increase in short-term risk to the general public resulting from the implementation of P-T is 0.57 health-effect per GW(e)-y [9]. This figure is comprised of (1)  $3 \times 10^{-3}$  health-effects/GW(e)-y from increases in routine (continuous) radiological releases due to the increased processing and increased concentrations of toxic actinides; and (2) 0.57 health-effect/GW(e)-y due to increases in nonradiological risks, principally petroleum combustion products from the generation of steam and heat at the waste treatment facilities and from physical damage during transportation. As a basis of comparison, the rate from natural background is about 1.0 health-effect/GW(e)-y.

#### 5.1.3 Benefits

The benefit of P-T is the reduction in the expected long-term dose from the geologic repository, assumed in this analysis to be located at the site of the Waste Isolation Pilot Plant (WIPP) in New Mexico. The measure of the long-term risk used in this study is the number of health effects expected from the repository over 1 million years, per unit of electrical capacity represented by the waste in the repository. This risk value is 5.16 health-effects/GW(e)-y for the reference case and 5.10 for the P-T case, giving a



benefit of 0.06 health-effect/GW(e)-y [10]. These values are about 0.08% of the health effects due to natural background. The benefit is principally derived from the reduction in the amount of  $^{226}\text{Ra}$  in the repository. This benefit is small because the nuclides that control the expected risk from the repository are  $^{99}\text{Tc}$  and  $^{129}\text{I}$ , which constitute 92% and 8% of the integrated 1-million-year risk, respectively. The risk value includes contributions from a slow-leach incident, a volcano growing through the repository, and the impact of a very large meteorite. Because the probability of either the volcano or the meteorite impact is small (about once every 100 billion years), these two events account for a small contribution to the total repository risk. In the more probable leach incident (about once every 7 million years), the actinides are retarded during their migration through the geosphere to such an extent that they do not emerge into the biosphere in significant quantities within a million years.

#### 5.1.4 Summary

Table 2 gives a summary of the costs, risks, and benefits of P-T expressed on a GW(e)-y basis. The risk values in terms of health effects were converted to man-rem using a conversion factor of  $2 \times 10^{-4}$  health effect/man-rem. [11].

Table 2. Summary of the costs, risk, and benefits of P-T per GW(e)-y

	Reference	P-T	Incremental
<u>Costs</u>			
Fuel cycle cost, \$	$178.9 \times 10^6$	$190.0 \times 10^6$	$11.1 \times 10^6$
<u>Short-Term Risk</u>			
Radiological dose, man-rem	4	20	16
Total risk, <sup>a</sup> man-rem	1700	4550	2850
Comparison: Natural background, man-rem	5000	5000	0
<u>Long-Term Benefit</u>			
Radiological dose reduction, man-rem <sup>b</sup>	25,800	25,500	300
Comparison: Natural background, man-rem <sup>b</sup>	$33.5 \times 10^6$	$33.5 \times 10^6$	0

<sup>a</sup>Includes nonradiological risks expressed as equivalent radiological impact using a conversion factor of 5000 man-rem/health effect.

<sup>b</sup>Expected dose integrated over 1 million years.

#### 5.2 Incentives for Actinide P-T

The method used to determine whether there are any incentives for implementing actinide P-T was to calculate the cost of reducing the expected long-term risk by 1 man-rem and compare it to the \$1000/man-rem guideline that

has been prescribed for use in determining whether additional effluent control systems on reactor plants are justified [12]. If the cost of P-T does not meet this guideline there are presumably alternative investments for the money that would save more lives than P-T; therefore, its implementation is not warranted.

It is evident from the data of Table 2 that there is more than one way to generate the cost per man-rem figure, principally because of the components of the short-term risk. For the purposes of this analysis, three values were developed. The first was the cost of reducing the expected long-term risk irrespective of the short-term risk:  $\$11,100,000/300 \text{ man-rem} = \$37,000/\text{man-rem}$ . The second value was based on the net radiological risk reduction, which is the long-term risk minus the short-term risk. This value is  $\$11,100,000/(300 - 16) \text{ man-rem}$ , or  $\$39,000/\text{man-rem}$ . The final value, which is based on the overall risk reduction, including the short-term nonradiological risks, is  $\$11,100,000/(300 - 2850) \text{ man-rem}$ , or  $-\$4400/\text{man-rem}$ . This negative value should be interpreted as meaning that, since the short-term risks of P-T exceed the long-term benefits, we must pay \$4400 to increase the overall risk by 1 man-rem. It should be noted that, if a nuclear plant were used to generate the process heat for the partitioning facilities, this value would be much smaller (in an absolute sense), although it would still be negative because of the risk from transportation accidents.

In summary, all three of the values fail by a wide margin to meet the \$1000/man-rem criterion that would justify P-T; thus there are no apparent incentives for its implementation. The justification for rejecting radiological protection options on this basis is supported by the 1972 BEIR report [11]:

"The public must be protected from radiation but not to the extent that the degree of protection provided results in the substitution of a worse hazard for the radiation avoided. Additionally, there should not be attempted the reduction of small risks even further at the cost of large sums of money that spent otherwise would clearly produce a greater benefit."

### 5.3 Sensitivity Analysis

The validity of the conclusion that there are no safety or cost incentives to implement P-T is subject to the sensitivity of the results to the assumptions and uncertainties in the analysis. The uncertainty in both the cost analysis and the short-term risk analysis is estimated to be about a factor of 2. The uncertainty in the cost analysis represents the possible error in cost estimation and the likely impact of partitioning improvements. The uncertainty in short-term risk, which is small relative to most risk analyses, results from uncertainties in the impact models used in the analysis. Such uncertainties are smaller than those in most risk studies because the principal contributors to the risk are routine effluents, which have a probability of 1.0. By using these maximum uncertainties in a manner most favorable to P-T, one can calculate that the expected long-term benefit of actinide P-T must be about 5500 man-rem/GW(e)-y to reach the \$1000/man-rem value, about a factor of 20 larger than the calculated value of 300 man-rem/GW(e)-y. The question is now reduced to considering the uncertainty of the long-term risk analysis.

The long-term risk analysis has a number of large, known conservatisms built into it. Principal among these are (1) the time horizon is extremely long, i.e. 1 million years; (2) the release fractions from the volcano and meteorite incidents are a factor of 10 to 20 greater than values which have been used elsewhere [13], and these directly affect the long-term benefits; (3) the fact that the repository risk model does not provide for the removal

of nuclides from the zones surrounding the repository by wind, river flow, etc.; (4) the population is assumed to remain in place both during and after volcanoes and meteorites; and (5) a conservatively small retardation factor is used for  $^{237}\text{Np}$  ( $K_d = 8.1$ ), the actinide most likely to emerge first into the biosphere following a leach incident. It should be noted that this approach was used simply because it was an expeditious way to examine the incentives for implementing P-T, and should not be construed as being necessarily appropriate for other studies of the same area or for studies of other areas.

On the other hand, there are other principal parameters where changes might significantly increase the benefits of P-T. First, changes in either the probability of the release fraction from the volcano or meteorite incidents, which are totally responsible for the calculated benefits of P-T, would directly affect the magnitude of the benefits. As noted above, the release fraction is believed to be conservative by a factor of 10 to 20, although the degree to which this value might vary is unknown.

A second class of parameters that might increase the calculated benefits of P-T is related to the leach incident. The 300 man-rem/GW(e)-y benefit from actinide P-T does not include any contribution from the leach incident since the actinides are retarded to such an extent that they do not emerge into the biosphere within 1 million years. However, by allowing changes in one or more of the leach parameters, the migration rate of the actinides can be accelerated to the point that they do emerge and contribute to the calculated leach incident consequences. The only parameter that could possibly accomplish this is an increase in the water migration velocity through the geosphere. It should be noted that a considerable increase in this value is required before any changes in the benefits would be observed, but thereafter the benefits would increase linearly with parameter increases. That is, there is a threshold below which the leach incident is insignificant. The water velocity assumed for this analysis is 1.46 m/year. The range of velocities measured at the WIPP site varied from essentially zero to a maximum of 4.6 m/year. Increasing the water velocity to 4.6 m/year would increase the benefits of P-T by a factor of 10 or less. However, an additional compensating factor is that the probability of a leach incident used in the analysis ( $2 \times 10^{-7}$ /year) is believed to be conservatively high. Thus, it would appear that the known conservatisms in the leach incident calculation would more than compensate for any possible uncertainties and unknown nonconservatisms in the analysis.

#### 5.4 Incentives for Technetium and Iodine P-T

The benefits of technetium and iodine P-T, based on the conservative assumptions described previously, were about 100 times that of actinide P-T, or about 30,000 man-rem/GW(e)-y. In addition, the costs and short-term risks of partitioning may be smaller since technetium occurs only in the HLLW and dissolver solids and iodine is found only in the dissolver off-gas. Thus, it is likely that the cost-risk/benefit values for technetium and iodine P-T would be less than the \$1000/man-rem criterion. This would indicate that, under the very conservative conditions used in the long-term risk analysis, there is an incentive for technetium P-T and, if iodine transmutation is feasible, there is an incentive for iodine P-T. It should be noted that this conclusion is a direct result of the conservative value used for the leach incident probability. Even if this probability were reduced by only a factor of 100, the incentives for both technetium and iodine P-T would likely be eliminated.

## 6. RESEARCH, DEVELOPMENT, AND DEMONSTRATION REQUIREMENTS

As might be expected, considerable RD&D would be required before actinide P-T could be commercially implemented. Partitioning studies are needed in a variety of areas. Principal among these are testing the integrated flow-sheet and searching for an alternative to the CEC process for separating actinides and lanthanides. Cross-section measurements and actinide transmutation studies are required to identify more precisely the best recycle modes and their likely impacts. Studies in the fabrication area, including test irradiations, are required to determine the acceptability of fuels containing higher-than-normal concentrations of neptunium, americium, and curium. A shipping cask must be built of the relatively unusual materials required to ship the neutron-active P-T fuels to ensure that they can be fabricated and that the cask meets applicable safety regulations. Finally, overall studies of the best methods for recycling the actinides would be required.

It is estimated that this RD&D would take about 15 years and that it should be possible to have the first partitioning facilities on-line in 20 years, assuming that the program was well-supported and that no licensing delays occurred. The limiting aspects in this entire process are the RD&D requirements and the design and construction of the partitioning facilities. The estimated cost of the RD&D, excluding the costs of the commercial-sized plants, is about \$900 million (1979 dollars). The largest portion of this money would go for a hot (radioactive) pilot plant for testing the integrated partitioning flowsheets.

## 7. CONCLUSIONS

### 7.1 Feasibility

1. The partitioning of actinides appears to be feasible based on the use of currently identified technology that has been experimentally tested and some of which has been used in full-scale operations.
2. The partitioning of technetium has not been adequately investigated, but there is presently no reason to believe that its recovery is not feasible.
3. Iodine partitioning is feasible using existing, demonstrated technology.
4. The transmutation of actinides appears to be feasible in thermal, fast, and fusion reactors subject to the acceptability of fuels containing higher-than-normal concentrations of neptunium, americium, and curium.
5. The transmutation of technetium appears to be feasible subject to the identification of an acceptable fuel form.
6. The transmutation of iodine is marginally feasible at best because of low transmutation rates, the volatility of iodine compounds, the production of xenon gas as a transmutation product, and the corrosiveness of iodine and its compounds.
7. The transportation of highly neutron-active P-T fuels appears to be feasible at a reasonable cost.
8. Partitioning-transmutation may not currently be implemented in the United States because of a nuclear policy which defers both reprocessing and plutonium recycle.

### 7.2 Incentives

1. The cost of actinide partitioning is relatively high, \$11.1 million/GW(e)-y, due to the variety of wastes that must be partitioned.
2. The short-term (contemporary) risks from P-T are substantial if the non-



radiological impacts are taken into account, amounting to 0.57 health-effect/GW(e)-y. The short-term radiological risks are small, amounting to 0.003 health-effect/GW(e)-y.

3. The long-term benefits (i.e., risk reduction) of P-T, using very conservative assumptions, is small, amounting to only 0.06 health-effect/GW(e)-y.
4. There are no incentives for actinide P-T, even if very conservative assumptions are used in the analysis. The cost of the actinide P-T benefits is \$39,000/man-rem if nonradiological risks are ignored, and the short-term risks exceed the long-term benefits integrated over a million years if nonradiological risks are included.
5. There may be incentives for technetium P-T if very conservative long-term risk analysis assumptions continue to be used and if partitioning and target fabrication processes can be developed.
6. There may be incentives for iodine P-T if very conservative long-term risk analysis assumptions continue to be used and if a feasible method for transmuting iodine can be identified.
7. Sensitivity analyses indicate that the above conclusions concerning the incentives for P-T are valid for a wide range of input assumptions and parameters.
8. The incentives for P-T are virtually independent of the transmutation device used. Thus, the existence of advanced devices would not alter the incentives.

### 7.3 Research, Development, and Demonstration Required

1. Approximately 20 years would be required to bring the first commercial-sized partitioning facilities on-line, assuming a well-supported program and no licensing delays.
2. Approximately 15 years of intensive RD&D would be required at a cost of about \$900 million (1979 dollars). The RD&D needed for partitioning requires the majority of the monies and is the limiting aspect of the schedule.
3. As a result of the lack of incentives for actinide P-T, additional RD&D in support of P-T is not warranted.

### 8. REFERENCES

1. A. G. Croff, J. O. Blomeke, B. C. Finney, and D. W. Tedder, Actinide Partitioning-Transmutation Program Final Report. I. Overall Assessment, ORNL-5566 (in press).
2. A. G. Croff et al., A Preliminary Assessment of Partitioning and Transmutation as a Waste Management Concept, ORNL/TM-5808 (September 1977).
3. W. D. Bond and R. E. Leuze, Feasibility Studies of the Partitioning of Commercial High-Level Wastes Generated in Spent Nuclear Fuel Reprocessing: Annual Progress Report for FY-1974, ORNL-5012 (January 1975).
4. H. C. Burkholder, M. O. Cloninger, D. A. Baker, and G. Jansen, Incentives for Partitioning High-Level Waste, BNWL-1927 (November 1975).
5. D. W. Tedder, B. C. Finney, and J. O. Blomeke, Actinide Partitioning-Transmutation Program Final Report. II. Partitioning Processes for LWR Fuel Reprocessing and Refabrication Plant Wastes, ORNL/TM-6982 (in press).\*

---

\*The distribution of this report must be coordinated with the Director, Office of Nuclear Waste Management, U.S. Department of Energy, Washington, D.C. 20545, U.S.A.

6. J. W. Wachter and A. G. Croff, Actinide Partitioning-Transmutation Program Final Report. III. Transmutation Studies, ORNL/TM-6983 (in press).
7. C. W. Alexander and A. G. Croff, Actinide Partitioning-Transmutation Program Final Report. IV. Miscellaneous Aspects, ORNL/TM-6984 (in press).
8. A. E. Smith and D. F. Davis (R. M. Parsons Co.), Actinide Partitioning-Transmutation Program Final Report. V. Preconceptual Designs and Costs of Partitioning Facilities and Shipping Casks, ORNL/TM-6985 (in press).
9. R. R. Fullwood and R. R. Jackson (Science Applications, Inc.), Actinide Partitioning-Transmutation Program Final Report. VI. Short-Term Risk Analysis of Reprocessing, Refabrication, and Transportation, ORNL/TM-6986 (in press).
10. S. E. Logan et al. (Los Alamos Technical Associates), Actinide Partitioning-Transmutation Program Final Report. VII. Long-Term Risk Analysis of the Geologic Repository, ORNL/TM-6987 (in press).
11. National Academy of Sciences Advisory Committee on the Biological Effects of Ionizing Radiations, "The Effects on Populations of Exposure to Low-Levels of Ionizing Radiation," National Academy of Sciences-National Research Council, Washington, D.C. (1972).
12. Code of Federal Regulations, Title 10, Part 50, Appendix I (January 1978).
13. U.S. Department of Energy, Technology for Commercial Radioactive Waste Management, DOE/ET-0028 (May 1979).

## 9. Captions

Fig. 1. Schematic diagram of the reference and P-T fuel cycles. ORNL DWG 79-1686.

Fig. 2. Generic actinide partitioning flowsheet. ORNL DWG 79-20557R1.

Table 1. Actinides in wastes of reference and P-T cycles as a percentage of facility feed.

Table 2. Summary of the costs, risk, and benefits of P-T per GW(e)-y.