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ACTINIDE RECYCLE POTENTIAL IN THE INTEGRAL FAST REACTOR (IFR) FUEL CYCLE *

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ABSTRACT

In the Integral Fast Reactor (IFR) development program, the entire reactor system—reactor, fuel cycle, and waste process is being developed and optimized at the same time as a single integral entity. The use of metallic fuel in the IFR allows a radically improved fuel cycle technology. Pyroprocessing, which utilizes high temperatures and molten salt and molten metal solvents, can be advantageously utilized for processing metal fuels because the product is metal suitable for fabrication into new fuel elements. The key step in the IFR process is electrorefining, which provides for recovery of the valuable fuel constituents, uranium and plutonium, and for removal of fission products. In the electrorefining operation, uranium and plutonium are selectively transported from an anode to a cathode, leaving impurity elements, mainly fission products, either in the anode compartment or in a molten salt electrolyte. A notable feature of the IFR process is that the actinide elements accompany plutonium through the process. This results in a major advantage in the high-level waste management, because these actinides are automatically recycled back into the reactor for in-situ burning. Based on the recent IFR process development, a preliminary assessment has also been made to investigate the feasibility of further adapting the pyrochemical processes to directly extract actinides from LWR spent fuel. The results of this assessment indicate very promising potential and two most promising flowsheet options have been identified for further research and development. This paper also summarizes current thinking on the rationale for actinide recycle, its ramifications on the geologic repository and the current high-level waste management plans, and the necessary development programs.

I. BACKGROUND

A multitude of studies and assessments of actinide partitioning and transmutation were carried out in late 1970's and early 1990's. Probably the most comprehensive of these was a study coordinated by Oak Ridge National

Laboratory.¹ The conclusions of this study were that only rather weak economic and safety incentives existed for partitioning and transmuting the actinides for waste management purposes, due to the facts that (1) partitioning processes were complicated and expensive, and (2) the geologic repository was assumed to contain actinides for hundreds of thousands of years.

Much has changed in the few years since then. A variety of developments now combine to warrant a renewed assessment of the actinide recycle. First of all, it has become increasingly difficult to provide to all parties the necessary assurance that the repository will contain essentially all radioactive materials until they have decayed. Assurance can almost certainly be provided to regulatory agencies by sound technical arguments, but it is difficult to convince the general public that the behavior of wastes stored in the ground can be modeled and predicted for even a few thousand years. From this point of view alone there would seem to be a clear benefit in reducing the long-term toxicity of the high-level wastes placed in the repository.

Secondly, the Nuclear Waste Policy Act of 1982 mandated that (1) EPA promulgate standards for protection of the general environment from off-site releases from radioactive materials in repositories and (2) NRC promulgate technical requirements and criteria, consistent with EPA standards, that NRC will apply in licensing of repositories. The place of actinide recycle today, therefore, must be reevaluated in the light of the technical performance requirements placed on the repository by these newly established NRC regulations² and EPA standards.³

Finally, IFR pyroprocessing has been developed only in recent years and it appears to have potential as a relatively uncomplicated, effective actinide recovery process. In fact, actinide recycling occurs naturally in the IFR fuel cycle. Although still very much developmental, the entire IFR fuel cycle will be demonstrated on prototype-scale in conjunction with

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the EBR-II and its refurbished Fuel Cycle Facility starting in late 1991. A logical extension to this work, therefore, is to establish whether this IFR pyrochemical processing can be applied to extracting actinides from LWR spent fuel.

II. RATIONALE FOR ACTINIDE RECYCLE

The LWR spent fuel waste consists of both fission product and actinide elements. Fission products comprise hundreds of various isotopes, which, along with energy, are the products of fissioning. Actinides (or more precisely transuranic elements) are produced from neutron capture, as opposed to neutron fission, reactions in the fuel.

The relative radiological toxicities of actinides and fission products contained in once-through LWR spent fuel are compared in Fig. 1, normalized to the toxicity of the natural uranium ore. Figure 1 was derived in the following manner:

The International Commission on Radiological Protection (ICRP) Publication 30 gives estimates of the cancer risk to various body organs resulting from ingestion of a radionuclide.⁴ When this is multiplied by the number of curies of each isotope in the LWR spent fuel, the result is an estimate of the number of cancer doses from each radionuclide in the spent fuel. These cancer doses are then summed over radionuclides in two categories, fission products and actinides, for 1 metric ton of the spent fuel. This gives the total potential radiological hazard if all radionuclides in 1 metric ton of the spent fuel are ingested. This potential hazard is then normalized relative to the cancer hazard, calculated in the same manner, associated with natural uranium ore from which the spent fuel originated. In the natural uranium ore the uranium is assumed to be in equilibrium with its daughter products.

As shown in Fig. 1, most of fission products have relatively short half-lives, and their radiological toxicity drops below that of their original uranium ore in a timespan of the order of 200 years. Actinides, on the other hand, have long half-lives and their radiological toxicity remains orders of magnitude higher than that due to fission products for millions of years. In this lies the incentive to separate actinides from the waste stream and recycle them back into a reactor for in-situ burning, leaving a waste free of actinides.

It should be noted at this point, however, that Fig. 1 does not represent actual radiological risk from a repository, for example, because it does not take any account of pathways to the human body, and in effect assumes total release. The term "toxicity" is used in Fig. 1

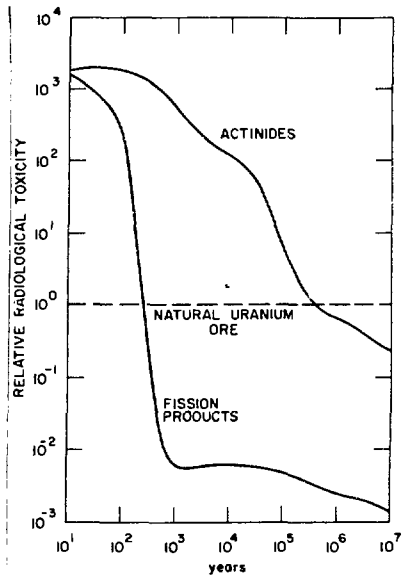


FIGURE 1. Relative Radiological Toxicity of Fission Products and Actinides Contained in the LWR Spent Fuel, Normalized to the Original Uranium Ore.

instead of "risk" because the word "risk" has an implicit connotation that the radioactive nuclides in the repository are released to the environment causing ingestion by humans and hence involves cancer risks. The term "toxicity" is meant to indicate the potential hazard contained in the materials in the repository, without addressing the issue of actual pathways from repository to human body.

Another, more concrete, way of evaluating the benefits of actinide recycle is in terms of the ability to satisfy the technical performance requirements placed on the geologic repository. The EPA standards establish containment requirements that cumulative releases of radionuclides to the accessible environment for 10,000 years after disposal have a likelihood of less than one chance in 10 of exceeding the quantities specified as "cumulative release limits."

A proposed NRC rulemaking involves direct incorporation of the EPA cumulative release limits within 10 CFR Part 60. Even before this direct incorporation, 10 CFR Part 60 requires the repository to conform to such generally applicable environmental standards for radioactivity as may have been established by EPA. The EPA cumulative release limit is therefore a very

important technical requirement placed on the geologic repository. The EPA cumulative release limits given in terms of curies per metric ton heavy metal (MTHM) of spent fuel are listed in Table 1.

The LWR spent fuel activities for various nuclides normalized to the EPA limits in Table 1 are summarized in Table 2. If this value is less than 1, it means that even if the entire inventory is released the EPA limit is not exceeded. If this value is 100, then only 1% of the inventory can be released before the limit is exceeded, and so on.

Following the 300-1000 years of containment period required by the NRC regulations, the long-lived fission products have decayed to the same magnitude as the cumulative release limit. However, the actinide (including plutonium) activity is about 10^4 times the cumulative release limit. The allowable release is 10^{-4} , or 0.01% over the 10,000-year period. Allowable annual release rates then would be 10^{-8} per year. This is a very stringent requirement to meet.

TABLE 1. Release Limits for Containment Requirements. (Cumulative releases to the accessible environment for 10,000 years after disposal)

Radionuclide	Release Limit per 1000 MTHM or Other Unit of Waste (Curies)
Americium-241 or -243	100
Carbon-14	100
Cesium-135 or -137	1000
Iodine-129	100
Neptunium-237	100
Plutonium-238, -239, -240, or -242	100
Radium-226	100
Strontium-90	1000
Technetium-99	10,000
Thorium-230 or -232	10
Tin-126	1000
Uranium-233, -234, -235, -236, or -238	100
Any other alpha-emitting radionuclide with a half-life greater than 20 years	100
Any other radionuclide with a half-life greater than 20 years that does not emit alpha particles	1000

If actinides are removed from the waste stream, then the EPA standards on cumulative release limits can be met more easily. Table 2 also indicates the desirable level of actinide decontamination. If 99.99% of actinides are removed from the spent fuel, then their activity will be in the order of unity after 1000 years. This is also consistent with the toxicity level considerations. Referring back to Fig. 1, if the actinides in the waste stream are reduced to 10^{-4} , their toxicity is in the same range as that of fission products.

III. IMPLICATIONS ON REPOSITORY

As discussed above, actinide recycle allows the technical performance requirements placed on the repository to be met more easily, and seems highly desirable from the point of view of public acceptance. Actinide recycle should make the entire geologic repository concept more viable. At the same time, however, actinide recycle is not a requirement for the high-level waste management, and actinide recycle does not replace the need for a geologic repository.

The geologic repository is needed, independent of actinide recycle, for the following reasons.

- The goal decontamination factor for the removal of actinides from the waste stream is 10^4 , that is, 99.99% removal of actinides. The residual 0.01% of actinides means waste that is far from being qualified as a non-TRU waste. A decontamination factor of the order of 10^6 would be needed for non-TRU qualification. Therefore, a geologic repository is needed to store even the residual actinides.
- In addition to residual actinides, there are a number of long-lived fission products, such as Tc-99, I-129, Cs-135, etc., that will always need to be stored.
- There are solidified defense high-level wastes and other civilian high-level wastes right now that need to be disposed in a geologic repository.

Therefore, actinide recycle must not be allowed to interfere with the present high-level waste management plans leading to the first geologic repository. Actinide recycle is a long-term option. It requires much further R&D to establish the technical, economic, and institutional practicality.

Although actinide recycle does not replace the need for a geologic repository, if successful it should simplify the long-term waste management strategy dealing with the need and timing of the second repository.

TABLE 2. LWR Spent Fuel Activity
(Normalized to the EPA "Cumulative Release Limit" Listed in TABLE 1)

	Activity at 10 yrs	Activity at 10 ³ yrs	Activity at 10 ⁴ yrs
Sr-90	60,000	0.0	0.0
Cs-137	90,000	0.0	0.0
I-129	0.3	0.3	0.3
Tc-99	1.4	1.4	1.4
Other Long-lived FP	1050	5.1	4.4
Actinides	76,000	19,000	4,000

The Nuclear Waste Policy Act (NWPA) of 1982 prohibits the emplacement of spent fuel in the first repository containing in excess of 70,000 metric tons of heavy metal until such time as a second repository is in operation. The intent was to prevent the first repository being the only one and hence expanding its capacity forever. However, if the radiological toxicity is reduced by orders of magnitude through actinide recycle, and the decay heat burden could be reduced through an interim storage of cesium and strontium, it is reasonable to suggest that the current NWPA be amended to allow more efficient use of the first repository capacity.

The characteristics of the geological formation in which the repository is to be located limits the quantity and configuration of the spent fuel or process wastes that are to be replaced therein. Analysis of the Yucca Mountain Site, for example, indicates that the maximum heat burden that the site can handle without damage to the rock (chemistry, structure, and mechanical strength) is about 57 kW/acre. For 10-year cooled spent fuel, the heat load considerations limit the boreholes to only 16 per acre. If the heat load is not a constraint, then more boreholes can be drilled. The structural considerations during borehole drilling then determine the maximum number of boreholes, which is conservatively estimated to be 130 per acre. This is a factor of 8 increase in the effective capacity. Furthermore, if heat load considerations were no longer limiting, the waste form could be compacted for each borehole to fairly easily gain a further factor of about 2.5. So the overall potential for the first repository capacity increase is of the order of a factor of 20, if heat loads were eliminated.

As shown in Table 3, the heat load is dominated by Cs and Sr in the short term, and by actinides in the long term. Therefore, if actinides are recycled and Cs and Sr are stored

in interim on the surface, the effective capacity of the first repository could be extended to the point where a second repository is not needed for a very long time. After their interim storage, the waste containing Cs and Sr can be put into the repository for permanent disposal.

Even after most of the long-lived actinides are separated from the waste stream, other long-lived fission products remain. A logical question is whether incentive exists to partition and transmute these fission products as well. As summarized in Table 2, the concentrations of these long-lived fission products are low and the activities are in the same range as the EPA cumulative release limits. Furthermore, their combined toxicity level is at least two orders of magnitude less than that of the natural uranium ore as shown earlier in Fig. 1. Therefore, there seems to be no strong incentive to partition and transmute these long-lived fission products.

However, it should also be noted that previous repository risk assessment has shown that Tc-99 and I-129 dominate the long-term risk. This is because during an assumed leach incident at the repository the models used for risk analysis predict that technetium and iodine migrate through the geosphere rapidly enough to reach the biosphere within one million years, while actinides are sorbed in the geosphere and do not reach biosphere within one million years. That is why the risk is dominated by Tc-99 (92%) and I-129 (8%). In perspective, however, the health effects of Tc-99 and I-129 that arise from a repository leach incident are about a factor of 20 less than the expected health effects from natural background.

IV. ACTINIDE RECYCLE IN THE IFR

A distinguishing element of the IFR concept is its unique fuel cycle based on metallic fuel

TABLE 3. Decay Heat of LWR Spent Fuel
(Watts/MTHM)

Year	Actinides	Sr and Cs	Others F.P.	Total
1	610	8270	3430	12310
5	280	1550	430	2260
10	280	940	80	1300
20	270	650	30	950
50	250	320	2	572
100	215	97	0	312
200	174	9	0	183
500	110	0	0	110

and pyroprocessing.⁵ As shown in Fig. 2, the IFR fuel cycle involves only a few steps. The key step in the IFR process is electrorefining. A cadmium pool in the bottom of the electrorefiner vessel serves as one electrode. The electrolyte is LiCl-KCl eutectic with about 2 mol % heavy metal chlorides, rare earth chlorides, and active metal (sodium, cesium, strontium) chlorides. Suitable electrodes for anodic dissolution of fuel and for product recovery are placed in the electrolyte.

Spent fuel pins are chopped and put in a basket for dissolution in the electrorefiner at 500°C. Cadmium chloride is then added to oxidize alkali, alkaline earth, and most rare earth metals to their chlorides, which become a part of the molten chloride electrolyte. Essentially pure uranium is electrotransported to a solid cathode and mixed U-Pu product is electrotransported to a liquid cadmium cathode. These cathodes are removed from the electrorefiner cell and retorted to vaporize the cadmium and any occluded salt and to consolidate the product by melting.

The chemistry of the pyroprocess is based on the relative ease of oxidation of the elements that make up the metal fuel. This is determined by the free energies of formation of chlorides of these elements. Alkali and alkaline earth metals are readily oxidized into the salt, and less easily oxidized noble elements remain as metals. The amount of oxidant can be adjusted so that the actinides and rare earths are found both as metals and in the salt, although actinides will mostly be found as metal and rare earths will mostly be chlorides. Thus, oxidation effects most of the separation of actinides from fission products.

Separate collection of the uranium product and the mixed actinide product is possible because uranium is slightly less easily oxidized than the other actinides, and because the

oxidation state is such that the salt contains a mixture of actinides. When electrotransport is used simultaneously to oxidize the metals to their chlorides at an anode and to reduce an equivalent amount of chloride to metal at a solid metal cathode, uranium is preferentially deposited and the other actinides are preferentially oxidized. The product is essentially pure uranium, contaminated with the salt.

When a crucible containing liquid cadmium is used as the cathode, uranium, plutonium, minor actinides, and some of rare earths are all stabilized as metals by interaction with the cadmium. The cadmium electrode product is a mixture of actinides with small amounts of rare earths. Thus, the recovery and recycle of the actinides occur naturally in the IFR pyroprocess.

The hard spectrum reactor characteristic of the IFR makes an ideal actinide burner. Of course, actinides in a given target element can be burned in any reactor type. However, large-scale actinide burning involving the cumulated actinides from the LWR spent fuel inventory cannot be done efficiently in the thermal spectrum reactor. Because of high capture to fission cross section ratios, substantial amounts of the actinides evolve into higher mass isotopes in the thermal spectrum reactor. This degrades reactivity of the actinide mixture. Therefore, if the reactor is fueled largely with actinides, there is a limit beyond which they will be unlikely to be placed in a thermal reactor because of reactivity poisoning. As shown in Table 4, after four cycle generations, the reactivity value of the actinide mixture has been reduced to only about 7% of standard enriched uranium fuel. This requires a 43.6% actinide content for the fuel to have equivalent reactivity value. By this time, less than 30% of the initial actinide have burned. Thermal reactors are not feasible as true actinide burners.

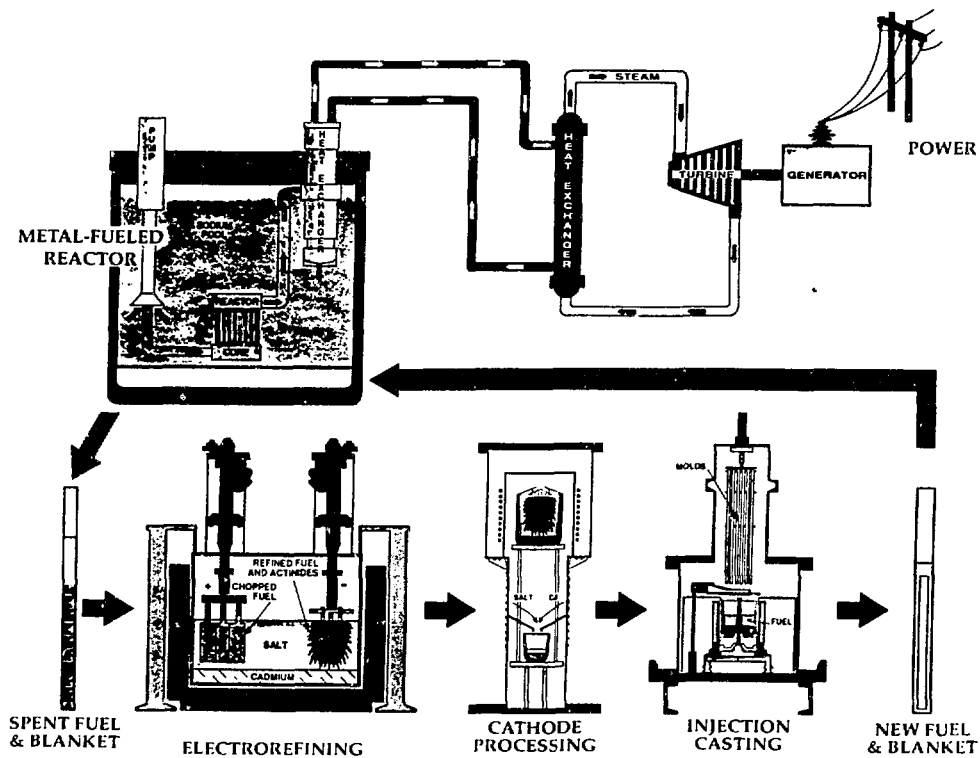


FIGURE 2. Schematic Illustration of the Integral Fast Reactor (IFR) Fuel Cycle.

TABLE 4. Actinide (Including Pu) Recycle in LWR

Recycle Generation	Actinide Reactivity Worth Relative to U-235	Actinide Loading Percent HM	Fraction of Initial Actinide Remaining
1	0.45	7.1	1.0
2	0.19	16.7	0.84
3	0.12	26.2	0.78
4	0.09	35.2	0.73
5	0.07	43.6	0.71

The LWR-generated actinides (plutonium, neptunium, americium, curium, etc.) can be used efficiently in the IFR spectrum because in this spectrum they have a considerable reactivity value. Further, because of the high fission to capture cross section ratios, they tend to burn rather than transmute and an equilibrium concentration of actinide is quickly established with continuous recycling. The IFR can be designed to operate in an actinide self-sustaining mode or as a net actinide burner.

The principal high-level wastes from the IFR pyroprocess fuel cycle are the metals and salts discharged from the electrorefiner. The metal waste consists of fuel cladding and perhaps a small amount of cadmium that is not recycled. This waste will also contain the noble metal fission products, most of the alloy zirconium, and small amounts of actinides. The electrorefiner salt will contain the halide, alkali-metal, alkaline earth and rare earth fission products along with some actinides. These waste streams are then converted to forms that are acceptable for disposal, as described below.

The salt waste is treated to reduce actinide content to less than 1 ppm by contact with a Li-Cd alloy, followed by filtration to remove insoluble impurities. After this treatment, the salt contains only the fission products cesium, strontium, and iodine. Its alpha activity will be below 100 nCi/g. The salt waste will then be immobilized in a suitable matrix and dispersed in metal or sintered ceramic matrix sealed in containers.

The metallic wastes from electrorefining will be combined with the Cd-Li used in treating the salt and the excess cadmium will be removed by retorting. The residue will be dispersed and immobilized in a corrosion-resistant metal matrix such as copper. This mixture will then be sealed in corrosion-resistant containers for disposal as high-level waste.

In the IFR process, cesium and strontium are already separated in the form of the salt waste, and therefore, there is no need to have additional separation processes developed if it were deemed desirable to implement the alternative waste management discussed earlier. The salt waste packages can be stored for an appropriate period to allow the decay heat reduction before they are permanently disposed in the geologic repository.

The status of the IFR process development has reached the point where a prototype demonstration of the entire IFR fuel cycle will be conducted in conjunction with EBR-II and its refurbished Fuel Cycle Facility, beginning in late 1991.

V. EXTENSION OF PYROPROCESSING TO LWR SPENT FUEL

IFR pyroprocessing appears to promise improvements in long-term waste management for the IFR itself. The next question that arises naturally is whether the approach can be extended to process LWR spent fuel. And, in fact, it turns out that there is an extensive experience base at Argonne in applying pyrochemical processes to oxide fuel.

In the late 1960's, the EBR-II fuel cycle facility operated for about five years using a simple drossing process, known as melt refining. In this process the electropositive fission products were separated from the fuel by reaction with a zirconium oxide crucible. The volatile elements were released and collected on a fume trap or condensed cryogenically from the cell atmosphere.

A pyrochemical process for recovering the actinides occluded with the dross, or crucible skull, was developed and demonstrated on an engineering scale with simulated fuel. This skull reclamation process employed liquid zinc-magnesium and molten chloride salt as process solvents. Also a blanket process for recovery of plutonium from metallic uranium blankets was demonstrated on a bench scale. This process involved the selective extraction of plutonium from molten uranium into an immiscible magnesium phase.

The techniques developed to process EBR-II skulls and blankets were then extended to processing uranium oxide and mixed oxide fast reactor fuels. Rapid methods for reducing these dense oxide fuels were demonstrated, and a liquid metal-fused salt extraction step was developed for isolating the uranium, plutonium, and fission products. This "salt transport process" was demonstrated on a laboratory scale, but funding terminated before a planned pilot plant demonstration could be completed.

The earlier pyrochemical process development efforts were discontinued because there was no clear advantage to producing a pure plutonium product stream over the traditional Purex process. Today, however, the processing goal has changed. In traditional reprocessing based on Purex, the goal was to produce a highly decontaminated, pure Pu product stream. However, when LWR processing is viewed as a waste management strategy, the goal is quite different. Neither a pure Pu product stream nor a high decontamination factor is required. In fact, just the opposite is desirable. The new process goals, when LWR spent fuel processing is viewed as a waste management strategy, are as follows:

- Direct extraction of all actinides (Pu, Np, Am, Cm, etc.) from the spent fuel as a single product stream.
- An actinide recovery target of 99.9% to 99.99%.
- The process should be incapable of producing pure Pu product.
- The process should be incapable of achieving a high decontamination factor for fission products.
- The process should be simple enough to achieve acceptable economics.

A preliminary assessment has been made to investigate the feasibility of using pyrochemical processes for directly extracting actinides from LWR spent fuel, satisfying the new process goals discussed above. It appears that the pyrochemical processes are exactly compatible with the new process goals and two promising flowsheet options have been identified: (1) a salt transport process and (2) a magnesium extraction process. These two processes are presented in Figs. 3 and 4.

The pyrochemical processes fit naturally to the LWR actinide extraction application and should provide significant advantages over the traditional Purex-based processes.

First, potentially all actinide elements are extracted in a single product stream, along with most rare earth fission products. A pure plutonium product is not possible. The product is highly radioactive and is not much more attractive than the original spent fuel as far as the diversion risk is concerned. The process as such therefore provides some nonproliferation protection.

Second, in these processes uranium remains as metal ingots with some noble fission product contamination. In this form the uranium can be easily stored for later recovery and use in the IFRs. The actinide extraction processes deal with only 1 or 2% of the total heavy metal. This small mass flow and the few process steps involved lead to compact equipment systems, small facility size, and portend favorable economics.

However, from the perspective of a U.S. utility, there is no immediate economic incentive in actinide recycling. With a 1 mill/kWh fee, the title to the spent fuel will be transferred to DOE for ultimate disposal. Initially, processing for actinide recycle will only add incremental cost. Thus the cost has to be kept to a minimum for the actinide recycle to be viable.

If actinide extraction is based on the traditional Purex-based technology, it appears

that even \$1000/kgHM processing cost is likely to be optimistic. But even this translates to 4.3 mills/kWh incremental cost to the LWRs. Accounting adjustments such as transferring this cost increment to the future LMRs is no solution because in the end utilities will operate both the LWRs and LMRs.

It is economically essential to develop a simple process that can extract actinides directly from the spent fuel. However, to be fair, the economic cost/benefit analysis for actinide recycle must be done for the entire system including the effects on the long term repository requirements.

VI. DEVELOPMENT NEEDS

An IFR economy can be justified and established without calling upon justification from LWR actinide recycle. However, LWR/IFR synergistic fuel cycle do provide advantages, and without the IFR fuel cycle, actinide recycling of the LWR spent fuel does not seem practical.

Specific policy decisions regarding LWR/IFR synergistic fuel cycle implementation are not possible at this time. The first repository is needed in any event. The present repository development program must continue and the IFR development program can proceed in parallel. IFR development and its associated actinide recycle technology must not be placed in the path of the repository program.

At the same time a good R&D program can be established now. It will provide the technical facts necessary to assess the practicality of actinide recycle and will be in time to allow timely policy decisions.

The IFR-based development program should have three major tasks:

- The program should begin on a practical LWR actinide extraction process, first developed, and then demonstrated.
- The presently planned IFR fuel cycle demonstration should be completed expeditiously as possible.
- Then an IFR demonstration project, including both the reactor plant and fuel cycle facility, should be put in place.

All three tasks must be completed successfully before the LWR/IFR synergistic fuel cycle can be implemented.

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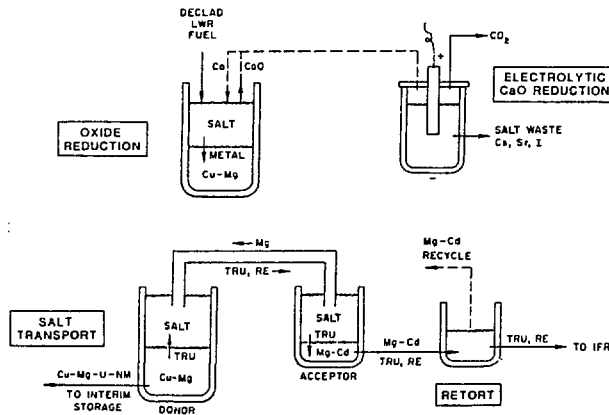


FIGURE 3. Salt Transport Conceptual Process for Extracting Actinides from LWR Spent Fuel.

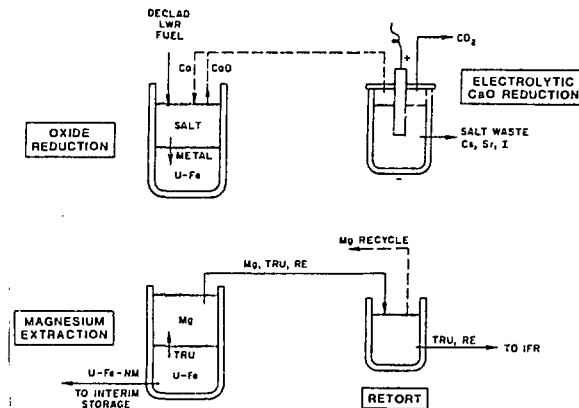


FIGURE 4. Magnesium Extraction Conceptual Process for Extracting Actinides from LWR Spent Fuel.