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# ACTINIDE CONSUMPTION: NUCLEAR RESOURCE CONSERVATION WITHOUT BREEDING

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## INTRODUCTION

For the past 40 years, nuclear power has been widely acknowledged as the essential basis for the energy economy of the 21st century. We are now at the point where investment decisions are 21st century decisions. The Department of Energy and industry groups have announced strategies for getting through the next decade, providing for a responsible entry into the 21st century. These analyses conclude that the next increment of nuclear power, based on Light Water Reactor (LWR) Technology, is economically justifiable[1]; there is ample uranium to support any realistic deployment[2], and massive programs are in place to address waste issues[3].

Looking beyond this decade, it is a short extrapolation to a time of concern both for resource availability and for waste accumulation. Chairman Seaborg's letter of November 20, 1962, transmitting the AEC's 1962 "Civilian Nuclear Power: a Report to the President"[4] is revealing in this regard:

> While the [AEC] has been proceeding on a considered course in general accord with its 10-year civilian power program adopted in 1958, that program is now on the threshold of attaining its primary objective of competitive nuclear power in high-fuel-cost areas by 1968. However, it became evident with the passage of time that our attention had probably for too long remained focused narrowly on short-term objectives. This restudy made it apparent that, for the long-term benefit of the country, and indeed of the whole world, it was time we placed relatively more emphasis on the longer-range and more difficult problem of breeder reactors, which can make use of nearly all of our uranium and thorium reserves, instead of the less than one per cent of the uranium and very little of the thorium utilized in the present types of reactors. Only by the use of breeders would we really solve the problem of adequate energy supply for future generations.

The logic of this approach was adopted as the basis of nuclear power development not only in the U.S., but in essentially every industrialized country. Since 1982, however, three major concerns have combined to frustrate breeder development: proliferation concerns, safety doubts, and capital costs of the reactor and associated fuel cycle. Perhaps the most insidious concern over the breeder is precisely that it will free nuclear power from any resource constraints and this would presumably would lead to an ever increasing nuclear waste problem.

A new approach, based on a metallic fast reactor fuel and pyrometallurgical processing of spent fuel is showing great potential and is approaching a critical demonstration phase. If successful, this approach will complement and validate the LWR reactor systems and the attendant infrastructure (including repository development) and will alleviate the dominant concerns over the acceptability of nuclear power.

## WHAT IS THE IFR CONCEPT

The Integral Fast Reactor (IFR) concept is a metal-fueled, sodium-cooled pool-type fast reactor supported by a pyrometallurgical reprocessing system[5]. The concept of a sodium cooled fast reactor is broadly demonstrated by the EBR-II[6] and FFTF[7] in the U.S.; DFR and PFR[8] in the UK; Phenix[9] and SuperPhenix[10] in France; BOR-60[11], BN-350[12], BN-600[13] in the USSR; and JOYO[14] in Japan. The metallic fuel is an evolution from early EBR-II fuels[5]. This fuel, a ternary U-Pu-Zr alloy, has been demonstrated to be highly reliable and fault tolerant even at very high burnups (160-180,000 MWd/MT)[15]. The fuel, coupled with the pool type reactor configuration, has been shown to have outstanding safety characteristics[16]: even with all active safety systems disabled, such a reactor can survive a loss of coolant flow, a loss of heat sink, or other major accidents. Design studies based on a small modular approach[17] show not only its impressive safety characteristics, but are projected to be economically competitive[18].

A distinguishing element of the IFR concept is its fuel cycle based on metallic fuel and pyroprocessing[19]. The key step in the IFR process is electrorefining, where the actinides from core and blanket fuels are recovered and separated from fission products. The electrorefiner device[20,21] is a steel crucible containing a pool of cadmium metal at 500°C overlain with a liquid mixture of chloride salts, primarily LiCl-50 wt % KCl-6% actinide chlorides. A commercial-scale electrorefiner would be about 1 m in diameter and would contain 1000 kg cadmium and 500 kg salt. The chopped core fuel is immersed in the salt and made anodic with respect to the cadmium pool. After anodic disolution and electrotransporting a portion of the uranium from the core elements onto a solid cathode, a ceramic crucible containing a separate and smaller cadmium pool is immersed in the electrorefiner salt as the cathode; the actinides are then electrotransported to the small cadmium cathode. A fraction of the rare earths necessarily accompany the TRU elements;

the amounts (estimated DF is >10) will not affect fuel performance or reactor operation, but will provide a high degree of safeguards protection. The noble metal fission products and Zr are not involved in the electroprocess, but remain as metal, free to settle as particulates into the lower cadmium pool. The alkali metal, alkaline earth, and rare earth fission products are dissolved in the salt as chlorides. The cladding hulls are removed from the electrorefiner and become a waste. The inseparable ensemble of TRU elements and uranium from the cadmium cathode is recovered by distilling of the cadmium and is used to make core fuel alloy.

Blanket elements are handled in the same way, except that uranium is extracted by using a solid steel cathodic collector immersed in the salt. The dendritic uranium deposit is mechanically stripped from the cathode and melted under vacuum to remove adhering salt and to prepare a uranium ingot for blanket fuel fabrication. The transuranium elements are allowed to accumulate as several batches of blanket fuel are processed and are then recovered in a cadmium cathode as discussed above. While the uranium deposit can be kept reasonably free of TRU carryover and tramp fission products, no adjustment of the chemistry of the process will lead to an effective separation among the transuranium elements, nor is a TRU product without significant uranium and fission product carryover feasible. Thus, the process is effectively as proliferation resistant as is spent reactor fuel. In fact it can be argued that returning plutonium to the fuel cycle precludes the safeguards threat that would be posed by 200 year cooled spent LWR fuel.

An IFR fuel cycle facility should have low overall release of radioactivity, because of the capability of containing all fission products, including those that are difficult to manage in conventional fuel recovery processes[3]. The fission gases, Xe, Kr, and T, are released when the fuel is chopped and then dissolved in the electrorefiner. Tritium is trapped by a purification system which converts oxygen to water and collects the water on molecular sieves. Because the volume of this water is small (a few hundred liters per year for reprocessing the fuel from a 1 GWe IFR), it can be stored until the tritium has decayed sufficiently to be released. Krypton and xenon can be trapped and concentrated by conventional cryogenic distillation methods. Iodine and C-14 are not expected to be volatilized in the pyroprocess; iodine will be contained in the salt as stable iodides, e.g., KI, and carbon will form stable carbides, e.g., ZrC, that should be retained in the metal wastes.

The principal wastes from the IFR pyroprocess fuel cycle are the metals and salts discharged from the electrorefiner. The metal waste consists of fuel cladding and a small amount of cadmium that is not recycled. This waste will also contain the noble metal fission products, a part of the zirconium from the fuel alloy, and a small fraction (estimated to be 1 part in $10^4$) of the actinides.

The electrorefiner salt as discharged from the electrorefiner will contain the halide, alkali metal, alkaline earth and rare earth fission products along with a minor amount of actinides. The salt waste is first treated in a molten salt-metal extraction process to recover more than 99% of the residual TRU elements, and then it is contacted with a Cd-Li alloy to remove any remaining actinides and the rare earths[22]; filtration is used 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; the current reference approach would be to trap the radioisotopes in a suitable matrix by ion exchange and then to disperse this ion exchange medium into an appropriate metal or sintered ceramic matrix, which would in turn be sealed in containers.

The metallic wastes from electrorefining will be combined with the Li-Cd 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 (depending on the intended repository chemistry). This mixture will then be sealed in corrosion-resistant containers for disposal as high-level waste.

It will be noted that in the IFR process, cesium and strontium are naturally separated in the form of the salt waste, and therefore, there would be no need to have an additional separation process developed if it were deemed desirable to store the salt waste packages for an appropriate period to allow decay heat reduction before disposal in a geologic repository. I-129 is the only long lived fission product that would be expected to be found in this waste stream.

With the projected low TRU carryover, it can be readily shown[23] that the long term repository requirements will not be controlled by the actinide content[24]. The dominant long lived fission products (Tc-99, C-14 and Sn-126) from the IFR pyroprocessing are in waste metal alloy, which can be further alloyed to meet repository requirements[25]. Thus, not only total actinide recycle but waste form segregation follows naturally from the IFR process.

In the hard spectrum of the IFR, all of the transuranium elements (Pu, Np, Am and Cm) are quality fuels and all isotopes contribute (as contrasted with a thermal spectrum, where even-mass isotopes build up to poison the nuclear reaction). Thus, the IFR system, by naturally recycling all the TRU elements and burning them as fuel, can extract essentially all of the energy resource from the initial feed. This is in contrast with the once-through LWR fuel cycle which extracts only 0.5 to 1 percent of the energy content of the mined uranium.

By straightforward adjustments in fuel composition and arrangement, the system can be readily adjusted to meet any overall fissile demand scenario, from being a rapid consumer of fissile material (conversion ratio as low as about 0.6) to a net producer (breeding ratio as high as about 1.3). The current reference is a self-sustaining fuel cycle, where the feed is natural or depleted uranium and the output is energy.

This approach is fundamentally different from the traditional partitioning and recycling techniques[26] in three regards:

- The process naturally recovers all of the transuranium elements.
- The process never involves separated plutonium.
- The process is basically simple and modular, permitting great flexibility and (presumably) economy.

Thus, while extensive reviews concluded that partitioning and transmutation could not be justified as a waste management strategy[27], those conclusions do not necessarily apply to pyroprocessing.

## HOW DOES THE IFR INTERFACE WITH THE LWR

There is an opportunity, but not a requirement, to couple the IFR fuel cycle with the LWR. The once through fuel cycle for the LWR leaves a residue of several hundred grams of plutonium for every kilogram fissioned. While the IFR can bootstrap itself from an enriched uranium start, resource considerations favor utilizing the residual plutonium and other TRU elements from LWR spent fuel or other sources.

A logical extension to the pyrometallurgical processing of IFR fuel is to establish whether a similar pyroprocessing concept can be applied to the extraction of actinides from spent LWR fuel. The processes being considered are essentially head-end processes to convert the LWR oxide fuel to metal and then to separate the TRU and rare earth elements from the bulk uranium, which would be stored for later use in IFRs. The product TRU plus rare earths would be fed directly into the IFR fuel cycle; no new types of waste streams are introduced. Based on initial laboratory scale work, two promising pyrochemical processes have been identified, and R&D to establish engineering feasibility has been initiated. If successful, this technology will permit a full integration of the LWR and IFR fuel cycles, with the LWRs providing fuel to the IFR and the IFR consuming the residual actinides. This would then represent the true symbiosis, where the wastes from the LWR are the catalyst for an energy supply that is free from resource constraints and which leaves a residue that after a few hundred years is less toxic than the ore from which the fuel cycle was started.

While highly desirable, this coupling to the LWR is not essential to the IFR concept. Having a portion of the nuclear energy supply base operating at resource efficiency approaching 100% is obviously preferable to having it all operate at 1/2 to 1% efficiency. For startup fuel, there are already substantial plutonium backlogs available in foreign stockpiles (the UK, for example, already has a utility stockpile of 30 MT of Pu from their civilian gas-cooled reactor program) and there is a very large weapons stockpile (IAEA has estimated that there are several hundred tons of weapons Pu) which may some day be surplus. Thus, existing inventories can support the startup of a reasonable fast reactor program.

## HOW PRACTICABLE IS THE IFR

The IFR is a proven reactor concept. The General Electric Company is well along in establishing licensing feasibility, and fundamental discussions on size optimization, optimization of safety parameters, and control concepts are progressing[17]. Fuel proof-of-principle testing, both for endurance and for fault tolerance, is complete; a simple fuel fabrication technique (injection casting) process has been used for many years at EBR-II using reconstituted irradiated fuel; efforts are now concentrating on statistical data accumulation and data required for manufacturing specifications and the licensing process. The production of uranium from simulated blanket fuel by electrorefining has been demonstrated at production scale without fission products. Laboratory scale plutonium separations have shown the feasibility of this part of the process. The fuel cycle, involving multiple recycle and equilibrium fission products will be demonstrated in a refurbished, existing facility at EBR-II. This process will be extended to recycle of target burnup (up to 20%), multiple recycle, and mixed actinide fuels. Barring technical setbacks, this will demonstrate the entire fuel cycle at a sufficient scale to permit confident estimation of commercial fuel cycle cost by 1995.

Parallel work under DOE's ALMR development program will have established by 1995 concept licensability for the reactor and its fuel cycle. This work will also have developed a demonstration plant design in sufficient detail to permit a rational commitment to construction.

The program to explore the feasibility of actinide recovery from spent LWR fuel is in its initial phase, but it is expected that technical feasibility could be demonstrated by about 1995; DOE has not yet committed funds to achieve this objective.

## WHAT CONCERNS WILL THE IFR ADDRESS?

There are at least six components to the justification for this new Technology:

- Reactor Safety: The reactor component of the IFR has demonstrated outstanding safety characteristics.
- Design and Process Simplification: The reactor safety features permit considerable design simplifications and the fuel cycle processes are fundamentally simpler than the traditional PUREX process; thus there should be significant savings in both capital and operational costs.

- Energy Resource Utilization: The process permits full utilization of the uranium resource involved by recovery of all of the transuranium and of the unburned uranium. Coupled with actinide recovery from LWR fuel, the IFR also provides for total resource recovery from LWR spent fuel without the need of the traditional breeder fuel cycle.

- Proliferation (Diversion) Resistance: The process is not compatible with obtaining weapons usable separated plutonium and is thus resistant to proliferation.

- Simplify Long-half-life Waste Disposal: The actinide content in the waste streams is sufficiently low that actinides do not dominate waste disposal considerations.

- Minimize "Mill Tailings" Problem: By more efficient uranium utilization, the need for uranium mining is drastically reduced along with the problem of mill tailings and release of radon to environment.

No one of these factors in isolation is sufficient to justify a near term investment of the magnitude necessary to demonstrate and commercialize these concepts. But in combination, there is again the possibility to fulfill the promise of nuclear power to "solve the problem of adequate energy supply for future generations."[4]

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