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THE IFR MODERN NUCLEAR FUEL CYCLE

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ABSTRACT

Nuclear power is an essential component of the world's energy supply. The IFR program, by returning to fundamentals, offers a fresh approach to closing the nuclear fuel cycle. This closed fuel cycle represents the ultimate in efficient resource utilization and environmental accountability.

INTRODUCTION

After nearly 40 years of design and operation, civilian nuclear power plants have settled into a few, well developed basic design concepts. The LWR has firmly established itself as a major contributor to the economic viability of no less than 16 nations around the world.⁽¹⁾

In general, these plants are operating on a once-through fuel cycle. Uranium is mined, processed, enriched by gaseous diffusion or centrifuges, fabricated, some 3% of the fuel is fissioned to produce heat, and the remainder set aside for later disposal. Considering the enrichment tails, this sequence results in less than 1% fuel utilization; the spent fuel presents challenges to those charged with assuring safe disposal. By plutonium recovery and recycle in thermal reactors, the effective resource utilization can be increased; perhaps doubled. Waste forms specifically engineered to retain fission products can be used. None of these processes have the potential that was envisioned for the closed fuel cycle of the breeder, which promised a 60 fold increase in fuel utilization.

A variety of fast reactors around the world (EBR-II,⁽²⁾ FFTF,⁽³⁾ PFR,⁽⁴⁾ Phenix,⁽⁵⁾ SuperPhenix,⁽⁶⁾ BN-350,⁽⁷⁾ BN-600⁽⁸⁾) have demonstrated that liquid metal cooled breeder reactor plants can operate effectively in utility environments. These plants have also demonstrated the extraordinary tolerance of this type of system to upsets and accidents. Closing the fuel cycle so as to achieve the fuel utilization inherent in the breeder has not kept pace.

A fresh look at closing the fuel cycle has opened exciting possibilities which go far beyond even the dramatic promise of the breeder: the utilization of effectively all of the energy potential of the ore

mined. If the fuel is fully recycled, it means that essentially no actinides remain to be disposed of in the high level wastes. Thus, to the extent that this approach can be realized, it represents the epitome of resource utilization and pollution prevention.

The IFR development was initiated as a means of capitalizing on the outstanding safety characteristics available with a metal fueled, sodium cooled fast reactor.⁽⁹⁾ To support this approach, it was necessary to demonstrate substantially improved nuclear fuel performance. These objectives (demonstrated safety characteristics and outstanding fuel performance) have been met. The metallic fuel also permits a simple and direct pyrometallurgical processing and recycle of actinides. The basic laboratory work to support demonstration of this recycle technology is largely complete; equipment is being fabricated, and within a year, the demonstration of the closure of nuclear fuel cycle will be underway.

WHAT IS THE IFR CONCEPT?

The initials IFR stand for Integral Fast Reactor, the name chosen for a new advanced reactor system that is being developed at Argonne National Laboratory. The word "Integral" is used to indicate that all the elements required to close the nuclear fuel cycle are being developed at the same time as an integral part of the system. That is, not only the reactor concept itself, but the entire fuel cycle is being developed, including systems for processing wastes. This is a generic technology in the same sense that the LWR is a generic reactor technology. The GE PRISM reactor concept⁽¹⁰⁾ is a particular realization of the reactor aspects of the IFR system.

The essential reactor characteristics are a metallic fuel alloy and sodium coolant in a pool type configuration. The particular fuel alloy is uranium, a recycle mixture of plutonium and higher actinides, approximately 10% Zr to assure an elevated melting point, and some tramp carryover of lanthanide fission products. The total transuranic content is adjusted for reactivity and breeding objectives.

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Metallc fuel has several unique characteristics. With the very high thermal conductivity, there is little temperature drop across the fuel; thus, there is essentially no stored energy. Metallc fuel swells under irradiation; this limited the lifetime for early metallc fuels, in that the swelling would cause unacceptable strain in the cladding.⁽¹¹⁾ However, by providing adequate room for swelling and a sodium bond to preserve conductivity, the fuel swelling is accommodated. After 1 to 2% burnup, fuel which originally occupied 75% of the available fuel cross section area will swell to bind with the clad; such fuel is too weak to strain the clad, and the voids interconnect to release most of the fission gas to a plenum.⁽¹²⁾ Such a fuel element is limited in burnup not by the fuel, nor by reactivity in a breeder, but ultimately only by irradiation damage to the clad.

Table I is a summary of fuel irradiation data on IFR type fuels as of early 1991. This data covers a range of Pu contents from zero to 26 w/o Pu; includes SS-316, D9, and HT-9 cladding. Except for a single pin deliberately operated at excessive temperatures, and several pins which have been deliberately defected, there have been no fuel failures. The EBR-II reactor is now fully fuelled with IFR type fuel. A number of assemblies have been or are being irradiated in FFTF. Considering the variety of parameters (including clad and Pu content), it can be concluded that this fuel type can be operated reliably to burnup well beyond the reference 10% core average burnup.

TABLE I. IFR Fuel Performance

Total Pins Irradiated or Under Irradiation	3000
Pins Greater than 10% Burnup	600
Pu Pins Irradiated or Under Irradiation	400
Pu Pins Greater than 10% Burnup	150

The limiting behavior of metallc fuels under severe accident conditions is also being investigated.⁽¹³⁾ In TREAT tests, IFR type fuel has been shown to withstand rapid transients to as much as 4 times normal power.⁽¹⁴⁾ In a specially designed test apparatus, irradiated fuel pins have been shown to survive over extended periods at up to 800°C⁽¹⁵⁾ (normal peak clad temperatures is less than 600°C). Deliberately defected fuel has been operated for over 200 days beyond cladding breach without problems; there is neither fuel erosion by the coolant nor enlargement of the failure as is characteristic with failed oxide fuels.⁽¹⁶⁾ Substantial progress has been made in modelling off-normal fuel and clad performance, so as to permit confident design. Combined with the self regulating characteristics and tolerance to insults available with an IFR core, the available data demonstrates the considerable design margins that are available to the designer.

The sodium coolant in a pool configuration complements the metal fuel and can provide an extraordinary tolerance for off normal and accident conditions.⁽¹⁷⁾ Sodium coolant has well known safety advantages, including high thermal conductivity, large margin to boiling, and an atmospheric pressure system (thus, no blowdown accidents). The large pool concept provides these plants with high thermal inertia, assuring that there is ample time for response to off-normal and accident conditions. Natural convection cooling at significant decay heat levels is readily achieved. Because of the high conductivity of the fuel,

there is little stored energy in the fuel. The low stored energy, coupled with the high margin to boiling, assures that natural convection will readily remove decay heat. In fact, for a typical core geometry, the stored heat energy is less than the energy necessary to raise the local sodium volume associated with any pin to its boiling temperatures.

These factors (large margins, high thermal inertia, and little stored energy), combined with a prudent design to emphasize reactivity feedback coefficients, lead to a reactor with outstanding stability and tolerance. This was dramatically illustrated in 1986 when the EBR-II was deliberately subjected to loss-of-flow and loss-of-heat sink transients with all engineered safety system including the SCRAM system disabled. The reactor passively terminated both of these events without challenging fuel or systems integrity. (These experiments are described in more detail in Ref. 18).

These safety and response characteristics offer the designer the potential of dramatic system simplifications relative to current generation reactors without compromise to overall safety. That is, it is not necessary to include emergency core cooling systems; emergency power systems are vastly simplified; decay heat removal is simpler; there is no potential for pipe whip accidents. Further, these can be near zero release plants (during 27 years of operation, there has been no make-up sodium added to the primary coolant system of EBR-II) with essentially no need for a liquid rad-waste treatment system. In addition to the capital cost advantages,⁽¹⁹⁾ these dramatic simplifications offer the potential of significant O&M savings. These systems also tend to be very clean as far as systems radioactive contamination; this is illustrated in Table II which lists personnel exposures for several sodium cooled power reactors as compared to LWR targets exposure levels.

TABLE II. Total Annual Dose Commitment

EBR-II (1970-1987)	14 ManRem
(1980-1987)	10
Phenix (16 year average)	8
(excluding major repairs)	5
Superphenix	<2
PWR (INPO 1990 goal)	288
BWR (INPO 1990 goal)	469

ACTINIDE RECYCLE

Spectacular as these advantages may be, they may be insufficient to justify commercial investment in a new technology. The unique potential of the IFR is in effectively closing the fuel cycle. This process is fundamentally different from the traditional PUREX type processing which was developed specifically to produce pure plutonium for weapons use. The IFR process has been selected on the basis of modern fuel utilization criteria.

The breeder fuel cycle was conceived as a means of extending our energy resource base by utilizing the bulk energy potential of natural uranium by converting U-238 to Pu-239. The liquid metal fast breeder reactor was adopted as a national priority at a time when the energy content of total known and reasonably assured uranium reserves, if used in LWRs, were comparable with petroleum reserves. In 1962, it was projected that 734 GW of nuclear capacity would be required by the year

2000⁽²⁰⁾ to meet projected electric demands. By 1967, orders for nuclear plants were progressing at a rate greater than projected in 1962, but no changes had been made in fossil or nuclear fuel resource estimates⁽²¹⁾. Impending exhaustion of low and moderate cost uranium demanded a rapid conversion from LWRs to LMFBRs, with the rate of introduction of LMFBRs being limited by available plutonium. By 1969 projections were for several hundreds of GW of breeders on line by the year 2000⁽²²⁾ with a substantial premium being justified by high breeding gain and rapid recycle.

The world looks different today. Massive uranium finds,⁽²³⁾ drastically altered growth and demand patterns⁽²⁴⁾ and technology deferrals leave us with very different criteria. There is still a projected exhaustion of uranium reserves, but it is not imminent; nuclear power does not enjoy an unquestioned cost advantage over all alternatives as it did in the late 60's; and there is a massive inventory of plutonium in spent fuel and in weapons stockpiles. Even though plutonium recovery technology is expensive and feared by some because of proliferation concerns, it is generally assumed that there are no constraints on plutonium availability. Thus, a high breeding gain is not necessarily an advantage - we may even wish to have plutonium burners. High specific power to limit inventory charges are far less important than capital costs.

By the late 1960s fuel recovery and recycle was considered to be an established technology and "the required fuel cycle capabilities should be well established when the LMFBR is introduced on a commercial basis in the 1980s,"⁽²²⁾ without further fundamental R&D. It was recognized that waste technologies would need to be developed, but in the 1967 Report to the President⁽²¹⁾ it is stated that "solidification and disposal technology for high-activity waste has reached the pilot plant and field demonstration phase," and "waste management costs will be a minor fraction of nuclear power costs and should not deter the development of safe and economical nuclear power".

These conclusions have been overtaken by new priorities.⁽²⁵⁾ Proliferation concerns, more exhaustive safety and radiation protection criteria, and demands for high assurance of minimal risk are now matters of highest priority. For fuel processing today, there is a priority for not producing a high purity product, so as to reduce proliferation concerns, and a very high priority on reducing the hazard content of the waste stream.

The technical feasibility of operating a thermal reactor on recycled fuel has been demonstrated, but the thermal neutron spectrum rapidly degrades the reactivity worth of the transuranium elements, making continued recycle impractical. In a fast reactor, all transuranium elements are high quality fuels. Many studies and assessments of actinide partitioning and transmutation were carried out in the late 1970's and early 1980's. An extensive bibliography of studies is contained in Reference 26. In these studies, the partitioning process (based on PUREX reprocessing) was considered to be complicated and expensive, and the geological repositories were presumed to prevent the release of actinides for hundreds of thousands of years. As a result, it was concluded that waste management considerations alone would not provide a sufficient economic and safety incentive for partitioning and transmuting wastes⁽²⁷⁾.

PYROPROCESSING

The development of pyroprocessing technology has led to renewed interest in actinide recycle.⁽²⁸⁾ A

unique aspect of this process is that it necessarily recovers an ensemble of all transuranic elements: plutonium, neptunium, americium, and curium. It is also not a "clean" process, in that there is necessarily a substantial uranium content and a partial carry-over of rare earth fission products in the product. The product is both highly radioactive and is unsuitable as a weapons material. Thus, the proliferation and diversion concerns associated with a commerce in separated plutonium do not arise. Further, techniques are available to strip the waste streams of their transuranics, greatly simplifying high level waste considerations. Being a much simpler process than PUREX, it is expected that the process will be substantially less costly than PUREX.

The key step in the process is electrorefining, where the actinides from core and blanket fuels are recovered and separated from fission products. The electrorefiner device^(29,30) being assembled for an integral demonstration of this process is a steel crucible containing a pool of cadmium metal at 500°C overlain with a liquid mixture of chloride salts, primarily LiCl-KCl. The demonstration electrorefiner is about 1 m in diameter and will 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 dissolution 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 unavoidable 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 first by using a solid steel cathodic collector immersed in the salt. Since the actinides on the cathode are fully exposed to the salt, uranium will be replaced by collected transuranics until the uranium concentration is reduced to a level comparable with that of the other actinides. 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.

This demonstration system is being installed in the Fuel Cycle Facility built for the EBR-II facility in Idaho. It will be used for a combination of development

and commercial scale demonstration. The equipment size is comparable with that which would be used in a production line supporting 400-500 MWe of power generation. The development program is scheduled to produce by 1995 a complete demonstration of the technical and economic feasibility of the entire IFR fuel cycle, including operation with high recycle fuel.

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 complete fuel cycle, involving multiple recycle and equilibrium fission products, will be demonstrated in the refurbished, Fuel Cycle Facility at EBR-II. This process will be extended to recycle of target burnup (up to 15%), 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.⁽³¹⁾

IFR WASTE STREAMS

The principal wastes from the IFR pyroprocess fuel cycle are metals and salts which are discharged from the electrorefiner when they are fully loaded with fission products. 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 10⁴) 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⁽³²⁾; 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; 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. Iodine-129 and Cesium-135 are the only long lived fission products that would be expected to be found in this waste stream. Without predisposal cooling of the Cs and Sr waste streams, it is evident that the repository physical capacity, which is ultimately determined by thermal considerations, will have only a modest advantage over alternate disposal waste forms, but will still enjoy the large reduction in long term hazard index.

With the projected low TRU carryover, it can be readily shown⁽³²⁾ that the long term repository requirements will not be controlled by the actinide content. 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. Thus, not only total actinide recycle but waste form segregation follows naturally from the IFR process.

Current repository analyses⁽³⁴⁾ show that actinides should be retained for the required 10,000 to 100,000 years under specified geologic conditions; programs are planned to characterize particular geologies. What actinide recovery and recycle offers is an easing of the burden of proof for a given repository.⁽³⁵⁾ It is still anticipated that a geologic repository will be required for the residual long lived fission products and the minor transuranic residence. The current repository program is also necessary for defense wastes, LWR waste not ultimately reserved for actinide recovery, and other high level wastes.

Two other waste considerations should be reiterated: the alloy waste form for noble metal fission products appears to be adaptable to anticipated repository conditions and the natural segregation of Cs and Sr permits consideration of engineered interim decay of these materials prior to disposal. The combination of segregation of the major decay heat load and elimination of almost all of the long term hazard will permit much greater flexibility and confidence in the use of geologic repositories.

LWR ACTINIDE RECOVERY

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. Just the PUREX process appears to be an expensive means of plutonium recovery, electrochemical processes do not appear to be well suited to the extraction of transuranics from spent LWR fuel, where the typical transuranic content is ~1%. 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 natural uranium.

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). The process can also be started from enriched uranium. Thus, existing inventories can support the startup of a reasonable fast reactor program.

CONCLUSIONS

The IFR concept has successfully built on a long heritage of fast reactor developments, to produce a technology which will close the nuclear fuel cycle. Not only will this fulfill the original promise of nuclear power of an inexhaustible energy resource, but the IFR is compatible with the highest standards of industrial and public safety and of environmental responsibility. The reactor technology is sufficiently advanced to permit the near term design and construction of a demonstration plant once high-recycle fuel demonstrations are available (about 1995). In that same time frame, the demonstration of the recycling of this fuel will be accomplished, using equipment of near commercial scale. Thus, the technology necessary to permit the design and construction of a prototype recycle facility will be available.

Programs are underway to verify the waste processing technology and to obtain certified waste forms. Parallel programs have been initiated to assess the practicability of pyrochemical recovery of actinides from spent LWR fuel.

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