FAST REACTOR FUEL PROCESSING

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INTRODUCTION

NOTICE

This paper is a status report on work now in progress on developing chemical processing and recycle fuel fabrication technology for liquid metal fast breeder reactor (LMFBR) fuel. With respect to chemical processing, there will be significant differences between the future LMFBR spent fuels and those from present light water reactors operating in the United States. These differences pose problems concerned mainly with the higher plutonium content and heat output from the expected LMFBR fuels.

It is natural to place major emphasis on the application of the Purex process for LMFBR fuel since nearly every major nuclear fuel processing facility now uses it. However, because of the differences mentioned above, research and development are necessary to make this approach economical for LMFBR fuel. For example, methods must be developed for removing and deactivating sodium; and advanced handling techniques are necessary to provide reliable cooling of the spent fuel elements until they are dissolved. The higher plutonium content provides a large economic incentive for processing at short decay times. However, this increases the problems of handling and containing shorter-lived fission products. The more volatile fission products, iodine, xenon, krypton, and tritium, are especially troublesome. The studies reported include development of reliable methods for cooling the spent fuel during shipping and handling, head-end processes for preparing the fuel for dissolution, dissolution methods, and solvent extraction technology. In the United States, particular emphasis is being placed on developing methods for reducing routine radioactive effluents from fuel processing plants to near zero.

With respect to fabrication of recycle fuel, the photon and neutron radiation from plutonium will require substantial shielding in commercial-scale fuel fabrication, and automation should be used. Methods for commercial fabrication and refabrication of fuel must be based on assessment of the relative performance of alternative fuel structures and the economics of the fabrication processes that produce those structures. Thus, irradiation data and process costs for mechanically blended pellets, coprecipitated pellets, and Sphere-Pac materials should be compared. At present, considerable data are available for the first two routes from the standpoints of processing costs and fuel performance; however, the Sphere-Pac process has had only limited application and the irradiation results for such material, while promising, are not comprehensive. It should be particularly noted that close attention should be given to methods by which process cost can be reduced by selection of a different fuel design. In addition, improvements in the economical production of LMFBR fuel tubing would have a significant impact on future costs, as would the development of fuel structure and a design that would permit attainment of the high burnups and heat rates required for competitive LMFBRs.

SIZE OF THE PROBLEM

Table I presents one prediction of the expected LMFBR fuel fabrication and processing loads in the United States. This projection assumes the introduction of commercial LMFBRs with properties similar to the Atomics International Follow-On design beginning in 1986 and the introduction of more advanced LMFBRs, similar to the General Electric Follow-On, by 1990. The table excludes fuel from the FFTF and the proposed demonstration plant. It can be seen that, although the U. S. fabrication load is expected to reach 1 metric ton/day by 1987, the U. S. processing load will not become significant before 1989; thus it will probably be prudent to stockpile spent fuel or process it in demonstration facilities until that time.

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Spent fuel will accumulate sooner in other countries, notably Britain, France, and the USSR, if they proceed with the construction of LMFBRs as planned. This will be discussed in more detail in a later section of this paper.

Table I. Predicted Fuel Fabrication and Reprocessing Loads
(metric tons/year)

Year	LMFBR		LWR
	Fabrication	Processing	
1986	140	4	5700
1987	240	7	6000
1988	390	67	6500
1989	580	165	7000
1990	820	300	7100
1991	1,350	475	7600
1995	4,700	2200	6600
2000	10,800	7250	4800

SPENT FUEL CHARACTERISTICS

The LMFBR concepts being developed are discussed elsewhere in this meeting, and a description of the reactor will not be given here. It is important, however, to consider the characteristics of the spent fuel that will be processed.

The reference U. S. fuel is UO_2 - PuO_2 clad in stainless steel. The core fuel element has three sections: the center section is the core containing uranium oxide plus about 20% plutonium oxide as the fissionable material, and the two end sections contain depleted uranium oxide that form the axial blanket of the reactor (Fig. 1). While it is practical to separate the core from the two end blanket sections and process them separately, there is no apparent advantage in doing so; hence, it may be assumed that they will be processed together. Elements containing depleted uranium oxide form the radial blanket which surrounds the core section of the reactor.

A comparison of projected LMFBR and LWR spent fuels is given in Table II. The expected average burnup for the General Electric Follow-On fuel is 100,000 Mwd/metric ton for the core, or about 42,000 Mwd/metric ton for the core-plus-blanket. This compares with 33,000 Mwd/metric ton for projected PWR fuels of the future. The decay heat from the LMFBR fuels, especially the core section at short decay times, is significantly greater than that of the LWR fuels. This is due to the higher specific power of the LMFBR as compared with LWRs.

Table II. Comparison of Fuel Discharged from Light Water Reactors and Future Liquid Metal Cooled Fast Breeder Reactors

	Typical LMFBR		Typical PWR with Recycle Pu	Typical PWR with Enriched UO_2
	Core	Mixed Core-Blankets		
Burnup, Mwd/metric ton fuel	100,000	42,000	33,000	33,000
Average specific power, kW/kg fuel	150	54	37.5	37.5
Decay heat, W/kg fuel				
30-day decay	190	75	65	50
150-day decay	80	30	30	20
Plutonium content, g/kg fuel	170	80	30	9

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ATOMICS INTERNATIONAL FOLLOW-ON SUBASSEMBLY (AI)

CHEMICAL PROCESSING

HEAD-END PROCESSING

Shipment of the spent IMFBR fuel by present technology would require a cooling period of about 1 year. The limiting factor is the temperature of the fuel cladding at the center of the bundle when only air is used as the coolant. Reducing the cooling period to 30-90 days will require a much more effective heat transfer medium, and our studies show liquid sodium, the reactor coolant, to be the most attractive possibility. Thus, fuel received at the processing plant may not only be contaminated with sodium but might actually be contained in a sodium-filled shipping cask. Under these circumstances, facilities and procedures for removal and deactivation of the external sodium at the processing plant will be required. This, we think, can be accomplished adequately by blowing off the massive quantities of sodium with an inert gas and deactivating the residual sodium with water vapor contained in an inert gas.

Beyond external sodium removal, the United States is pursuing development of two alternative head-end processing methods.

Mechanical Head-End Processing

This scheme¹⁻³ uses three steps (see Fig. 2) to prepare the fuel for dissolution: (1) mechanical disassembly; (2) shearing of the cladding to expose the fuel; and (3) a new process, voloxidation, for removal of tritium, removal of most of the iodine and noble gases, and final sodium deactivation prior to dissolution.

Disassembly. - Disassembly can be accomplished by various means, ranging from cropping of end adapters to subdividing the fuel unit into individual fuel pins. The purposes of disassembly prior to shearing are to minimize heat dissipation problems by reducing the number of fuel rods per unit and to minimize the difficulty of shearing.

Cropping operations can be performed rather simply through the use of an abrasive disk saw. The end adapters alone might be removed by such a procedure, or, conceivably, the gas plenums could also be removed, leaving a bundle of fuel rods contained within a relatively short length of shroud.

Alternatively, the fuel rods could be removed from the shroud. Most LWR fuels do not employ a shroud; consequently, this operation has little significance at present. Removal of the fuel rods from the shroud by a simple pulling operation is expected to be impractical because of anticipated swelling and bowing of the fuel. Hence, longitudinal cutting of the shroud is indicated. We have shown that this can be accomplished by grinding or by using plasma-arc torch system.

Shearing. - Intact IMFBR subassemblies can be sheared in a large unit having a force capability of about 250 tons. In early tests, the shroud fragments produced during shearing were sufficiently long and twisted to collect and lodge in the discharge port of the shear. Terminal pieces with lengths up to 3 in., which would require dissolution times of 20 to 40 hr to leach the contained fuel, were also produced. The development of a new blade design and the use of a larger discharge chute eliminated the first problem. Improved methods of restraining the fuel during shearing are now being developed to reduce the lengths of terminal pieces.

If subassemblies are dismantled prior to shearing, a very small, inexpensive multirod (up to 100 fuel rods) shear can be used. With this shear, fuel restraint is facilitated, heat dissipation problems are minimized, and the lengths of the terminal pieces of fuel are significantly reduced. A multirod (25 rods) prototype unit developed at ORNL has been operated at rates ranging from 40 to 120 cuts/min to shear stainless-steel-clad, porcelain-filled prototype fuel and has made 10,000 cuts without noticeable wear on the blade. Sheared pieces as short as 1/4 in. were successfully produced.

Removal of Volatile Fission Products (Voloxidation). - The voloxidation processing step⁴ is in an early state of development. The objective of this step is to remove the tritium from the fuel into a relatively small volume of dry gas, from which it can be efficiently removed in concentrated form for storage or permanent disposal. Some iodine, krypton, and xenon will also be evolved.

In this process step, the UO_2 - PuO_2 fuel is heated in an oxidizing atmosphere to a temperature of 450°C. During the oxidation to U_3O_8 - PuO_2 , a change in crystal structure takes place and the volatile fission gases are released. As an added benefit, the voloxidation step is also expected to deactivate any sodium present with the sheared fuel.

LMFBR MECHANICAL HEAD-END PROCESSING FLOWSHEET AND MAJOR PROBLEMS

The behavior of the volatile radionuclides was studied with four specially irradiated stainless-steel-clad fuel rods that originally contained sol-gel-derived 15% PuO₂--85% UO₂ microspheres. These rods had been irradiated in the Engineering Test Reactor to a burnup of about 20,000 Mwd/metric ton and then cooled for 32 to 65 days. About 50% of the ⁸⁵Kr and ¹³³Xe but essentially none of the other radionuclides were released to the off-gas when the fuel rods were sheared. From 8 to 67% of the ¹³¹I and from 11 to 65% of the ¹³⁴⁻¹³⁷Cs had migrated to and condensed in the cooler portions of the fuel rods (the gas plenum). The variations were apparently due to the temperature history of the fuel during irradiation.

In voloxidation tests, the sheared fuel segments were heated in a stream of helium and then oxidized at 450, 650, or 750°C for 4 to 8 hr in pure oxygen. In most of these tests, the weight gain upon oxidation (~3.7%) indicated stoichiometric conversion of the UO₂ to U₃O₈. In each case, less than 0.1% of the tritium originally produced in the fuel remained after voloxidation. From 32 to 75% of the ¹³¹I in the sheared fuel was removed, with the best result (75%) being obtained in a single test at 650°C. About 50% of the ⁸⁵Kr and ¹³³Xe in the sheared fuel rod segments was removed by the voloxidation treatments. Based on the total amounts found in shearing, voloxidation, and dissolution, a total of about 75% of the ⁸⁵Kr and ¹³³Xe and from 63 to 81% of the ¹³¹I were released prior to dissolution (Table III).

Table III. Residual Volatile Fission Products in Reactor Fuel After Shearing and Voloxidation

Fission Product	Residual % of Calculated Yield	
	LWR (<1% PuO ₂)	LMFBR (15-30% PuO ₂)
³ H	<0.1	<0.1
⁸⁵ Kr and ¹³³ Xe	30	25
¹³¹ I	20-40	20-40

Temperature and oxidizing conditions, as well as the effect of the presence of sodium, are currently being assessed.

Molten Zinc Decladding

An alternative head-end processing approach⁵⁻⁷ based on cladding dissolution is being pursued at Argonne National Laboratory (ANL). The dissolvent is molten zinc, at 800°C, contained in a tungsten vessel. Dissolution is rapid (about 2 mm/hr), and attack proceeds past the 8% solubility limit for stainless steel by dissolution of the nickel component and release of easily suspended metal grains. (The slurry is mobile with up to 20% solids.)

Application of the cladding dissolution concept is essentially as follows. Three fuel elements with the bottom hardware cropped off are drawn up into a turret, the bottom is closed, and the turret is transferred into an operating cell. Both the turret and the cell have argon atmospheres. The turret is moved into position at the top of a heated vessel, locks are opened in both the turret and the vessel, and the assemblies are lowered into molten zinc. The zinc is covered by a layer of molten calcium chloride to suppress zinc vaporization. After the shroud and cladding have been dissolved, the zinc slurry is pressure-siphoned out of the furnace vessel into large carbon-steel pipes. Most of the zinc is distilled from the pipes for recycle; the solid residue is sealed in the pipes for ultimate disposal.

The oxide fuel that remains in the vessel with the salt layer still retains about half of its original content of volatile fission products. The remaining volatiles are released when the oxides are reduced by the addition of a zinc-magnesium-calcium alloy. The reaction requires agitation of the vessel contents. Calcium is the reactant, and the resulting calcium oxide dissolves in the salt, while the uranium and plutonium metals form an alloy with the zinc-magnesium. The latter is pressure-siphoned from the vessel to an evaporation step for removal of excess zinc-magnesium, then cast into ingots for dissolution in nitric acid. The contaminated salt layer is transferred to large steel pipes and sealed for disposal.

A primary advantage of this process is that it removes essentially all of the volatile fission products from the fuel prior to dissolution. The iodine reacts with zinc vapor and is collected in the salt layer; measured decontamination factors exceed 10⁶. Krypton, xenon, and tritium accumulate in the argon cover gas and can be pumped to pressure cylinders for storage. No special measures for

removing sodium remaining on (or in) the fuel will be required since sodium has no influence on the decladding process.

DISSOLUTION

The rate of dissolution of PuO_2 - UO_2 fuels in nitric acid is affected by the fabrication and reactor histories. The independent effect of fabrication history on the rate of dissolution of unirradiated fuel is being determined in statistical factorial experiments⁵ at the Hanford Engineering Development Laboratory (HEDL). Likewise, the permanency of undesirable fabrication effects which may still exist after irradiation, as well as the influence of irradiation conditions, is being studied at ORNL.

Of six fabrication variables investigated for unirradiated mechanically blended fuel, the sintering temperature, the source of PuO_2 , and the plutonium content had major effects on dissolubility. As used here, dissolubility refers to the fractions of plutonium and uranium that dissolve when the oxide fuel is exposed to boiling 12 M HNO_3 for two 6-hr treatments. The effects of the rate of temperature rise during sintering, sintering time, and pellet-press pressure were minor, as shown in Fig. 3.

Preferential dissolution of uranium was found to occur in both irradiated and unirradiated samples. Greater than 99 wt % of the original plutonium remained undissolved in the worst case. Vol-oxidation has only a modest effect on the dissolubility of unirradiated fuel. To date, results obtained in experiments with irradiated fuel indicate the following: Plutonium recoveries of 99.8% or higher can usually be obtained by leaching for up to 12 hr with boiling 8 M HNO_3 . Most of the fuel dissolves rapidly; however, a small fraction of the plutonium dissolves quite slowly and, in some cases, is essentially insoluble in 8 M HNO_3 . Uranium recoveries are almost always higher than plutonium recoveries. In instances when high plutonium losses occur, apparently some of the plutonium in the fuel is segregated from the uranium and hence is in a refractory form. The refractory residues can be dissolved by adding 0.05 M HF to the leach solution. Thus, in general, dissolution is more complete after irradiation, and the effects of fabrication variables are minimized.

SOLVENT EXTRACTION OF LMFBR FUELS

The purification of LMFBR fuel solutions by solvent extraction can be accomplished by the Purex process, using the same general sequence of steps employed for LWR fuel solutions. The principal differences will center around the higher plutonium content of the LMFBR fuel solutions and their increased radioactivity at short cooling times. Associated with high plutonium content are problems relating to criticality, plutonium polymer formation, plutonium reductant, and equipment capacity in plants designed for processing fuel containing only small amounts of plutonium. The high specific radioactivity of LMFBR feed solutions has also caused concern about damage to the organic solvent and diluents used in the extraction process.

Criticality

Although criticality can be controlled by administrative procedures in a solvent extraction facility, the more desirable system involves the use of geometry and/or fixed poisons in combination with chemical phenomena known to limit fissile concentration. The problem of criticality control in both pulsed columns and centrifugal contactors is presently under study. It is calculated that solvent extraction contactors (pulsed columns or centrifugal contactors) having diameters of 10 in. or less will require plutonium concentrations greater than 1000 g/liter for criticality, provided that the ^{240}Pu content is as high as 12% and that there is minimal reflection of the contactor and minimal interaction with other vessels containing fissile materials. Normally, the ^{240}Pu content will be significantly greater than 12%. Further, plutonium concentrations as great as 1000 g/liter can never be encountered in a solvent extraction column without attendant precipitation and accumulation of solids.

Solvent Stability

Tributyl phosphate (TBP) in contact with nitric acid solutions decomposes slowly to form principally dibutylphosphoric acid (DBP), along with much smaller amounts of monobutylphosphoric acid (MBP). The rates of these reactions are enhanced in a high radiation field. When present in sufficient concentrations, the DBP and MBP decrease decontamination from fission products and increase uranium and plutonium retention by the solvent during stripping. They contribute to the formation of "cruds" and emulsions which, under extreme conditions, can cause shutdown of the solvent extraction system. The normal Purex flowsheet includes an alkaline wash cycle⁹ to remove DBP and MBP from the solvent and to prevent their accumulation to the level where such process problems are encountered.

THE EFFECT OF PuO_2 CONTENT AND SINTERING TEMPERATURE ON
DISSOLUBILITY OF $\text{PuO}_2\text{-UO}_2$ FUEL CONTAINING
CALCINED OXALATE PuO_2

We estimate that, in processing 30-day-cooled mixed-core-and-blanket material in pulsed columns, the solvent radiation dose would be about $0.1 \text{ W-hr liter}^{-1} \text{ cycle}^{-1}$. Considerably higher doses would result from processing more concentrated feeds, particularly those derived from dissolution of core material alone. In any case, however, it appears that the solvent dose during processing in pulsed columns should not exceed $0.5 \text{ W-hr liter}^{-1} \text{ cycle}^{-1}$. Exhaustive tests at ORNL have confirmed that adverse effects on decontamination factors and uranium retention are barely detectable at $1 \text{ W-hr liter}^{-1} \text{ cycle}^{-1}$ and are not cumulative; that is, the degradation products responsible for the effect are removed by the alkaline wash treatment.

To provide further assurance that the solvent can tolerate expected irradiation doses, batch cyclic irradiation studies¹⁰ were conducted in 15% TPP--n-dodecane at $0.3 \text{ W-hr liter}^{-1} \text{ cycle}^{-1}$ to an integrated dose of about $6.5 \text{ W-hr liter}^{-1}$. Tests were also made¹¹ in a continuous system consisting of three banks of mixer-settlers preceded by a single-stage irradiation contactor. As expected, no adverse effects on solvent performance were observed.

Centrifugal Contactors

In the expectation that the high plutonium content of LMFBF fuel will lead to criticality problems, ANL is developing a mixer-settler with small holdup volume¹² for use in the plutonium isolation step of the Purex process. This device is patterned after the multistage centrifugal mixer-settler bank that was installed in parallel with the Purex 1A mixer-settler bank¹³⁻¹⁴ at the Savannah River Plant (SRP) in 1966 and has been in constant service since then. Maintenance requirements of the SRP centrifugal contactor, as described by Orth et al., have been minor. The principal operating advantage of the centrifugal contactor is ease of startup and shutdown. The design being studied retains the air control weir but has a smaller-diameter bowl (current version, 4 in.) turning at higher speeds (currently up to 4000 rpm). The intent of the program is to gather enough information to allow prediction of the processing capacity of a unit of small dimensions and feasible speed. The 4-in. unit has a holdup of about 2 liters; at 3000 rpm, the residence time per stage can be as short as about 5 sec for typical Purex liquids. Eventually, a bank of perhaps six units will be constructed to permit demonstration of bank control systems and remote operation.

CONTROL OF RADIOACTIVE EFFLUENTS

The routine discharge of solid, liquid, and gaseous waste materials containing detectable amounts of radioactivity is one of the major issues facing the nuclear power industry in the United States today. This problem will become more acute for the nuclear fuel reprocessor as the amount of fuel to be reprocessed, particularly the amount in a single facility, increases and as the cooling time decreases. For example, Table III compares the iodine problem encountered in a present LWR fuel processing plant handling 150-day-cooled fuel with that for a future LMFBF fuel processing plant handling 30-day-cooled fuel. It should be emphasized that the required containment factors presented in Table III are based on the present maximum permissible release rates and, consequently, should probably be increased by two orders of magnitude for future use.

Present regulations do not require retention of ^{85}Kr and ^3H until large plant sizes are reached; again, however, a containment factor of at least 100 seems reasonable for the future.

Effluent Control

A preliminary flowsheet for treating the gaseous wastes from a future plant processing LMFBF fuel is outlined in Fig. 4. Such a system can be effective and economical only if off-gas volumes are reduced from present-day standards of tens of thousands of cfm to hundreds of cfm. This can be accomplished by sealing the cells and recycling the off-gas, as illustrated in Fig. 4. This flowsheet also illustrates how water may be recycled to reduce the volume of liquid effluent that must be treated to remove radioactivity and various noxious materials.

As has been previously mentioned, greater than 99% of the tritium is liberated in the voloxidation step in a small dry stream of gas from which it can be effectively removed (e.g., by adsorption on a molecular sieve). Iodine, xenon, and krypton are evolved in all three process steps prior to solvent extraction. The krypton can be removed to the degree indicated after the gas stream has been cleaned of reactive gases by scrubbing with a fluorocarbon.¹⁵⁻¹⁶ The iodine, however, presents a greater problem because of its complex chemistry and because of the high degree of containment (a factor of $>10^7$ for short-cooled fuel) required under present regulations.

The preliminary flowsheet (Fig. 4) indicates that the off-gas streams will probably be segregated on the basis of iodine content and treated accordingly. The off-gas from the head-end steps and the

FUEL REPROCESSING PLANT-EFFLUENT CONTROL FLOWSHEET

dissolver will contain >99.9% of the iodine. The preferred methods for treating these streams for iodine removal (see Table IV) are: scrubbing with 16 N HNO_3 in a distillation tower, scrubbing with nitric acid containing mercuric nitrate at near-room-temperature, and adsorption by a silver-impregnated zeolite. Each of these methods has been shown to provide a decontamination factor of $>10^4$ for gas streams containing iodine. Silver zeolite is selected for use in the final step since it is much more resistant to oxidation and poisoning than charcoal and, therefore, more reliable.

Table IV. Methods for Removing Iodine from LMFBR Fuel Processing Plant Off-Gas

	Probable Flow (cfm)	DF Expected	Preferred Methods
Primary system	50	10^4	HNO_3 or HNO_3 -- mercuric nitrate scrubber
Secondary system	200	10^4	HNO_3 or HNO_3 -- mercuric nitrate scrubber
Final treatment	500	10^4	Silver zeolite

Results of laboratory tests indicate that 16 N HNO_3 will be effective as the primary treatment and will not be sensitive to impurities expected in the off-gas. Also, a scrub with 10 N HNO_3 containing mercuric nitrate is effective.

Pilot-plant-scale tests of these and other methods are in progress at ORNL. Results to date have demonstrated iodine retention factors as high as 10^6 (the limit of detection) and have shown that the use of successive iodine removal processes can provide improved containment.

The nature of liquid and solid wastes expected from future LMFBR processing plants will differ little from that of present wastes. In future designs, more emphasis will be placed on routing all radioactivity to the high-level waste streams in order to minimize the amounts of intermediate- and low-level wastes.

PLANS OF OTHER COUNTRIES

The Germans plan to operate their WAK plant at Karlsruhe to gain experience and to improve its performance with regard to processing advanced LWR fuels; eventually, they plan to adapt the plant for processing fast breeder fuel of the SNR type.¹⁷ In particular, they are engaged in studies of fuel receiving, cleaning, storage, and head-end processing. Their approach is quite similar to that previously described in the program being carried out in the United States. They plan to use the shear-leach process, which involves sodium removal, whole-bundle shearing, voloxidation, and leaching with nitric acid. They are also investigating methods for improving the treatment of effluents.

France^{18,19} and the USSR^{20,21} are evaluating both the Purex and the voloxidation processes for treating their fast breeder fuels. Although the Russians intend to process their early fuel in existing plants, they are also developing technology for a plant designed solely for fast reactor fuels. In this way, they are following the same route pursued in Germany and the United States, including improved effluent control. In a volatility approach, the Russians are studying melt de-cladding followed by fluorination. Their scheme calls for volatilization of the uranium by fluorination at 350°C, leaving the plutonium behind as PuF_4 . They are considering the use of both solid sorbents and distillation as means for purifying the uranium, but they have made few comments about the difficulties of purifying the plutonium.

The French fast fuel processing pilot plant, ATL, at La Hague Center has been in operation since 1968. Two fuel charges from Rapsodie have been successfully processed there. A fluoride volatility process is being tested in their Attila hot-cell equipment. In this process, the fuel is chopped, separated from the cladding by milling, and fluorinated in a fluid bed. Using spent Rapsodie fuel, the French have been successful in recovering clean uranium but, like others, have had difficulty decontaminating the plutonium. The French plan to use the new chop-leach head-end facility they are installing on the La Hague plant to process their early fast fuels assuming, of course, a suitably long decay period for these fuels.

The British²² plan to utilize the new chop-leach head-end facility on the Windscale plant to process early fast fuels but have not reported development of processes specifically for use with such fuels.

FUEL FABRICATION AND FUEL ELEMENT TECHNOLOGY

E. E. Kintner told fast reactor fuel experts at the American Nuclear Society Conference held in New Orleans in April 1971 that, after 20 years of fast breeder reactor development in the United States, "We do not yet have the technical understanding necessary to design any fast reactor fuel element with great confidence and will not have that technical understanding necessary to design the fuel elements needed for competitive nuclear breeder industry unless we find ways to make more effective use of applied resources."²³ He related the recent experiences of LMFBR fuel designers to those of Dante, who one day realized that he had strayed from the "true way" into the "dark woods of error." Most of his comments related to understanding fuel and cladding performance, but all of them have a direct bearing on the processes involved in the fabrication of fuel. Fuel performance is, of course, greatly dependent on the fuel form, structure, and chemistry, as well as similar characteristics of the cladding. It is appropriate, therefore, to review the status of the technology with regard to processes involved in the fabrication of fuel and in fuel performance. It would also be instructive to consider the options that still remain for fabrication technology and fuel design.

SUMMARY OF EXPERIENCE WITH FAST BREEDER FUEL

P. Murray²⁴ has reviewed comprehensively the experience to date with LMFBR ceramic fuels, which are the ones of primary interest for commercial LMFBR application. In his review he pointed out the technical involvement of a number of countries, including Russia, United Kingdom, United States, France, West Germany, Japan, and Italy, as well as supporting efforts in other countries.

His review may be summarized as follows. Fuel rod irradiations in test reactors have demonstrated the high performance capability of mixed-oxide stainless steel systems. Fission gas release and fuel swelling can be accommodated, and the benefits arising from irradiation-induced effects of swelling and creep in stainless steel cladding give confidence that high-burnup targets for LMFBR fuel can be achieved. Fuel-cladding chemical interaction has emerged as a potential problem that may limit the peak cladding temperatures to which mixed-oxide fuel rods are designed and operated. Further detailed results on these limits are needed, particularly in the high burnup (10% FIMA) range. Reactions caused by fuel-sodium interactions under dynamic cooling conditions do not appear to cause major operating problems. However, definition of fuel rod thermal performance models requires additional experimental data to minimize the uncertainties.

Stainless steel swelling and irradiation-induced creep have, in effect, focused attention on the fuel assembly and on the behavior of the fuel rod spacer and flow duct systems. These are vital areas in which more precise materials irradiation data and interpretation for design are necessary. Present uncertainties can be overcome by a design giving more reliable irradiation data concerning the high fluences required for LMFBR application. Nevertheless, accommodation of swelling does introduce an economic penalty that highlights the importance of long-term metallurgical solutions to the swelling problems.

The experience with irradiations of oxide fuels in fast reactors has been summarized²³ and is given in Table V. In the irradiation program in the United States, several uranium-plutonium-carbide and uranium-plutonium-nitride irradiations are also being performed, as indicated in Table VI.²⁴ The data presented in these tables show that substantial experience has been gained in fabricating fuel rods for irradiation, although the amount of experience with the advanced fuels is limited. Most of the experience in fabricating uranium-plutonium oxide fuels has been obtained by using either the mechanically blended or the coprecipitation route.

In addition to the fuel fabrication experience indicated in Tables V and VI, essential experience is being obtained through the procurement of fuel for the Fast Flux Test Facility (FFTF). The program for developing high-quality fast reactor fuels in support of the FFTF was reported by W. E. Roake et al.²⁵ at the Fourth Geneva Conference. In this paper, Roake pointed out that strong emphasis is placed on product specifications, process description and control, and quality assurance procedures and practices. The flowsheet for the FFTF fuel is based on the use of mechanically mixed oxides. However, substantial experience has been gained with the coprecipitated mixed oxide

material; in particular, the coprecipitation process was used by the Kerr-McGee Corporation for the production of mixed oxides for the zero power reactor fuel rods during 1970 and 1971 at their Cimarron Plutonium Facility. Kerr-McGee produced 13,000 fuel rods with the nominal composition (Pu-85 wt % U)O₂ and 6000 fuel rods with the nominal composition (Pu-70 wt % U)O₂. In addition, Nuclear Fuel Services has used the coprecipitation process for the production of SEFOR fuel, and General Electric²⁶ and NUMEC²⁷ have also used the coprecipitation process for production of mixed oxide fuels. Substantial experience exists for the laboratory-scale preparation and fabrication of mixed-oxide fuels by sol-gel processes.²⁸⁻³⁰

Table V. Oxide Irradiations in Fast Reactors^a

Country	Reactor	Number of Rods Irradiated	Number of Rods Failed
U.S.S.R.	BR-5 and BOR-60	4600	120-150
U.K.	DFR	800	50
France	Rapsodie and DFR	8000	1
U.S.A.	EBR-II and SEFOR	1500	10
Other	Rapsodie and DFR	200	3

^aAbstracted from Nuclear Engineering International, p. 496
(June 1971).

Table VI. Fuel Rods Irradiated to Various Burnups in EBR-II^a

Type of Fuel	Up to 5 at. %		To 5 - 10 at. %		Beyond 10 at. %		Total	
	Encap- sulated	Unencap- sulated	Encap- sulated	Unencap- sulated	Encap- sulated	Unencap- sulated	Encap- sulated	Unencap- sulated
Oxide	147	550	127	241	39 ^b	0	313	800
Carbide	49	0	71	0	8	0	128	0
Nitride	18	0	0	0	0	0	18	0
Metal + cermet	70	196	4	38	0	0	74	234
Total	284	756	202	279	47	0	533	1034

^aStatus as of December 1971.

^bPeak cladding fluences up to 0.95×10^{23} neutrons/cm² ($E > 0.1$ MeV) have been achieved.

PROCESS OPTIONS FOR MANUFACTURE OF LMFBR FUEL RODS

The principal competing flowsheets for fabrication of LMFBR core-axial blanket fuel elements are: the coprecipitation flowsheet, the mechanically mixed flowsheet, and the sol-gel pellet and sol-gel Sphere-Pac flowsheets. In addition to these indicated routes, vibratory compaction of angular particles or shards has been considered and rejected by most investigators.

When the three principal routes for preparing pellets are compared, the differences are found to lie only in the preparation of the fuel materials. The coprecipitation and mechanically mixed routes are quite similar in that prepressing and granulation are required in order to prepare a material suitable for pressing to obtain products with the lower stable densities that are required for LMFBR fuel. In the sol-gel pelletizing route, this step is accomplished by adjustment of the calcination procedure, which is conducted in the temperature range 400 to 700°C on dried gel material. The principal disadvantage of the mechanically mixed route is the need for inspections and qualified procedures to ensure homogeneous mixture of the UO₂ and PuO₂. The principal problem in the coprecipitation route lies in control of the simultaneous precipitation of plutonium and uranium intermediate products. However, as previously noted, a coprecipitation step has been used in very large-scale operations, and the results indicate that the mechanically mixed route will prove to be satisfactory in the FFTF program.²⁶

In the sol-gel route, the subsequent preparation of a suitable powder for pelletizing is more straightforward than the other two routes even though the preparation of the sol may be a more complicated operation than for either precipitation or coprecipitation. The only mechanical operation necessary is a grinding step, which has been accomplished in a fluid energy mill and does not require screening.³¹ To summarize the status of the three processes, both the coprecipitation and the mechanically blended routes involve a large number of mechanical operations. However, operations using these routes have been conducted on a large scale, and a great deal of industrial experience concerning them is available. The sol-gel pelletizing route, on the other hand, involves a greater number of chemical operations and less mechanical handling, but only results from laboratory-scale operations are available.

The Sphere-Pac route for fabrication of core axial blanket LMFBR fuel elements consists of the preparation of plutonia and uranium sols, blending them, and forming two sizes of microspheres, typically 400 and 40 μm , from the mixed sol. Several procedures for preparing such microspheres have been reported in the literature.²⁸⁻³⁰ Microspheres with densities 95 to 99% of theoretical are produced by a drying and sintering procedure carried out over the temperature range 200 to 1400°C. Adjustment of the fuel chemistry such as the oxygen-to-metal ratio is done in a manner identical to that for pellets.

Sphere-Pac is a technique for sequentially loading the two sizes of microspheres into a fuel column using low-energy vibration. The basic principles of packing spheres into tubes have been described by McGlary³² and Ayer.³³ In their procedure, coarse microspheres are first loaded into the fuel tube and then fine microspheres are infiltrated into the interstices. During the loading of the fine fraction, the coarse bed is retained by a screen over the bottom of a funnel that rests on top of the coarse bed. Fuel smear densities ranging from 80 to 85% of theoretical density are readily obtained in this method. Sease and his co-workers³¹ have reported an improvement of this process, called the "U-Fine" process, which is a form of Sphere-Pac loading in which all the plutonium is contained in the coarse microspheres. The fine fraction consists only of pure UO_2 . A comparison of the U-Fine method with conventional Sphere-Pac processes is given in Fig. 5. Fuel inhomogeneity should not be a problem since coarse microspheres for the nominal composition in the core of $(\text{U}-20\% \text{Pu})\text{O}_2$ would have to contain only 27% plutonium. The principal advantages of the process are that no fine plutonium material is necessary and that the quantity of material that must be handled with a high degree of containment and shielding is reduced.

Differences in the economics of the various preparation procedures are not substantial and probably lie within the error of the estimates. Both Sease³¹ and his co-workers and G. Colombi and L. Podo³⁴ have assessed the difference in cost between low-energy Sphere-Pac process and pelletizing routes. Sease concluded that the cost differential is in the range of \$20 to \$25/kg heavy metal in a plant producing 500 kg of core material in finished elements per day. In their study, Colombi and Podo saw little economic difference in favor of sol-gel processes. It should be pointed out, however, that more detailed assessment should be made of the alternative processes with particular attention to any differences in process losses, the requirement for internal recycling of fuel material, and dissolubility of the spent fuel in fuel reprocessing. In addition, there may be some differences in the degree of contamination and the relative difficulty associated with it among the various processes.

RELATIVE FUEL PERFORMANCE

As previously noted, a large number of mixed oxide fuel rods have been fabricated and irradiation-tested. In most cases, the fuel pellets for these rods were prepared by coprecipitation and mechanical blending routes. Accordingly, most of the data being accumulated are for pellets, and this experience generally indicates that there may be some problems with respect to maximum temperature of the fuel, primarily due to fuel-cladding chemical interaction. Although most of the experience is being obtained on pellet fuels, the future application of a fuel system to the LMFBR should consider other alternatives; in particular, results from irradiation of Sphere-Pac fuels are encouraging in that these fuels have exhibited better compatibility with cladding and better fuel-cladding thermal conductance.³⁵ However, these results have been obtained with a relatively small number of fuel rods, and any conclusion should await the results of postirradiation examination of additional rods, which are presently being irradiated in the EBR-II.

NONFUEL HARDWARE MANUFACTURING

Because of the stringent service conditions expected for the various hardware contained in LMFBR fuel elements, it is necessary that detailed attention be given to the metallurgical characteristics of the materials used and to strict quality assurance procedures for the manufacture of such materials. Roake²⁶ has reported the progress of the FFTF hardware procurement program in obtaining increased levels of quality, and this improvement can be expected to continue as industrial experience is

Comparison of Conventional with U-Fine Sphere-Pac Process

gained. The commercial application of IMFBRs will require the continual assessment of the technology of the structural materials since developments can be expected to lead to cladding materials, for example, with less susceptibility to chemical attack by the fuel and less susceptibility to swelling. It should also be noted that the hardware costs, particularly for tubing, account for a substantial part of the fuel fabrication costs; therefore, economies in processing suitable tubing and other hardware must be obtained if the goal of a competitive IMFBR is to be reached. In fact, the fuel element hardware can account for more than 50% of the total costs of manufacturing IMFBR core axial blanket fuel elements.

FUTURE PROJECTIONS FOR THE IMFBR FUEL FABRICATION INDUSTRY

The questions involving the future of the IMFBR fuel fabrication industry concern (1) fuel fabrication demands, and hence scale of operation; (2) fuel fabrication plant design; (3) probable cost; and (4) probable fuel performance. In addition, an analysis of the type of economies that can be reached through certain improvements is of great interest since such an assessment would serve as a base line for the cost benefit analysis of research and development expenditure in the future.

The projected fabrication demand, based on assumed reactor economy, was presented in Table I. In addition to this, reports of the system analysis task force³⁶ and the fuel recycle task force³⁷ show comprehensively the type of demands required for the industry in the future as well as provide an estimate of the cost of fuel fabrication. At least one commercial plant with capacity for fabricating core fuel at the rate of 0.5 metric ton/day or greater can be expected by 1986. At least six commercial plants with capacities for fabricating a total of 12 metric tons/day of core fuel are expected by the year 2000.

Bell and Nichols³⁸ considered the capabilities of various types of operations classified according to mode of operation, that is, glove boxes, semiremote operation, or remote operation (operations that have been defined by Shuck et al.³⁹). In their analysis, they estimated maximum capacities of fuel fabrication lines preparing UO_2 - PuO_2 as a function of shield thickness. These data are shown in Table VII. The calculated shielding requirement for the manufacture of IMFBR core fuel at a capacity of 500 kg/day was 22 in. of concrete. (That shielding will be required is unquestioned.) For the future, the problem is to determine the most economical method of incorporating the required shielding and the most economical method for the design of the process in order to minimize exposure of the operators. Process designs, therefore, should emphasize automation and mechanization, close integration instrumentation to minimize holdup of materials.

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Table VII. Estimated Maximum Capacities of Lines for Preparing UO_2 - PuO_2 for IMFBRs^a

Type of Operation	IMFBR Core (LWR Recycle Pu): $Pu/(U + Pu) = 0.22$	
	Concrete Shield	
	Thickness (in.)	Capacity [kg (UO_2 - PuO_2)/day]
Gloved enclosures ^b	0 ^c	7
Semiremote ^b	12	30
Remote ^d	16	100
	22	500
	24	1000

^aAssumed operator dose rate = 40 mrem/week.

^bAssumes that process equipment is installed 1 ft from inner wall of enclosure.

^cAssumes 1/4 in. of lead for shielding soft gamma radiation.

^dAssumes that process vessels (which contain no more than 20% of daily throughput) are installed 2 ft from the inner surface of the shield and that the dose rate at the operating surface is 1.0 mrem/hr.

Frankhouse, in his paper,⁴⁰ concluded that the NUMEC experience with plutonium fuel fabrication does not provide the basis for design of an oxide fuel rod production facility but does allow guidelines to be formulated. He pointed out that, other than isotopic composition of plutonium, such practical considerations as age of fuel, concentration of plutonium in fuel, fuel surface area-to-volume ratio, and fuel mass must be taken into account. Plant design per se can vary from glove-box enclosures with shielding, general enclosure shielding, and shielding on process equipment to completely remote operation. A thorough evaluation of capital cost vs operating cost must be made in order to find the optimum solution to the problem. At this point, it would appear that remote operation with automatic equipment would be a prudent choice for large-scale plants. This would allow for a number of contingencies regarding reduction of permissible operator exposure as well as unexpected increases in radiation associated with the fuels to be fabricated. Operation of the HTGR fuel recycle pilot plant projected for the period 1977-1980 should help define the cost of remotely fabricating nuclear fuels.⁴¹

The fuel recycle task force³⁷ has developed base cost information for the fabrication of LMFBFR fuel elements. Typical results from this work are shown in Fig. 6. These costs, which were developed with the use of FABCOST 9 computer code,^{42,43} use a very low fuel-element hardware cost (e.g., approximately \$1.25 per foot of stainless steel tubing). Substantial differences are indicated in the fuel fabrication costs for the various referenced reactors. Most of these differences are attributable to variations in the dimensions of the fuel rods. For example, although the preparation of carbide fuel forms are more expensive, subsequent fabrication is less expensive on a weight basis of fuel because the high thermal conductivity of the carbide permits the use of relatively large-diameter pins with high loadings of fuel per unit of length. The effect of hardware cost, particularly tubing cost, is indicated in Fig. 7. This figure shows the reference case for an LMFBFR in the systems analysis task force study compared with a high cost for tubing representing present-day cost, and with an intermediate cost that represents at least a probable cost for the future. At a production rate of 1000 kg of heavy metal per day, escalation of the tubing cost from \$1.26/ft to \$6/ft doubles the fuel fabrication cost.

The value of trade-off in terms of final power cost is an important assessment. Figure 8, taken from McVey et al., is a plot of fuel cycle cost, in mills/kWhr, vs core burnup for several fuel cycle costs in terms of costs per kilogram of heavy metal. Applying data of this type to the worth of various changes for LMFBFR core fuel is instructive. Table VIII shows the effects of making several important changes with respect to scale of operation, process selection, hardware costs, and burnup capability of the fuel. Changes such as increasing the scale of operation by a factor of 2, changing the mode of fabrication, and using a different fabrication process when considering only the processing costs result in a decrease of cost on the order of 5 to 6%. Reduction of tubing costs from the present rate of the order of \$6/ft to \$3/ft gives an improvement of approximately 20%; a reduction to \$1.25/ft gives an improvement of approximately 50%. It is notable also that burnup capability greatly affects the power cost. Therefore, one would conclude from these results that significant attention should be given to methods of reducing the cost of fuel element hardware, particularly tubing, and to selection of particular fuels which will enable the reactors to operate at maximum specific power and achieve a higher burnup.

Table VIII. Worth of Various Changes for LMFBFR Core Fuel^a at 1000-kg/day Scale

	\$/kg	mills/kWhr(e)	%
Basic reference cost	530	0.830	
Increase in scale of operation to 2000 kg/day	-30	-0.047	-5.7
Use of glove box facility instead of remote operation	-29	-0.045	-5.5
Use of Sphere-Pac process instead of mechanically blended powders	-24	-0.038	-4.5
Reduction in tubing cost from \$6/ft to \$1.25/ft	-268	-0.420	-50.6
Reduction in tubing cost from \$6/ft to \$3/ft	-98	-0.153	-18.5
Decrease in average burnup from 10 at. % to 6 at. %		+0.35	+42.2

^a10% FIMA average burnup, exhibit 55 - SATF Study (WASH-1098).

Cost of Fabricating LMFBR Fuel

Effect of Tubing Cost on LMFBR Core Fuel Fabrication Cost

Fig. 8. Effect of Burnup and Ex-Reactor Cost on Fuel Cycle Costs.