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MASTER

LMFBR FUEL CYCLE STUDIES

PROGRESS REPORT FOR OCTOBER, NO. 8

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Contract No. W-7405-eng-26

CHEMICAL TECHNOLOGY DIVISION METALS AND CERAMICS DIVISION

LMFBR FUEL CYCLE STUDIES PROGRESS REPORT FOR OCTOBER, NO. 8

I. AQUEOUS FUEL REPROCESSING

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II. FUEL MATERIAL PREPARATION

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III. FUEL FABRICATION AND EVALUATION

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NOVEMBER 1969

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CONTENTS

	<u>Page</u>
ABSTRACT	1
HIGHLIGHTS	1
I. AQUEOUS FUEL REPROCESSING	
1. Shipping (Task 1)	4
2. Receiving and Storage (Task 2)	7
3. Head-End Processing of LMFBF Fuels (Task 3)	8
4. Volatile Fission Product Removal (Task 4)	16
5. Dissolving (Task 5).	20
6. Feed Preparation (Task 6)	23
7. Solvent Extraction (Task 7)	25
8. Plutonium Purification (Task 8)	30
9. Waste Treatment and Storage (Task 9)	30
10. Off-Gas Treatment (Task 10)	31
11. Radiation and Shielding (Task 11)	36
12. Criticality (Task 12)	44
13. Engineering Studies (Task 13)	47
II. FUEL MATERIAL PREPARATION	
1. Conversion Studies	58
2. Sol-Gel Processes for Preparation of LMFBF Recycle Fuel	61
III. FUEL FABRICATION AND EVALUATION	
1. Fabrication Development.	70
2. Fuel Evaluation	86

LMFBR FUEL CYCLE STUDIES PROGRESS REPORT FOR OCTOBER, NO. 8

ABSTRACT

This report continues a series outlining progress in the development of methods for the reprocessing and fabrication of LMFBR fuels. Development work is reported on problems of irradiated fuel transport to the processing facility, the dissolution of the fuel and the chemical recovery of the $\text{PuO}_2\text{-UO}_2$ values, the containment of volatile fission products, product purification, preparation of fuel material by the sol-gel process, conversion of fuel processing plant product nitrate solutions to solids suitable for shipping and for fuel fabrication, fuel fabrication of sol-gel materials, and fuel evaluation studies, both in-pile and out-of-pile. Pertinent experimental results are presented for the information of those immediately concerned with the field. Detailed description of experimental work and data are included in topical reports and in the Chemical Technology Division and Metals and Ceramics Division Annual Reports.

HIGHLIGHTS

Division I

A 1/6-scale cask model was dropped a second time from a height of 30 ft onto an unyielding surface. Increased deformation was slight and there was no discernible increase in the barely detectable (via helium mass spectrometer) leak rate present after the first drop test. (Sects. 1.3 and 1.4)

Equations were derived for the calculation of new more accurate HEX black body view factors using Hottel's crossed-string method. (Sect. 3.1)

In the processing of short-cooled LMFBR fuel specimens, more than half of the total ^{131}I found was in the gas plenum. Up to 74% of the amount remaining in the fuel sections was released during a voloxidation treatment. (Sect. 4.1)

Essentially quantitative recoveries of uranium and plutonium were obtained by dissolving irradiated uranium mononitride in boiling 8 M HNO_3 for about 2 hr. (Sect. 5.1)

About 99.9% of the ^{131}I remaining in the feed solution after dissolution of IMFBR fuel specimens was volatilized by sparging after addition of relatively large amount of KI followed by addition of NaNO_2 . (Sect. 6.2)

Passing air containing dodecane vapor through Hopcalite oxidation catalyst at 350°C completely oxidized all organic material to CO_2 and H_2O . With no Hopcalite present, all of the dodecane was destroyed at 350°C but some intermediate organic oxidation products were formed. (Sect. 10.1)

ORIGEN calculations were performed for the AI follow-on reactor for the inner core, outer core, axial blanket, inner and outer radial blankets and for the homogenized discharge from the reactor (based on the refueling schedule). (Sect. 11.2)

A series of criticality calculations reveals that an array of 36 IMFBR core elements in a sodium-filled, iron-shielded cask will be appreciably subcritical but that the source neutron multiplication factor may be in the range of 3 to 15. The interspersing of radial blanket elements or the use of lithium instead of sodium as the coolant are effective means for reducing the source neutron multiplication and, hence, the neutron shielding requirements. (Sect. 11.4)

Material balance flowsheets have been prepared for reprocessing of FTR fuel cooled 180 days. Concept definition of a facility for receipt of fuel subassemblies shipped in a sodium-filled cask is essentially complete. (Sect. 13.1)

Division II

It was established that sol prepared from fresh 1 M U(IV) nitrate-formate feed solution ($\text{NO}_3^-/\text{U} = 2$; $\text{HCOOH}/\text{U} = 0.5$) had a lower U(IV) content than those prepared from feed that had been aged overnight. Chemical tests on feeds aged under argon for up to 4 days showed no self-oxidation of the feeds. The aged feeds produced sols with $87 \pm 1\%$ U(IV) content. (Sect. 2.1)

Formic acid has been definitely associated with undesirable urania microsphere clustering in the sphere forming column. A caustic scrub to remove formic acid (and coincidentally, nitric acid) has been added to the experimental engineering microsphere forming equipment. After the initial difficulty with caustic entrainment in the scrub 2-ethyl-1-hexanol was overcome by changing from spray columns to a packed column, a caustic scrub was found to behave quite satisfactorily in removing formic acid. This improvement should effectively remove a significant sphere forming problem. (Sect. 2.3)

Division III

Two Series 1 TREAT experiments, which are to compare the transient behavior of unirradiated sol-gel-derived Sphere-Pac and pelletized (U,Pu)O₂ fuels, were shipped to the reactor site on October 14, 1969. (Sect. 1.2.1)

Work was started on the preparation of hardware and fuel for the Series 2 unencapsulated EBR-II tests, which are to compare (U,Pu)O₂ fuels fabricated by different processes. (Sect. 1.2.2)

Gas release measurements on (U, Pu)O₂ microspheres suggest that the largest apparent gas evolution occurs at or below 900°C, that at above 1200°C some of the gas is reabsorbed, and that at 1600°C or above gas is again evolved. (Sect. 2.1.1)

Five fuel rods containing Sphere-Pac fuels being irradiated in the EBR-II in subassembly X050 have been exposed to 6400 Mwd of operation. This is equivalent to a calculated peak burnup of 3.4% FIMA. A request for "Approval-in-Principle" to extend the burnup for three of these tests from 5% FIMA was submitted to the USAEC this month. (Sect. 2.2.5)

I. AQUEOUS FUEL REPROCESSING

(W. E. Unger, R. E. Blanco, D. J. Crouse, A. R. Irvine, C. D. Watson)

1. SHIPPING (TASK 1)

(A. R. Irvine, J. D. Rollins, R. S. Lowrie,
D. C. Watkin, J. H. Evans)

The objective of Task 1 is to assure that an economic and safe method of shipment of LMFBR spent fuel will be available when needed for transport of fuel from the demonstration and early commercial LMFBR's. The work involves analytical studies of the various facets of the problem; design, construction, and test of components, and of assemblies; and preliminary design of prototype casks.

The bulk of the effort on this task is directed toward expanding the store of information on the effectiveness of sodium as a primary coolant and of cask design features which can assure containment of coolant (and fission products) within the cask fuel cavity. Other coolants (including gases, liquids, and solids) have been considered, but they do not appear to be as attractive as liquid sodium.

Work performed during this report period was primarily in the areas of Tasks 1.1 through 1.4, which deal with evaluation and test of heat dissipation methods and of cask integrity.

Reportable accomplishments include the following:

1.1 Evaluation of Heat Dissipation Methods (Task 1.1)

- A. Preliminary heat transfer calculations were made in order to compare the thermal characteristics of LWR fuel with those of LMFBR fuel, and accordingly to illustrate the necessity for a liquid coolant in the case of LMFBR fuel shipment. Table 1.1 lists the results of the calculations (approximate) which indicate that the maximum fuel clad temperature reached in an LWR fuel subassembly which has cooled for 150 days is several hundred degrees less than that reached in an LMFBR fuel subassembly which has cooled for 30 days. It is further shown that decay periods of approximately 800 and 1400 days, respectively, would be required to reduce the maximum clad temperature in AI Reference Oxide LMFBR (a 217 rod cluster irradiated to 80,000 Mwd/T) and GE LMFBR (a 331 rod cluster irradiated to 100,000 Mwd/T) type fuel subassemblies to that of a 150-day cooled LWR fuel subassembly (a 225 rod square array irradiated to 33,000 Mwd/T). The reasons for the higher fuel clad temperatures in the LMFBR type subassemblies include: a more compact fuel pin array (triangular as opposed to square) more rows of fuel pins, higher linear heat generation rates

(even for the case of equal decay periods), and, in the case of the GE fuel subassembly a smaller pitch to pin diameter ratio. The need is, therefore, evident for an effective heat transport medium (e.g., liquid sodium) inside the fuel cavity of LMFBR in order to transport fuel subassemblies safely after short cooling periods.

Table 1.1 Maximum Fuel Clad Temperature in Average LWR and LMFBR Fuel Subassemblies As a Function of Decay Time for Various Shroud Temperatures

Fuel Shroud Temp., °F	Decay Time, days	Maximum Fuel Clad Temp., ⁽¹⁾ °F		
		PWR ⁽²⁾	AI Ref. Oxide LMFBR ⁽³⁾	GE LMFBR ⁽⁴⁾
400	30	1180	1990	2370
400	150	890	1490	1790
400	~ 800	—	890	1070
400	~1420	—	—	890
800	30	1280	2020	2390
800	150	1060	1570	1830
800	~ 790	—	1060	1200
800	~1400	—	—	1060

⁽¹⁾ Excluding axial peaking factors.

⁽²⁾ Burnup: 33,000 Mwd/Metric Tonne, specific power: 33 kw/kg, 225 pin array.

⁽³⁾ Burnup: 80,000 Mwd/Metric Tonne, specific power: 175 kw/kg, 217 pin array.

⁽⁴⁾ Same parameters used as for AI fuel in ⁽³⁾ above with the exception of the pin array which included 331 pins.

1.2 Heat Dissipation Tests (Task 1.2)

- B. A series of tests to ascertain the extent that the baffles in the reduced scale cask mockup interfere with the thermal convective flow of the mercury heat transfer medium within the fuel cavity were completed. Data from these tests are being analyzed and will be reported next month.

- C. The second startup of the electrically heated mockup of an Atomics International Reference Oxide fuel assembly has been further delayed due to continued slippage in the heater delivery schedule. On the basis of test results (at ORNL during the first week of October) on 9 tube heaters (supplied by the manufacturer) in which 7 out of 9 heaters survived cyclic test to a temperature of 1500°F, ORNL requested delivery at the maximum possible rate (50 heaters per week). However, continued problems with delivery of insulators (MgO) to the heater manufacturer have shifted the scheduled date for delivery of the final batch of heaters to late December.
- D. Work on the full-scale, half-length, 37-subassembly, sodium-cooled, electrically-heated cask mockup is being held in abeyance due to budgetary restrictions.

1.3 and 1.4 Cask Integrity Studies and Tests (Tasks 1.3 and 1.4)

- E. Additional tests were performed with energy absorbers of the tube-filled pipe design. Test specimens 3 in. in length with the outer envelope made of 1-1/4-, 2-, and 2-1/2-in. sched 40 pipe, respectively, were deformed under the impact of drop hammers ranging in weight from 25 lb to 482 lb dropped from a height of 30 ft. Both empty and tube-filled (included 9, 22, and 33 tubes, respectively, of 3/8 in. OD, 0.065 in. thick 304 stainless steel wall) pipe specimens were utilized. Results indicate that deformation of the outer envelope (pipe) accounts for approximately 5-10 per cent of the total energy absorbed by the tube-filled specimen.

Data from the above tests, together with those previously reported in ORNL-TM-2748, are being utilized as input to a structural analysis computer program developed at ORNL. In essence, the program performs a double numerical integration (trapezoidal rule) of the force-time data which are recorded during impact testing by means of a strain gauge-load cell and oscilloscope. Output data include linear deformation (displacement) as a function of time. From the force and displacement data, absorbed energy is computed by means of a final integration. Preliminary code results have been obtained thus far; graphical representations of these data are being constructed.

- F. Cask modeling tests on impact specimens of 1/9.38 scale have been delayed sequentially by (1) unavailability of an adequate load cell, and (2) failure of the lifting mechanism (winch) at the ORNL Drop Tower Facility. Fabrication of a load cell capable of handling 2×10^6 lb has been completed

and calibration is imminent. A replacement winch (with a 3000 lb capacity) for the ORNL Drop Tower Facility has been ordered and delivery is anticipated for early November.

- G. The drop test of the 1/6 scale model cask reported during the previous work period (see ORNL-TM-2748) was repeated with the cask at the same attitude (i.e., to permit successive impact on the deformed region). A subsequent He leak check indicated that no additional leakage occurred through the Grayloc type seal as a result of the second consecutive drop test.

Fabrication was initiated on a housing for a 1 in. diam Parker metal seal which will be incorporated into a 1/6 scale model cask in a fashion similar to that used previously for Grayloc type seals (see Fig. 1 of ORNL-TM-2552). Drop tests with the latter cask (housing a Parker type seal) are planned together with additional casks utilizing Grayloc type seals following installation of a replacement winch at the ORNL Drop Tower Facility.

Work during the following report period will continue along the same general lines. Additional test data on post-test seal integrity will be obtained from 1/6 scale model casks as well as impact deformation data from cask modeling tests. Analytical investigations of heat dissipation for different types of reactor fuel subassemblies will continue.

2. RECEIVING AND STORAGE (TASK 2) (A. R. Irvine and C. D. Watson)

This task is concerned with the means for rapid, effective economical, and safe operation of receiving and storage facilities for LMFBR fuels. The character of the work to be performed under this task will be determined largely by the outcome of investigations performed for the shipping and for the head-end processing tasks. Conversely, these other two tasks will be required to take into consideration the effect of variables in their area on the task of receiving and storage. This work will take cognizance of related work on fuel handling and sodium removal that will be performed under Elements 3 and 5, respectively, of the LMFBR program plan.

The work done on this task is coincidental with the work on receiving and storage at an existing reprocessing facility, which is reported under Task 13.1.

3. HEAD-END PROCESSING OF LMFBR FUELS (TASK 3) (C. D. Watson)

The objective of this task is to develop economic head-end processing steps, in preparation for Purex recovery methods, for long- and short-decayed fuels.

In general all experimental work is being deferred pending completion of a Head-End Engineering Evaluation Study which is now under way.

In this report period: Equations for calculating new black body view factors for HEX were completed and preliminary calculations begun; design drawings of mechanical head-end concepts and experimental equipment were cataloged for use some months hence; and, a melt decay run was made using crushed UO_2 .

Reportable accomplishments include:

3.1. Decay Heat Dissipation (Task 3.1) (R. L. Cox)

New and more accurate black body and gray body view factors are being calculated for inclusion in an ORNL report on the computer code, HEX. Hottel's crossed-string method¹ was used to derive equations for the black body view factors or configuration factors for radiation exchange between a rod and each of the rods in the first three rows surrounding it in an array of infinitely long parallel cylinders on a triangular pitch. The equations are presented below. Please refer to Fig. 3-1 where F_{ij} is the fraction of the radiation emitted from body "i" which reaches body "j" by direct radiation and PDR is the pitch to diameter ratio of the rods.

$$F_{11} = 0, \text{ all PDR} \quad (1)$$

$$F_{12} = \frac{1}{\pi} \left(\frac{\pi}{6} + \tan^{-1} \sqrt{(\text{PDR})^2 - 1} - \sqrt{(\text{PDR})^2 - 1} \right), \quad (2)$$

$$1 \leq \text{PDR} \leq 2/\sqrt{3}$$

$$F_{12} = \frac{1}{\pi} \left(\frac{\pi}{2} - \text{PDR} - \tan^{-1} \sqrt{(\text{PDR})^2 - 1} + \sqrt{(\text{PDR})^2 - 1} \right), \quad (3)$$

$$\text{PDR} \geq 2/\sqrt{3}$$

ORNL DWG 69-12125

Fig. 3-1 Designation of Rods in Configuration Factor Determinations.

$$F_{13} = \frac{1}{\pi} \left(\sqrt{(\text{PDR})^2 - 1} - \tan^{-1} \sqrt{(\text{PDR})^2 - 1} \right), \quad (4)$$

$$1 \leq \text{PDR} \leq 2/\sqrt{3}$$

$$F_{13} = \frac{1}{\pi} \left[\sqrt{3(\text{PDR})^2 - 1} - \tan^{-1} \sqrt{3(\text{PDR})^2 - 1} \right. \\ \left. - 2 \left(\sqrt{(\text{PDR})^2 - 1} - \tan^{-1} \sqrt{(\text{PDR})^2 - 1} \right) - \frac{\pi}{6} \right], \quad (5)$$

$$2/\sqrt{3} \leq \text{PDR} \leq 2$$

$$F_{13} = \frac{1}{\pi} \left(\frac{\pi}{2} - \sqrt{3} \text{PDR} - \tan^{-1} \sqrt{3(\text{PDR})^2 - 1} + \sqrt{3(\text{PDR})^2 - 1} \right), \quad (6)$$

$$\text{PDR} \geq 2$$

$$F_{14} = 0, \text{ all PDR} \quad (7)$$

$$F_{15} = 0, \quad 1 \leq \text{PDR} \leq 2/\sqrt{3} \quad (8)$$

$$F_{15} = \frac{1}{2\pi} \left[\sqrt{7(\text{PDR})^2 - 1} - \tan^{-1} \sqrt{7(\text{PDR})^2 - 1} \right. \\ \left. - 2 \left(\sqrt{3(\text{PDR})^2 - 1} - \tan^{-1} \sqrt{3(\text{PDR})^2 - 1} \right) + \sqrt{(\text{PDR})^2 - 1} \right. \\ \left. - \tan^{-1} \sqrt{(\text{PDR})^2 - 1} + \tan^{-1} \sqrt{\frac{2}{9}} - \frac{\pi}{6} \right], \quad (9)$$

$$2/\sqrt{3} \leq \text{PDR} \leq 2$$

$$F_{15} = \frac{1}{\pi} \left(\sqrt{7(\text{PDR})^2 - 1} - \tan^{-1} \sqrt{7(\text{PDR})^2 - 1} - \sqrt{3(\text{PDR})^2 - 1} \right. \\ \left. + \tan^{-1} \sqrt{3(\text{PDR})^2 - 1} - \sqrt{(\text{PDR})^2 - 1} + \tan^{-1} \sqrt{(\text{PDR})^2 - 1} \right. \\ \left. - \frac{\pi}{3} \right), \quad (10)$$

$$2 \leq \text{PDR} \leq 2\sqrt{21}/3$$

$$F_{15} = \frac{1}{\pi} \left(\frac{\pi}{2} - \sqrt{7} \text{PDR} - \tan^{-1} \sqrt{7(\text{PDR})^2 - 1} + \sqrt{7(\text{PDR})^2 - 1} \right), \quad (11)$$

$$\text{PDR} \geq 2\sqrt{21}/3$$

$$F_{16} = 0, \text{ all PDR} \quad (12)$$

Some preliminary calculations were made and compared to black body view factors using the old method and there were significant differences. The next step will be to use the configuration factors given by the equations to calculate the gray body interchange factors for this geometry.

The computer code ORIGEN² was used to calculate actinide and fission product concentrations and heat generation rates as a function of decay time for spent fuel discharged from the proposed Atomics International follow-on reactor. Calculations were performed for each of the regions in the reactor (inner core, outer core, axial blanket, inner and outer radial blankets) as well as for a homogenized discharge from the reactor based on the refueling schedule. Summary results for the homogenized case are reported in Irradiation Studies Section 11, Subsection 11.2 (Task 11.2).

3.2 Dismantling of Multitubular Assemblies (Task 3.2) (G. A. West, R. S. Lowrie)

All experimental work for dismantling LMFBR fuel assemblies has been deferred pending the completion of Head-End Engineering Evaluation Studies. A listing of both the concept and experimental equipment drawings that have been generated for dismantling studies is presented for reference purpose.

- A. Abrasive sawing and slitting concepts for removing the fuel assembly end-sections and longitudinal slitting of the sheath to rid the downstream processing of the massive waste metal. The modifications and clamping fixtures have been completed and the prototype sheath and assembly are essentially complete.

- M-12200-EK-011-D Modification to saw to adapt to 25-in. blades.
- 012-D Holding fixture for hex. assemblies.
- 013-D Prototype sheath for hex. core assembly.
- 014-D Prototype hex. assembly.
- 015-D Prototype hex. sheath for blanket assembly.

- B. In-cell fuel assembly dismantling unit concept of receiving the fuel from storage and removing excess hardware and freeing the fuel rods for single or multi-rod shearing. No fabrication or experimental work is now planned on this unit during this fiscal year.

- M-11244-EL-100-E Dismantling Unit - Assembly
- 101-E Dismantling Unit - End elevation
- 102-E Dismantling Unit - Side elevation
- 103-E Dismantling Unit - Saw, hold-down clamp concept
- 104-E Dismantling Unit - Saw-blade changer concept
- 105-E Dismantling Unit - Dual slitting saw concept
- 106-E Dismantling Unit - Dual slitting saw details

3.3 Shearing (Task 3.3) (G. A. West, R. S. Lowrie)

The experimental work for shearing LMFBR type fuels has also been deferred pending completion of the Head-End Engineering Evaluation Studies. A listing of conceptual and experimental equipment drawings that have been generated for an investigation of shear design parameters follow:

- A. ORNL 250-ton shear modifications for intact shearing of LMFBR fuel assemblies. New shear blades were designed and shear stroke increased 2-5/16-in. for a 12-1/2-in. total stroke.
- M-11244-EL-052-D Shear Blade - "Interrupted Sharks Tooth"
 - 053-D Shear Blade - "Straight Sharks Tooth"
 - 054-D Shear Blade - "Elongated Stepped Tooth"
 - 055-D Shear Blade - Shear Gagging Mechanism
 - 056-D Shear Fixed-Blade Holder and Gag Guide Concept
 - 057-D Shear - Traveling Knives
 - 058-D Shear - Piston Bushing and Key (completed)
 - 059-D Shear - Fixed Knives
 - 060-D Shear - Fixed Knife Drill Jig (completed)
 - 061-D Shear - Fixed Knife Liners
 - 062-D Shear - Traveling Knife Liners
 - 063-D Shear - Fixed Knife and Liner Holder
 - 064-D Shear - Side (Feed) Plate Modifications
 - 065-D Shear - Single Gag for 5.450-in. hex Fuel
 - 066-D Shear - Inner and Outer Gags for 5.450-in. hex Fuel
 - 067-D Shear - Feed Magazine for 5.450-in. hex Fuel
- M-11244-EK-011-D Gag Compression Test Fixture
- 012-D Prototype LMFBR hex Fuel Test Assembly
 - 013-D Prototype LMFBR hex Fuel Assembly
 - 014-D Shear - Feed Pusher Head
- B. Multi-rod shear designed for shearing 25 to 100 LMFBR core fuel rods. All items have been completed except for DWG-295-D (Kick-back Test Installation).
- M-11244-EJ-210-D Shear - Assembly, "5 x 5 Tube Bundle"
 - 211-D Shear - Housing Detail
 - 212-D Shear - Knife Holder
 - 213-D Shear - Hyd. Cylinder and Misc. Details
 - 214-D Shear - Misc. Details
 - M-11244-EK-015-D Shear - Electrical Control Wiring Diagram
 - M-11244-EJ-225-D Shear - Installation
 - 226-D Shear - Support Details
 - 227-D Shear - Feed Magazine and Push Bar
 - 291-D Shear-Hyd. Cylinder Modifications and Support Bracket

- 292-D Shear - Double Solenoid Control Valve Installation
- 293-D Shear - Hydraulic Supply Line Modifications
- 294-D Shear - Hydraulic Piping Installation Support Detail
- 295-D Shear "Kick-back Test Installation

C. Single-rod Experimental Shear. Fabrication, installation and testing has been deferred until the end of this fiscal year.

- M-12200-EK-100-E Experimental Shear - Assembly
- 101-D Experimental Shear - Spring and Misc. Detail
- 102-D Experimental Shear - Shaft, Frame and Misc. Detail
- 103-D Experimental Shear - Blade, Holder and Misc. Detail
- 104-D Experimental Shear - Support Stand
- 105-D Experimental Shear - Load-cell, Bracket and Misc. Detail
- 106-D Experimental Shear - Alternate Assembly and Details

D. Miscellaneous - (Rotary Leacher). Fabrication, installation and testing has been deferred indefinitely.

- M-11244-EL-046- Rotary Leacher Assembly
- 047- Rotary Leacher - Outer Shell Detail
- 048- Rotary Leacher - Inner Spiral and Drum Detail
- 049- Rotary Leacher - Misc. Details
- 050- Rotary Leacher - Support Stand
- 051- Rotary Leacher - Misc. Details and Parts List

3.7. Alternative Head-End Process (Task 3.7) (S. D. Clinton, A. R. Irvine)

Melt-decladding is being considered as a backup process for the shear-leach head-end. By heating the fuel assemblies above the melting point of stainless steel (1450°C) it may be possible to volatilize iodine, xenon, krypton, and tritium and to separate the molten cladding from the urania-plutonia core material.

A test was made in an induction furnace at 1460°C with urania pellets and fragments enclosed in a 5-in. length of stainless steel tubing. Two urania pellets were placed at each end of the specimen, and the center 2-in. section of tubing was filled with 10.9 g of UO_2 fragments in the size range of 300 to 600 microns. The specimen was inserted in a 2.5-in.-diam cylindrical alumina crucible inclined at an angle of 10 degrees with the horizontal. About 75 wt % of the stainless

steel (total of 38 g) separated from the urania and collected at the lower end of the crucible, and there was no apparent indication that urania fragments had been carried by the molten steel. The urania fragments were either fused together or impregnated with molten steel during the run and remained between the pellets as a cylindrical shape. A better understanding of the results of this run will have to await chemical analysis.

Analytical results from a previous melt declad experiment,³ in which nine specimens containing urania pellets were heated to 1540°C, indicated a total UO_2 loss of 0.017 wt % to the separated stainless steel. The original specimens consisted of 388 g of UO_2 pellets and 243 g of steel. About 183 g (~ 75%) of the molten steel flowed away from the pellets which were resting on a 10 degree incline. Essentially all of the UO_2 loss was determined to be at the upper end of the separated steel with the lower portion (172 g) of steel containing less than 1 ppm of uranium.

During the next month, one or two melt declad runs will be made with stainless steel pins containing crushed and sized urania fragments as a further check on the feasibility of separating molten cladding from oxide fuels under adverse conditions.

References

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2. W. E. Unger, et al., Aqueous Processing of LMFBR Fuels, Progress Report, April 1969, No. 2, ORNL-TM-2585, p. 32.
3. LMFBR Fuel Cycle Studies Report for September, No. 7, ORNL-2748.

4. VOLATILE FISSION PRODUCT REMOVAL (TASK 4) (D. J. Crouse, C. D. Watson)

The objective of Task 4 is to develop a head-end processing method for removing iodine, xenon, krypton, and tritium from the fuel prior to aqueous processing. Early removal of these gases from the fuel into a relatively small volume of gas would greatly facilitate off-gas treatment. This is of particular importance with respect to iodine control because of the very high plant retention factors that will be required for ^{131}I when treating short-cooled fuels.

This month accumulation and interpretation of the data from the hot-cell tests with short-cooled LMFBR fuel rod specimens was continued. The amount of ^{85}Kr released during shearing of the fuel rods ranged from 1.28 to 1.89 mCi/g U + Pu. Up to 68% of the total ^{131}I was found in the gas plenum. From 40 to 74% of the ^{131}I in the fuel rods was removed by the voloxidation treatment, with the highest removal being obtained in a test at 650°C.

4.1 Volatilization from Oxide Fuels (Task 4.1) (V. C. A. Vaughn, J. H. Goode, L. A. Byrd, G. D. Davis, W. B. Howerton, O. L. Kirkland, R. C. Lovelace)

Additional data from hot-cell experiments on the shearing, oxidation, and dissolution of four short-cooled (32-65 days) stainless steel-clad fuel rods, containing sol-gel 15% PuO_2 --85% UO_2 microspheres, have been analyzed. Further observations on the behavior of fission products may be made as data reduction continues.

Fuel Powdering on Shearing. -- Material balances in two experiments indicated that when the 0.25-in.-diam fuel rods were sheared to 1/4-in. lengths, 73 and 75% of the fuel particles fell from the "pillowed" cladding segments. A third rod, cut to 3/4-in. lengths released only 20% of the fuel as a loose powder.

Release of Noble Gases During Shearing. -- The noble gases released on shearing the pins were carried to an evacuated gas holder by a helium purge stream. A charcoal trap was in place to catch any ^{131}I that was released at the same time. Replicate samples of the gas were counted directly for ^{133}Xe and ^{85}Kr . The charcoal trap (held at cell temperature $\sim 30^\circ\text{C}$) retained a significant fraction of the ^{133}Xe .

The quantities of noble gases released on shearing are given in Table 4-1. The data for pins 49 and 46 appear to be consistent with the anticipated flux curve shape. The ^{85}Kr value from pin 50 and the ^{133}Xe from pin 51 appear to be low. In run FR43, it is known that there was a preferential loss of ^{133}Xe from the charcoal trap; however, there is no apparent reason for the low value of ^{85}Kr in pin 50. We have not completed the analysis of noble gas releases during voloxidation and dissolution so it is not possible to report material balance checks at this time.

Table 4-1. Release of Noble Gases on Shearing
Short-Cooled LMFBR Fuel Pins

Rod No. ^a	Run No.	⁸⁵ Kr		¹³³ Xe		Ratio ¹³³ Xe/ ⁸⁵ Kr
		Conc. ^b	Relative Amt.	Conc. ^b	Relative Amt.	
49	FR45	1.28	1.00	66.2	1.00	51.4
46	FR44	1.89	1.48	96.4	1.46	51.0
50	FR46	1.56 ^c	1.22 ^c	88.2	1.33	56.5
51	FR43	1.66	1.30	57.2 ^d	0.86 ^d	34.4

^aArranged in order, top to bottom, in the irradiation holder.

^bmCi/g U + Pu

^cThis number appears low; assuming that the ¹³³Xe number is correct and a ¹³³Xe/⁸⁵Kr ratio of 51, the calculated amount of ⁸⁵Kr would be 1.72 mCi/g U + Pu and the relative amount would be 1.35.

^dAssuming the ⁸⁵Kr number is correct and a ¹³³Xe/⁸⁵Kr ratio of 51, the calculated amount of ¹³³Xe would be 84.7 mCi/g U + Pu and the relative amount would be 1.28.

Iodine Release During Shearing. - Iodine releases during shearing were small (~0.5%) and of the same magnitude as the fraction of fuel powder left in the shear bowl. This indicates that the iodine was not released preferentially to the gas phase.

Iodine Release During Voloxidation. - During irradiation some of the ¹³¹I escapes to the gas plenum, while the remainder is left within the fueled section of the rod. In the tests described below, the plenum with its iodine content was excluded from the voloxidizer experiments. The sheared fuel sections of each rod were split into two representative fractions, each containing cladding and powdered fuel. Each fraction was heated to temperature in a stream of helium and then voloxidized at 450°C, 650°C, or 750°C for 4 or 8 hr in pure oxygen at 0.1 CFH. The outlet gas passed to a gas scrubber (0.1 M HNO₃--0.05 M Hg²⁺, 1 liter/hr), then to an iodized charcoal trap, and finally to a gas holder. The scrubber D.F. was about 6 x 10³, based on the ¹³¹I found on the charcoal trap and in the scrubber effluent. After voloxidation, the fuel and cladding were removed from the burner and dissolved. The burner was leached with CCl₄ and then with 4 M HNO₃--0.05 M Hg²⁺ solution.

The amount of iodine removed in the voloxidation step in these runs ranged from about 40 to 74% (Table 4-2). The results of A and B fractions (duplicate runs) were reasonably consistent. The highest removal (74%) was obtained at 650°C; only one run, however, was made at this temperature and the apparent better results as compared to those obtained at 450°C and 750°C need to be confirmed in future experiments. There was no apparent significant effect of changing the length of the shearing cut from 1/4- to 3/4-in.

The amount of ^{131}I recovered by leaching the gas plenum was 4.45, 1.46, and 5.52 Ci, respectively, for runs 43, 44, and 46, compared to ^{131}I recoveries from the total rod (including the plenum) of 7.23, 5.18, and 8.13 Ci, respectively. We expect that the iodine in the gas plenum would volatilize readily and, if the plenum sections had been given the voloxidation treatment (as would probably be done in actual processing), the percentage removal of iodine in voloxidation would have been considerably higher than the values listed in Table 4-2.

The iodine material balances for these experiments are being checked to see if any discrepancies can be located. We are also awaiting RIBDOR calculations to give us an estimate of the starting quantities of fission products including ^{131}I . We have observed a tendency for ^{131}I to remain in all pieces of equipment. There are instances in our data where some ^{131}I was held up from one part of an experiment to the following one. We will refine our techniques to minimize this carry forward in future runs.

Analysis of Equipment Requirements (S. D. Clinton)

An electrically-heated rotary kiln for investigating voloxidation process parameters on a scale of up to 5 kg of $\text{UO}_2\text{--PuO}_2/\text{hr}$ has been ordered from the C. O. Bartlett and Snow Company, Cleveland, Ohio. Delivery is scheduled for January 1970. The unit is capable of heating sheared LMFBF fuel sections to 750 to 1000°C in either an oxidizing or inert atmosphere.

Table 4.2. ^{131}I Removal by Voloxidation Treatment

Run No.	Voloxidation Conditions	^{131}I Removed by Voloxidation ^a (Ci)	Total ^{131}I Found in Voloxidation and Dissolution Steps (Ci)	^{131}I Removed in Voloxidation (%) ^b
43A	1/4-in. cuts,	0.74	1.65	44.8
43B	4 hr at 750°C	0.53	1.13	46.9
43 (Combined)		(1.27)	(2.78)	(45.6)
44A	3/4-in. cuts,	0.87	1.62	53.7
44B	8 hr at 750°C	0.98	2.10	46.6
44 (Combined)		(1.85)	(3.72)	(49.8)
46A	1/4-in. cuts,	0.72	1.57	45.9
46B	8 hr at 450°C	0.41	1.04	39.4
46 (Combined)		(1.13)	(2.61)	(43.3)
48	1/4-in. cuts, 8 hr at 650°C	1.30	1.75	74.3

^aAmount caught in scrubbers and from leaches of the equipment.

^b% of total fed to voloxidizer.

5. DISSOLVING (TASK 5)

(D. J. Crouse, C. D. Watson)

The objective of Task 5 is to ensure that LMFBR fuels can be dissolved in nitric acid with high metal recoveries. Since the dissolution characteristics of the fuels can vary widely depending on their plutonium content, method of preparation, and irradiation histories, extensive leaching data are being obtained to define the effects of the many variables. A thorough understanding of iodine chemistry in the dissolver system is needed as a guide for providing effective iodine control. The dissolver equipment must be designed and operated within rather narrow limitations imposed by criticality control and off-gas considerations. It appears that satisfactory solution of these problems can best be accomplished using a continuous dissolver and this approach is being emphasized. Evolution of a successful dissolver will require development of equipment for dependably moving the sheared stainless steel hulls and other solids through the system, and development of seals for isolating the system to prevent excessive in-leakage of diluent gases.

This month data are reported for the dissolution of irradiated uranium mononitride. Dissolution was rapid in boiling 8 M HNO_3 and recoveries of uranium and plutonium were essentially complete. Study of the solubility of niobium in nitric acid indicated that only a minor fraction of the ^{96}Nb in short-cooled fuel will be dissolved.

5.1 Dissolution Data (Task 5.1)

Dissolution of Irradiated Uranium Mononitride (M. B. Sears)

On a long-term basis nitride or carbide fuels appear to have many advantages over oxide fuel for an LMFBR. They result in much higher breeding ratios (1.43 for carbide, 1.40 for nitride vs 1.25 for oxide), and have much higher thermal conductivities, and much higher melting points.¹ Even though a carbide-fueled reactor has a slightly better breeding ratio than one fueled with nitride, preliminary data for uranium mononitride indicate that the nitride may be superior overall. Uranium mononitride is more compatible than carbide with claddings, swells less during irradiation so less void space is needed in the fuel element resulting in better heat transfer out of the element, can be operated at higher temperatures without breakaway swelling, and is easier (and presumably cheaper) to manufacture.¹ Hopefully these same properties would also apply to the mixed (Pu - U) nitride fuel needed for an LMFBR.

Our earlier studies^{2,3} showed that unirradiated uranium mononitride can be readily dissolved in nitric acid, yielding a uranyl nitrate solvent extraction feed solution. Powdered uranium mononitride samples have also been burned in oxygen for analytical purposes. Thus, the cold work did not reveal that serious difficulties would be encountered in handling nitride fuel in a conventional fuel reprocessing plant.

No published reports are available on the chemical behavior of irradiated uranium mononitride. In cooperation with the group of C. E. Lamb of the Analytical Chemistry Division, scouting tests were made on the dissolution behavior of uranium mononitride that had been irradiated to a burnup of approximately 1.8 atom %. The uranium mononitride had been prepared from the element and was in the form of small pellets weighing about 4.5 g.

There were no visible signs of reaction after 40 min exposure of the irradiated uranium mononitride to water near its boiling point. With 4 M HNO_3 there were no visible signs of reaction after 15 min at room temperature; however, after heating almost to boiling, gas was evolved slowly from the specimen. As the nitric acid concentration increased because of the preferential evaporation of water, the reaction rate increased. With 8 M HNO_3 gas evolution began at once at room temperature, but was too slow to be of practical value in fuel dissolution; after 30 min this experiment was also heated nearly to boiling. There was rapid gas evolution and crumbling of the specimen in the hot 8 M HNO_3 , yielding a very dark solution (similar in appearance to those obtained from leaching uranium carbide) and finely divided black solids that looked somewhat like graphite flakes. The solids formed a film on the walls of the beaker. Some 15.8 M HNO_3 was then added to the experiments with water and 4 M HNO_3 raising the acid concentrations above 8 M, and, after boiling approximately 1 hr, these too yielded very dark solutions and the finely divided black solids. The uranium nitride was probably destroyed by this treatment since gas evolution had ceased, but the solutions were so opaque that it was impossible to inspect visually for unreacted solids other than those on the sides of the beakers. When additional boiling in what was probably azeotropic nitric acid failed to destroy the fine black solids, the solutions were evaporated down and concentrated HClO_4 was added. The first pass with the boiling HClO_4 -- HNO_3 mixture destroyed the dark color in the solutions, but failed to dissolve all the black solids. A second treatment with boiling HClO_4 -- HNO_3 on the insoluble residue alone completed the dissolution of the sample.

Only one quantitative experiment has been made. In this test a 2-g piece of the irradiated uranium mononitride was reacted with refluxing 8 M HNO_3 under a slow helium purge. Dissolution of the uranium nitride was essentially complete within 1.5 hr as indicated by the disappearance of the brown NO_2 (N_2O_4) product gas from the system; however, a black residue remained. The mixture was allowed to reflux for an additional 15 minutes after all the NO_2 had been flushed from the system by the purge gas. After cooling for 15 min, solids were removed by filtering through No. 42 filter paper. The black precipitate transferred "cleanly" (i.e., like fission products rather than graphite which had been suspected). After washing and drying, it corresponded to 0.24% of the original pellet weight and contained 0.002% of the total uranium, $\leq 0.007\%$ of the total plutonium, and 2% of the gross- γ activity. Radiochemical analysis showed that it contained 23% of the ^{106}Ru , 0.3 % of the $^{95}\text{Zr-Nb}$, and $<1\%$ of the ^{137}Cs , ^{141}Ce , and ^{144}Ce . The nitric acid solution appeared clear (through the yellow windows of the hot cell). It contained only 1.2×10^{-5} mg of ^{14}C per gram of fuel; ^{14}C is produced by the (n,p) reaction on ^{14}N . A NaCl trap on the off-gas collected 0.005% of the gross- γ activity. This

apparently was mostly spray carryover, since it was primarily nonvolatile species such as ^{95}Zr , ^{95}Nb , ^{144}Ce , and ^{137}Cs .

The only potential problem uncovered in this experiment was the sparking of the irradiated nitride. For example, 2-in.-long sparks were observed when the specimen was dropped a few inches into a 200-ml flask. Finely divided uranium nitride is known to be pyrophoric; however, we have never encountered sparking in handling +100 mesh particles of un-irradiated nitride in air. The dissolution itself proceeded smoothly. Unfortunately, it was necessary to use all the uranyl nitrate solution for analyses, so that none was left for solvent extraction tests.

Solubility of Niobium in Nitric Acid Solutions (W. Davis, Jr., A. H. Kibbey)

In processing high-burnup fuels at Dounreay, it was noticed that ^{95}Nb tended to be retained in the dissolver. The hold-up was variable and large variations in feed activity could be explained by non-uniform release of ^{95}Nb from the dissolver where it was very probable that it had not been truly in solution. As a guide to similar problems that may be encountered in LMFB processing, we are determining the solubility of niobium in feed solutions.

In the first tests, small volumes of a concentrated solution of $\text{K}_3\text{Nb}_6\text{O}_{19} \cdot 16 \text{H}_2\text{O}$ in water were added to 1, 3, and 5 M HNO_3 to precipitate niobium and the solutions were filtered. The filtrates were analyzed spectrochemically and showed niobium solubilities of 0.24, 0.42, and 1.2 mg/liter, respectively, for 1, 3, and 5 M HNO_3 . These values are much lower than those reported in the literature.⁴ Consequently, additional tests were made in which the niobium solution was added as before but the solutions containing niobium precipitate were refluxed at the boiling point for 3 days prior to cooling to room temperature and filtering. The clear filtrates, when analyzed as before, showed niobium solubilities in 1, 3, and 5 M HNO_3 of 0.30, 0.72, and 0.16 mg/liter.

The values in 1 and 3 M HNO_3 are in good agreement with the results from the initial tests but there is a significant discrepancy in the 5 M HNO_3 data. Based on the fission yield and 30-day cooling data reported previously⁵ and assuming a feed solution with 65 g of U + Pu per liter, total dissolution of the niobium would result in a concentration of about 5 mg per liter of feed solution. This greatly exceeds the above solubilities and indicates that most of the ^{95}Nb would remain in the leach residue.

References

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2. L. M. Ferris, J. Inorg. Nucl. Chem. 30, 2661 (1968).

3. M. B. Sears, ORNL, unpublished data.
4. Seidell and Linke, "Solubilities of Inorganic and Metal Organic Compounds," 4th Edition, Vol II, PP. 1181-82.
5. W. E. Unger, et al., LMFBR Fuel Cycle Studies Progress Report No. 6, ORNL-TM-2710 (October 1969).

6. FEED PREPARATION (TASK 6) (D. J. Crouse)

The aqueous feed discharged from the dissolver will contain solids (undissolved fission products, corrosion products, etc.) and will probably require clarification prior to solvent extraction. Preparation of the feed for solvent extraction also will include adjustment of the plutonium valence and the nitric acid concentration, and a treatment to remove iodine. This task also covers feed preparation for subsequent process cycles.

This month additional laboratory data were obtained on volatilizing iodine from nitric acid solutions. In addition, volatilization of as high as 99.9% of the ^{131}I from a nitric acid solution of short-cooled irradiated oxide fuel was demonstrated.

6.2 Iodine Control (Task 6.2) (G. I. Cathers, C. J. Shipman)

Volatilizing essentially all of the ^{131}I and ^{129}I from the nitric acid dissolver solution seems essential to effective iodine control in the chemical processing plant since it will make attainment of high iodine retention factors easier and simplify waste treatment.

Measurement of the partial pressure of iodine over nitric acid solutions is being attempted as the best means of evaluating the difficulty of volatilizing molecular iodine (I_2) from dissolver solution. A transpiration technique, whereby gas is bubbled at a slow rate through a test solution, is being used to measure partial pressures. The change of iodine concentration is followed by using ^{131}I tracer and measuring the activity of the solution.

It was reported previously that the decrease of iodine concentration usually follows the expected first-order relationship but that the results were not very reproducible with respect to either the magnitude of the first-order constant or to the concentration at which deviation from the first-order effect occurred. We have recently overcome this irreproducibility to a large degree by using N_2O_3 in nitrogen as the sparging gas. This apparently results in the necessary concentrations of "nitrose" gases in the nitric acid solution required to keep the iodine in the I_2 form. It has been found to give much better results than intermittent additions

of NaNO_2 solution. The N_2O_3 (partly dissociated to NO and NO_2) gives an equilibrium solution of NO , NO_2 , N_2O_4 , and HNO_2 . Nitrogen is being used to avoid the oxidation effect of oxygen in air, and the chain of reactions accompanying the reaction of NO with O_2 .

First-order transpiration rate constants of 0.0037, 0.0066, 0.0090, and 0.013 have now been determined, respectively, at 40, 50, 60, and 70°C. These were measured using 22 ml/min N_2O_3 and 45 ml/min N_2 with 250 ml 4 M HNO_3 containing 5×10^{-4} M I_2 . The constants at 70°C for 1, 4, and 8 M HNO_3 were respectively 0.014, 0.013, and 0.0032. These results indicate generally that iodine volatilizes quite readily even at a temperature as low as 40°C. Also, 1 and 4 M HNO_3 solutions do not behave very differently, in contrast to 8 M HNO_3 where the I_2 was evidently complexed to some degree.

One run was made with N_2O_3 sparging at 70°C; the iodine was first heated in 16 M HNO_3 to convert it to iodate or periodate before dilution to 4 M HNO_3 . A first-order constant of only 0.002 was measured, showing definitely that the higher valence forms of iodine are difficult to reduce with nitrite to the volatile I_2 state.

Conversion of these first-order constants to partial pressures requires knowledge of the partial pressure of water over these solutions and of the equilibrium volume of "nitrose" gas generated by use of N_2O_3 . This work will be carried on as part of the program. The question of why non-linearity from the first-order effect occurs at some point in every transpiration experiment (but not always at the same I_2 carrier concentration) also needs further work. Some spectrophotometer runs are being planned to confirm that following the radioactivity of the solution is giving correct information. There is much evidence in the literature concerning the difficulty of exchanging ^{131}I with carrier iodine, and this could be the reason for some of the deviations. A calibration run on a spectrophotometer showed that Beer's Law (absorbance proportional to concentration) was obeyed quite closely at 467 mμ.

Removal of Iodine from Solutions of Irradiated Fuel (J. H. Goode, V. C. A. Vaughn, L. A. Byrd, G. D. Davis, W. B. Howerton, O. L. Kirkland, R. C. Lovelace)

Two experiments were made to test the removal of ^{131}I from nitric acid feed solutions derived by leaching short-cooled LMFBR fuel specimens. The oxide fuel was irradiated to about 20,000 Mwd/ton and had been given the voloxidation treatment (450 to 750°C) prior to dissolution. The composited dissolver solution was 7 M in HNO_3 and contained 85 g of U, 13 g of Pu, and about 0.1 Ci of ^{131}I per liter. The iodine removal tests were made about 75 days after discharge of fuel from the reactor and several weeks after dissolution of the fuel.

Sparging a 100-ml aliquot of solution for 6 hr at 70°C with 150 cm³ min⁻¹ of air volatilized only about 4% of the ^{131}I . Continuing the sparging of this solution after addition of a large amount (0.1 M) of KI volatilized 99.7% of the remaining iodine. The subsequent addition of

NaNO_2 (to 0.1 M) and sparging volatilized another 0.2%, giving a total removal of 99.9%. These results suggest that most of the ^{131}I in the aged dissolver solution was present in the non-volatile iodate form.

In the second test, NaNO_2 (0.01 M) was added to a 100-ml aliquot of dissolver solution. Air sparging at 70°C for 6 hr removed essentially no ^{131}I . When KI (0.01 M) was then added to the solution and sparging at 70°C was continued for 6 hr, about 90% of the ^{131}I was volatilized.

To produce a solvent extraction feed 3 M in HNO_3 and containing 95 g of U and 20 g of Pu per liter, the dissolver solution subsequently was evaporated to about 25% of its original volume (~126°C), and then diluted with 0.5 M HNO_3 to the correct heavy metal concentrations. The distillation step volatilized 75% of the ^{131}I present in the solution, yielding a feed containing about 0.01 Ci ^{131}I per liter (90 days decay).

7. SOLVENT EXTRACTION (TASK 7) (D. J. Crouse, C. D. Watson)

The objective of Task 7 is to establish that LMFBR fuels can be processed successfully by solvent extraction methods. Initial emphasis is on development of solvent extraction flowsheets suitable for interim processing of LMFBR fuels in existing plants. Present Purex flowsheets are being modified where necessary to provide for the high plutonium content of these fuels. The wide solvent extraction experience accumulated at production sites is being assessed and factored into these studies. Particular emphasis is being given to experimental evaluation of iodine behavior in solvent extraction and to the effect of solvent damage on process performance, especially when processing short-cooled fuels. Different solvent extraction contactors are being evaluated with respect to their relative merits for processing LMFBR fuels.

This month preliminary data are presented for the cyclic solvent extraction tests made with feed solutions derived from LMFBR fuel specimens that had been irradiated to 20,000 Mwd/ton and cooled about 90 days. Plutonium recoveries in extraction were >99.9% and phase separation was good and did not change significantly with cycling or with change in the solvent radiation exposure. There was considerable retention of plutonium and uranium by the solvent after cycling in one test but this may represent a spurious result. Laboratory studies of the degradation of secondary amines and the rate of degradation of TBP were continued.

7.2 Solvent Stability (Task 7.2) (J. H. Goode, V. C. A. Vaughn, L. A. Byrd, G. D. Davis, W. B. Howerton, O. L. Kirkland, R. C. Lovelace)

Data on the two cyclic batch cross-current solvent extraction experiments that we performed with a solution of PuO_2 -- UO_2 fuel irradiated to

about 20,000 Mwd/ton and cooled 90 days are still incomplete. In these tests the 15% TBP--85% n-dodecane solvent was contacted with feed under conditions that gave an estimated radiation dose to the solvent of 0.15 whr liter⁻¹ cycle⁻¹ in Run 1 and 0.45 whr liter⁻¹ cycle⁻¹ in Run 2; the different solvent exposures between runs resulted from a change of the extraction contact time from 13 min in the first run to 39 min in the second. The solvent was then scrubbed, stripped, and washed (twice with 0.1 M Na₂CO₃ and once with 3 M HNO₃) before being subjected to the next extraction cycle (Table 7-1). This was continued for four cycles to give cumulative solvent exposures of 0.60 and 1.84 whr/liter, respectively, in Runs 1 and 2. The aqueous phase from the first extraction contact was contacted with six successive volumes of fresh solvent to complete the recovery of plutonium.

The fission product power level of the feed solution was calculated to be 2.13 $\beta + \gamma$ watts/liter. This calculation is based on the actual β and γ radiochemical spectra and average energies of the individual major fission products. This agrees with the value of about 2 watts/liter obtained by calorimetric measurements. The higher value estimated last month (2.85 watts/liter) had been calculated from the gross beta and gross gamma counting rates and assuming an average energy of 0.5 Mev.

The available data indicate:

- (1) Uranium losses to the raffinate after seven extraction contacts were less than 0.001% in all cycles of each run. The plutonium losses to the raffinate in the first run ranged from 0.02 to 0.03%; in the second run the plutonium loss increased from about 0.01% in the first cycle to 0.05% in each of the next three cycles.
- (2) In the first run, losses of uranium and plutonium to the stripped solvent were <0.02% in the first cycle; because of insufficient sample volumes, these losses could not be determined for the remaining three cycles. Based on analysis of the washed solvent following the fourth cycle, however, there was no significant retention of plutonium and uranium by the washed solvent. In the second run, losses of plutonium to the stripped solvent were low (<0.02%) in the first two cycles. However, in cycles 3 and 4, the losses increased to 0.04% and 0.43%, respectively. Uranium losses to the stripped solvent were low (<0.02%) in the first three cycles but increased to 0.35% in the last cycle. The high retention of plutonium and uranium by the solvent in the last cycle is yet unexplained. The carbonate wash treatments were not effective in removing the retained plutonium and uranium from the solvent, indicating that the metal retention was caused by diluent degradation products rather than by dibutyl phosphoric acid (degradation product of TBP). It is not clear how such a relatively large amount of degradation product could have been formed in the last cycle; conceivably some contamination of the system occurred.

- (3) Fission product decontamination factors were generally obscured by ^{233}Pa which extracted and stripped with the uranium and plutonium. The ^{233}Pa was accidentally introduced into the fuel column by thermal diffusion of ThO_2 from the insulator pellets when the temperature of the fuel rods reached a higher temperature than expected during irradiation.¹ Decontamination from $^{140}\text{BaLa}$ and rare earths appeared to be on the order of 10^3 , or about that achieved in similar tests last year. We are obtaining selected RCA analyses to determine other fission product DF's.
- (4) Phase separation times for the extraction, scrubbing, and stripping contacts did not change significantly with cycling or with the level of radiation exposure:

<u>Operation</u>	<u>Phase Separation Time (sec)</u>
Extraction	44 ± 4
Scrubbing	23 ± 2
Stripping	29 ± 4

Table 7-1. Solvent Extraction Experimental Conditions^a

Feed (1.0 volume):	3 M HNO_3 ; 95 g U/liter, 20 g Pu/liter, 1.8×10^{11} Gr β c/m/ml, 1.26×10^{11} Gr γ c/m/ml.
Scrub (0.3):	2 M HNO_3 , 21 g U/liter, 5 g Pu/liter
Solvent (1.8):	15% TBP in <u>n</u> -dodecane
Strip (0.9):	0.10 M HNO_3
Solvent Wash (0.45):	0.1 M Na_2CO_3 (twice) 3.0 M HNO_3 (once)

^aOne complete solvent cycle consisted of one extraction contact, four scrub contacts, seven strip contacts, and the solvent wash.

Decomposition of TBP and Precipitation of Butyl Phosphates in Aqueous Nitric Acid Solutions (W. Davis, Jr., A. H. Kibbey)

In a one- or two-phase system composed of water, nitric acid, and tributyl phosphate, the latter decomposes into dibutyl-, monobutyl-, and ortho-phosphoric acids. The extent of this decomposition is a function of

concentrations, temperature, and time. If zirconium nitrate is added to such a system, the zirconium will form complexes with the phosphates. Depending upon the quantities of these complexes, they will either be dissolved in the TBP phase, if this is present in large amount, or will precipitate as a white solid, if very little TBP is present. Such precipitation apparently does occur in existing Purex process solvent extraction plants. We have been primarily interested in determining the rate of decomposition of TBP in the presence and absence of radiation in the absence of zirconium, on the one hand, and of precipitation of zirconium butyl phosphates in the aqueous phase, on the other, at temperatures that might exist in process solvent extraction equipment, since there is so little information in the literature concerning these aspects of fuel processing. To keep the aqueous phase saturated with TBP, we have maintained enough TBP to form a second phase, typically 1 ml TBP/300 ml aqueous solution.

According to data summarized by Burger,² the rate constant for TBP decomposition in the aqueous phase is about tenfold higher than the constant for decomposition in the diluent-free, equilibrium TBP phase for aqueous 1 to 5 molar solutions of nitric acid. However, since the concentration of TBP dissolved in the aqueous phase is only about 10^{-3} M while that in the organic phase is about 3.5 M, the quantity of TBP decomposed per unit volume of TBP is about 350 times the quantity of TBP decomposed per unit volume of aqueous phase. Thus, in our experiments there is about as much decomposition of TBP in the 1 ml of TBP phase as in the 300 ml of aqueous phase.

Hardy and Scargill³ have shown that the distribution coefficient of HDBP between 1 to 3 M HNO_3 solution and the diluent-free TBP phase favors the TBP phase to the extent $D_{\text{HDBP}} \approx 100$. Therefore, in a two-phase system consisting of 1 ml of TBP and 300 ml of aqueous nitric acid, about 1/4 of the HDBP formed by TBP decomposition will be in the 1 ml of TBP phase and about 3/4 will be in the aqueous phase. The net result is, therefore, a concentration of HDBP in the aqueous phase about 1.5 times as great as would be observed if the experiment were performed with no organic phase present (but with the aqueous phase kept saturated with TBP). This bias of a factor of 1.5 is well within the limits of uncertainty of our values and of literature values on TBP decomposition rates.

We have now completed experiments on the decomposition of TBP, while exposed to ^{60}Co gamma-radiation, in 1, 3, and 5 M HNO_3 at 50°C. The absorbed γ -ray power density in the 3 M HNO_3 solution was 0.0334 w/liter; that in the recently completed 5 M HNO_3 experiment was 0.0278 w/liter. Within a factor of 2, there was no difference in the rates of TBP decomposition in the presence or absence of radiation at these relatively low irradiation levels. For comparison, Groenier⁴ has estimated an aqueous phase power density of solutions prepared from 30-day-cooled, reference AI-LMFBR fuel is 3.5 and 10 w/liter, respectively, for feeds containing 65 and 200 grams per liter of U + Pu. Dilution of this power density by the addition of 2 or 2.5 volumes of solvent and 0.3 volume of scrub solution for each volume of feed reduces this power density to 1.4 to 3.4 w/liter. This range is 40 to 120 times that of the system we are now using.

Chromatographic Studies of Solvent Degradation Products (J. G. Moore)

We are continuing our investigation of the degradation of secondary amines in nitric acid systems. Gas chromatographic studies were started using the degraded solutions of ditridecylamine (DTDA) in diethylbenzene (DEB) previously described.¹ The initial chromatograms were made using temperature programming from 60 to 270°C and a column 4 ft long and 2 mm in diameter containing 0.3% Apiezon L on 100 to 120 mesh Corning GLC 110 Textured Surface Glass Beads. The feed solution (0.3 M DTDA in DEB) showed three peaks, two for diethylbenzene with a retention time of 2 and 3 1/2 min and one peak with a retention time of about 32 min due to the amine. After contacting the solution 4 hr at 40°C with 7 M HNO₃ containing 0.05 M NaNO₂, a chromatogram showed the same peaks at 2, 3 1/2, and 32 min plus a series of small ones at 7 to 18 min and a medium sized peak at 38 min. By comparison with a chromatogram of DEB that had contacted the same nitric acid-nitrite solution, it was found that the small peaks at 7 to 18 min resulted from the DEB and made up about 0.5% of the total peak area of the DEB. The 38-min peak resulted from amine degradation and comprised about 23% of the total peak area of the DTDA. Additional studies are underway to determine if these peaks can be broken down further by using different stationary phases on the glass support.

The tertiary amine content of amine mixtures are commonly determined by titrating aliquots of the mixture before and after reacting with acetic anhydride. Primary and secondary amines react with the anhydride whereas the tertiary does not:

Preliminary chromatographic studies with amine mixtures indicate that acetic anhydride may be used to differentiate peaks due to primary and/or secondary amines from those due to tertiary amines by injecting an aliquot of the anhydride along with the amine mixture. Chromatograms were made of a benzene solution containing diethylbenzene plus heptadecyl-, di(tridecyl)-, and tri(n-octyl)amines with and without acetic anhydride present. The peaks due to the heptadecyl and di(tridecyl) amines were displaced about 5 min when acetic anhydride was present. There was no change in the position or shape of the peak due to the tertiary amine.

References

1. W. E. Unger, et al., LMFBR Fuel Cycle Studies Progress Report for September, No. 7, ORNL-TM-2748.
2. L. L. Burger, The Chemistry of Tributyl Phosphate, A Review, HW-40910 (Oct. 27, 1955).
3. C. J. Hardy and D. Scargill, "Studies on Mono- and Di-n-Butylphosphoric Acids-II," J. Inorg. Nucl. Chem. 11, 128 (1959).
4. W. E. Unger, et al., Aqueous Processing of LMFBR Fuels, Progress Report, March 1969, No. 1, ORNL-TM-2552.

8. PLUTONIUM PURIFICATION (TASK 8)
(D. J. Crouse)

This task covers the process steps in purifying plutonium, starting with the plutonium product solution from the first Purex cycle and continuing through preparation of a product solution of adequate purity and concentration for delivery to the fuel refabrication operation.

Plutonium chemistry is reasonably well-known although the high plutonium content of LMFBR fuels makes some process revisions desirable. The large amounts of plutonium to be handled provide strong incentive for improving the efficiency of the plutonium purification processes since the cost of these operations will represent a much larger fraction of the total reprocessing costs than previously. In particular there appears to be a need for a higher capacity process than ion exchange for final decontamination of the plutonium.

No reportable progress was made on this task this month.

9. WASTE TREATMENT AND STORAGE (TASK 9)

Progress on this task is reported separately.

10. OFF-GAS TREATMENT (TASK 10)

(D. J. Crouse, C. D. Watson)

Retention of iodine is the major problem in treating the off-gas from processing of short-cooled LMFBF fuels since plant retention factors of 10^7 to 10^8 would be required. Retention of most of the xenon and krypton, and eventually tritium, also may be required for future large processing plants. Logic diagrams illustrating alternative processing methods, based on present knowledge, for removing the gases from off-gas streams were presented previously.¹ All promising methods will be evaluated and additional chemical and engineering data developed where necessary. Studies are active in the areas of (1) scrubbing iodine species from gas streams with mercuric nitrate--nitric acid and alkaline solutions, 2) decomposition of organic solvent vapors and organic iodides by catalytic oxidation, and (3) testing of solid iodine adsorbents.

This month it was shown that passing air containing dodecane vapor through a glass tube at 350°C completely eliminates the dodecane but results in some formation of intermediate organic products. With Hopcalite oxidation catalyst present at 350°C, the dodecane is all converted to carbon dioxide and water. In a 30-hr run at 100°C on trapping methyl iodide in a 2-in. bed of silver zeolite, the trapping efficiency was high (>99.9%) even though the bed loading reached 7 mg of iodine per gram of silver zeolite. An iodine DF of $>10^4$ was obtained for scrubbing elemental iodine from air with mercuric nitrate solution in a bubble column with about 37-in. depth of liquid.

10.1 Iodine (Task 10.1)

Catalytic Oxidation of Organic Vapors (G. I. Cathers, M. R. Bennett)

Work on the removal of organic material from air by oxidation over Hopcalite catalyst was continued. Attention was given particularly to the question of whether some low molecular weight organic oxidation products might be found in addition to CO_2 and H_2O . This is of interest in evaluating whether changes in catalyst surface area might affect the catalyst performance.

Tests were made at different residence times at 350°C with Hopcalite catalyst having a degraded surface area of only about 12 m²/g. Air saturated with dodecane at room temperature was used as the test gas. Carbon dioxide analyses, used to measure the completeness of combustion, indicated that residence times of as little as 0.02 sec (1/10 of normal time) resulted in essentially complete combustion. This raised the question of whether oxidation of 100 to 200 ppm dodecane carried in air would proceed at 350°C in the absence of a catalyst. The literature indicates that hydrocarbon oxidation reactions proceed at macro concentrations at temperatures well below 350°C.²

A series of dodecane oxidation runs at several temperatures ranging from 130 to 350°C confirmed that rapid oxidation without Hopcalite catalyst does occur at the upper part of this temperature range. These tests were of 6-hr duration at an air flow rate of 50 ml/min with the unoxidized dodecane being trapped in a dry ice bath and then analyzed by gas chromatography. At 350°C complete disappearance of the dodecane was observed; at 300°C the percent oxidation was calculated to be 25 to 66%, depending on the value assumed for the saturation concentration of dodecane in air (i.e., the concentration of dodecane in the air entering the oxidation system). At temperatures below 300°C, the results were also ambiguous due to the uncertainty in the saturation value.

Another instrumental method was used to check the above observations. A Hydrocarbon Analyzer, made by Beckman and having a high sensitivity flame ionization detector, was connected to the test equipment to monitor the unoxidized organic material in the air stream. The response of this instrument is proportional to carbon atom input in organic compounds; it does not react to CO_2 . The test results generally confirmed the gas chromatographic results; however, there was an important distinction. The Hydrocarbon Analyzer indicated 89% oxidation of dodecane at 350°C with no catalyst present whereas the gas chromatographic method had indicated 100%. The distinction between the two methods is that gas chromatography measures the dodecane specifically whereas the Hydrocarbon Analyzer also measures intermediate organic oxidation products. Using a Hopcalite catalyst bed at 350°C, the Hydrocarbon Analyzer showed essentially 100% oxidation. A temperature profile run with no catalyst present indicated that about 90% of the dodecane was oxidized in the temperature range of 230 to 500°C.

On the basis of the gas chromatographic and Hydrocarbon Analyzer tests we conclude that dodecane carried at its saturation pressure in air is rather easily degraded completely in the temperature range of 300 to 500°C without any catalyst being required. Approximately 10% of the material is incompletely oxidized to CO_2 . Use of a catalyst bed results in complete oxidation of all organic material to CO_2 .

It can be speculated from the above that methyl iodide (and other organic iodides) may not be as difficult to destroy as is commonly assumed. The problem with respect to application of this system for off-gas treatment is perhaps really one of preventing elemental iodine from recombining with low molecular weight oxidation products as they emerge from the oxidation process.

Trapping of Methyl Iodide by Silver Zeolite (R. E. Adams, R. D. Ackley, Zell Combs)

Silver zeolite, which is prepared by treating Linde Molecular Sieves (Type 13X) with AgNO_3 solution, is highly effective for trapping iodine in the form of CH_3I at temperatures of 100°C and higher. One potential drawback, of course, is that this material would be expensive to produce

(due to the high price of silver). Thus, its effective capacity for ^{131}I and ^{129}I is needed to aid in evaluating its usefulness. Studies to define the capacity are described below.

In one group of tests, air containing ^{131}I -labeled CH_3I at a concentration of 3 ppm was passed through two parallel 2-in. beds of silver zeolite, one at 100°C and one at 200°C , for successive time periods of 4, 4, 4, and 18 hr. Methyl iodide trapping efficiencies corresponding to each of the time periods were calculated from gamma-counting data on the test beds and on the collection beds that followed the test beds. The results are presented in Table 10-1. No significance is presently attached to the apparent increase in trapping efficiency with time during the earlier portion of the 30-hr period. However, the slightly lower values observed for the final 18 hr of exposure are probably indicative of a lowering in efficiency with increased iodine loading of the bed; the total loading in these tests approximated 7 mg (as I) per gram of silver zeolite. Results appeared slightly better at 100°C than at 200°C .

Presently, experimental work is underway in which the effect of considerably higher methyl iodide loadings is being investigated. In addition, similar tests with other organic iodides, such as butyl iodide, are planned.

Table 10-1. Efficiency of Silver Zeolite for Trapping Methyl Iodide^a as a Function of Exposure Time

Temp. ($^\circ\text{C}$)	Face Velocity of Air (fpm)	Incremental Trapping Efficiency for a 2-in. Bed Depth (%)			
		0-4 hr	4-8 hr	8-12 hr	12-30 hr
100	50	99.99	99.99+	99.99+	99.95
200	63	99.94	99.99	99.99+	99.89

^a Methyl iodide injection rate was approximately 0.25 mg CH_3I per gram of zeolite per hr.

Scrubbing Iodine from Air with Mercuric Nitrate Solutions (D. J. Crouse, J. M. Schmitt, W. B. Howerton)

A series of four tests was made in which 100 cc/min of air containing about 1 mg of ^{131}I -labeled I_2 per liter was bubbled up through a cylinder (~1-in. diam) containing 0.1 M $\text{Hg}(\text{NO}_3)_2$ --0.1 M HNO_3 solution. The volume of solution in the cylinder in each test was changed so as to vary the path length that the bubbles traveled before they emerged from the

solution. Changing the depth of the solution above the gas dispersal point from about 6 to 37.5 in., increased the ^{131}I decontamination factor from 1.9×10^3 to 2.8×10^4 (Table 10-2). The results for Run 2 of this series appear out-of-line with the results for the other runs.

Distribution of Iodine Between Nitric Acid and Air. - The distribution of elemental iodine between 0.1 M HNO_3 solution and air was measured by adding 0.1 M HNO_3 containing ^{131}I -labeled I_2 to a large flask, shaking the flask to allow equilibration of iodine between the phases, and sampling the aqueous phase for analysis. In a series of ten measurements in which the iodine concentration in the aqueous phase at equilibrium varied from about 3×10^{-4} to 3×10^{-3} N, the measured D_G^L (ratio of ^{131}I concentration in the aqueous to its concentration in the gas phase) ranged from 73 to 89. This is the approximate value of the distribution coefficient that has been measured by others between the gas phase and acidic solutions of different composition. Similar tests are being made to measure the distribution of methyl iodide between air and 0.1 M HNO_3 . Values reported in the literature for other acidic solutions indicate that the D_G^L for methyl iodide should be in the range of 4.

Scrubbing of Methyl Iodide from Air. - Previously³ an iodine DF of about 700 was obtained by bubbling air containing methyl iodide through about a 14-in. depth of 0.1 M $\text{Hg}(\text{NO}_3)_2$ --0.1 M HNO_3 solution in the manner described above for experiments on elemental iodine removal. This test indicated that highly efficient removal of methyl iodide could be obtained easily in plant operation. Recent calculations indicate, however, that this experiment may be misleading and, rather than easy, it would be difficult to achieve a high methyl iodide DF using, for example, a packed column operated with the gas phase continuous. The efficiency of methyl iodide removal is primarily controlled by the relatively slow reaction of methyl iodide with mercuric ion in the aqueous phase to form the mercury-iodine complex. Extrapolation of the data⁴ of Bennett and Slusher indicates that the half-time for this reaction at 30°C should be in the order of 0.1 min when the mercury concentration is 0.1 M. With a half-time of this magnitude, coupled with the relatively low methyl iodide absorption coefficient (D_G^L) of about 4, high DF's across the column can be obtained (at tolerable solution pumping rates) only when $\frac{V_{LH}}{V_G^*}$ is considerably larger

than unity, where V_{LH} is the liquid volume hold-up in the column and V_G^* is the gas volume throughput over the reaction half-time. This means that, to achieve high DF's, use of either a very long column or quite low linear gas flow velocities are needed. The methyl iodide removal efficiency can be improved by increasing the mercury concentration and/or the temperature in order to increase the reaction rate in solution. Students from the MIT Practice School at ORNL will be studying this problem during the coming month with the objective of defining the range of performance that can be expected for packed columns and considering the relative merits of other types of gas-liquid contactors.

The limitations described above for methyl iodide apparently do not apply (or are much less severe) to scrubbing of elemental iodine from the gas stream. Although the rate of reaction of mercury with I_2 in the aqueous phase has not been measured, it apparently is rapid compared to its reaction with methyl iodide. In addition, the relatively high D_G^L for I_2 of about 80 (compared to 4 for methyl iodide) is important in retaining the I_2 in solution long enough for the fixation reaction to occur.

Table 10-2. Scrubbing Elemental Iodine from an Air Stream with Mercuric Nitrate Solution

Run	Run Time (hr)	Volume of Solution (ml)	Depth of Solution (in.) ^a	Decontamination Factor
1	2	88	6	1.9×10^3
2	3.5	215	16.5	1.6×10^3
3	3.5	335	27.2	1.3×10^4
4	5	463	37.5	2.8×10^4

^a Measured from the gas dispersal point to the top of the liquid during air flow.

References

1. W. E. Unger, et al., Aqueous Processing of LMFBF Fuels Progress Report March 1969, No. 1, ORNL-TM-2552.
2. W. Jost (ed.), Low Temperature Oxidation, Chapt. 8, Gordon and Breach Science Publishers, 1965.
3. W. E. Unger, et al., LMFBF Fuel Cycle Studies Progress Report No. 6, ORNL-TM-2710 (October 1969).
4. W. E. Unger, et al., LMFBF Fuel Cycle Studies Progress Report No. 5, ORNL-TM-2671 (August 1969).

11. RADIATION AND SHIELDING (J. P. Nichols and C. D. Watson)

This task consists of analytical studies with the objective of developing information on the radiation properties and shielding requirements of LMFBR fuels outside the reactor. The work involves compilation and correlation of basic nuclear data including decay schemes, nuclear cross sections, radiation sources and energy spectra, and thermal power (Task 11.1); development of computer programs for calculating transient concentrations of radionuclides in LMFBR fuel (Task 11.2); calculation of dose attenuation kerma for shield materials of interest (Task 11.3); and application of these data to LMFBR fuel cycle processes (Task 11.4).

11.2 Irradiation Studies (R. L. Cox)

The isotopic composition, activity, and rate of heat generation of fuel discharged from the AI Follow-On LMFBR were calculated for the inner core, outer core, axial blanket, and inner and outer radial blankets as well as for a blend of these regions based on the refueling schedule. The as-charged composition, specific power (based on a capacity factor of 0.8), irradiation time, and weight fractions of the as-charged reactor loading discharged from each region are given in Table 11-1. Tables 11-2 to 11-6 summarize the mass, radioactivity, and thermal power of actinides and fission products per metric ton of blended fuel at discharge (the column headed "initial" in the tables) and at decay times of 30, 90, 150, 210, and 270 days.

11.4 Shielding Calculations for Fuel Shipping Cask (E. D. Arnold, H. F. Soard)

The ANISN shielding calculations for the 36 element cask for the AI Follow-On LMFBR design have been initiated. To date we have completed the criticality calculation and the fixed plus fission source calculation. The fixed source only calculations are to be run. The calculated value of k_{eff} using the 27 neutron group-20 gamma group cross section set was 0.75 for an array containing 37 fuel elements. This value of k agrees well with results of more precise calculations presented in Section 12.2.

Additional cross section sets for Li, Mn, Cr, Ni and Cd have been generated. These cross-sections are to be used for calculating the effects of Li substitution for Na in the cavity.

Table 11-1. Data for AI Follow-on LMFBR

Region	U ²³⁵	U ²³⁸	Pu ²³⁹	Pu ²⁴⁰	Pu ²⁴¹	Pu ²⁴²	Specific Power	Irradiation Time	Discharge Weight
	Gram-atoms/Metric ton of (U + Pu) charged						MW/MT	Days	Fraction
Inner core	7.34	3635.	374.	145.	27.7	10.7	105.3	730	.2462
Outer core	6.92	3422.	516.	199.	38.0	15.1	86.7	730	.2612
Axial blanket	8.41	4193.	0	0	0	0	4.58	730	.3491
Inner radial blanket	7.67	4194.	0	0	0	0	8.28	1825	.0884
Outer radial blanket	8.21	4194.	0	0	0	0	4.06	2555	.0551

Table 11-2 Masses of Actinides

AI FOLLOW-CN HOMOGENIZED DISCHARGE FROM REACTOR									
PCWR= 51.12 MW/MT, BURNUP= 38528. MWD/MT, FLUX= 2.62E 15 N/CM**2-SEC									
NUCLIDE CONCENTRATIONS, GRAMS / METRIC TON (U + PU)									
	INITIAL	30. D	90. D	150. C	210. D	270. D			
U235	1.71E 03	1.71E 03	1.71E 03	1.71E 03	1.71E 03	1.71E 03			
U236	8.20E 01	8.22E 01	8.26E 01	8.30E 01	8.33E 01	8.37E 01			
U238	8.65E 05	8.65E 05	8.65E 05	8.65E 05	8.65E 05	8.65E 05			
NP237	1.52E 02	1.54E 02	1.54E 02	1.55E 02	1.55E 02	1.55E 02			
PU238	3.16E 01	3.32E 01	3.56E 01	3.75E 01	3.90E 01	4.01E 01			
PU239	6.51E 04	6.54E 04	6.54E 04	6.54E 04	6.54E 04	6.54E 04			
PU240	2.20E 04	2.20E 04	2.20E 04	2.20E 04	2.20E 04	2.20E 04			
PU241	3.27E 03	3.26E 03	3.23E 03	3.20E 03	3.17E 03	3.15E 03			
PU242	1.65E 03	1.65E 03	1.65E 03	1.65E 03	1.65E 03	1.65E 03			
AM241	3.15E 02	3.29E 02	3.58E 02	3.86E 02	4.14E 02	4.41E 02			
AM243	1.39E 02	1.39E 02	1.39E 02	1.39E 02	1.39E 02	1.39E 02			
SUBTOT	9.59E 05	9.59E 05	9.59E 05	9.59E 05	9.59E 05	9.59E 05			
TOTALS	9.59E 05	9.59E 05	9.59E 05	9.59E 05	9.59E 05	9.59E 05			

Table 11-3 Decay Heats of Actinides

AI FOLLOW-CN HOMOGENIZED DISCHARGE FROM REACTOR									
POWER= 51.12 MW/MT, BURNUP= 38528. MWD/MT, FLUX= 2.62E 15 N/CM**2-SEC									
NUCLIDE THERMAL POWER, WATTS / METRIC TON (U + PU)									
	INITIAL	30. D	90. D	150. C	210. D	270. D			
PU238	1.77E 01	1.86E 01	1.99E 01	2.10E 01	2.18E 01	2.24E 01			
PU239	1.24E 02	1.25E 02	1.25E 02	1.25E 02	1.25E 02	1.25E 02			
PU240	1.51E 02	1.51E 02	1.51E 02	1.51E 02	1.51E 02	1.51E 02			
PU241	1.55E 01	1.54E 01	1.53E 01	1.52E 01	1.50E 01	1.49E 01			
PU242	1.90E 01	1.90E 01	1.90E 01	1.90E 01	1.90E 01	1.90E 01			
AM241	3.41E 01	3.56E 01	3.87E 01	4.17E 01	4.47E 01	4.77E 01			
CM242	1.55E 02	1.38E 02	1.07E 02	8.27E 01	6.41E 01	4.97E 01			
CM244	2.58E 00	2.57E 00	2.55E 00	2.54E 00	2.52E 00	2.51E 00			
SUBTOT	5.01E 02	4.86E 02	4.59E 02	4.39E 02	4.24E 02	4.13E 02			
TOTALS	2.25E 05	5.20E 02	4.59E 02	4.39E 02	4.24E 02	4.14E 02			

Table 11-4 Masses of Fission Products

AI FOLLOW-CN HOMOGENIZED DISCHARGE FROM REACTOR							
POWER= 51.12 MW/MT, BURNUP= 38528. MWD/MT, FLUX= 2.62E 15 N/CM**2-SEC							
NUCLIDE CONCENTRATIONS, GRAMS / METRIC TON (U + PU)							
	INITIAL	30. D	90. D	150. D	210. D	270. D	
KR 83	7.66E 01	7.66E 01	7.66E 01	7.66E 01	7.66E 01	7.66E 01	
KR 84	1.34E 02	1.34E 02	1.34E 02	1.34E 02	1.34E 02	1.34E 02	
KR 85	3.03E 01	3.02E 01	2.98E 01	2.95E 01	2.92E 01	2.89E 01	
RB 85	5.25E 01	5.26E 01	5.30E 01	5.33E 01	5.36E 01	5.39E 01	
KR 86	1.27E 02	1.27E 02	1.27E 02	1.27E 02	1.27E 02	1.27E 02	
RB 87	1.59E 02	1.59E 02	1.59E 02	1.59E 02	1.59E 02	1.59E 02	
SR 88	2.42E 02	2.42E 02	2.42E 02	2.42E 02	2.42E 02	2.42E 02	
Y 89	2.62E 02	2.71E 02	2.82E 02	2.87E 02	2.89E 02	2.90E 02	
SR 90	3.61E 02	3.60E 02	3.59E 02	3.57E 02	3.56E 02	3.54E 02	
ZR 90	9.54E 00	1.07E 01	1.21E 01	1.36E 01	1.50E 01	1.65E 01	
ZR 91	3.64E 02	3.78E 02	3.94E 02	4.02E 02	4.06E 02	4.08E 02	
ZR 92	5.30E 02	5.31E 02	5.31E 02	5.31E 02	5.31E 02	5.31E 02	
ZR 93	6.55E 02	6.55E 02	6.55E 02	6.55E 02	6.55E 02	6.55E 02	
ZR 94	7.43E 02	7.43E 02	7.43E 02	7.43E 02	7.43E 02	7.43E 02	
ZR 95	1.17E 02	8.49E 01	4.49E 01	2.36E 01	1.25E 01	6.57E 00	
NB 95	6.25E 01	5.84E 01	3.99E 01	2.38E 01	1.34E 01	7.32E 00	
MO 95	7.79E 02	8.15E 02	8.74E 02	9.11E 02	9.32E 02	9.44E 02	
ZR 96	8.65E 02	8.65E 02	8.65E 02	8.65E 02	8.65E 02	8.65E 02	
MO 96	1.80E 01	1.80E 01	1.80E 01	1.80E 01	1.80E 01	1.80E 01	
MC 97	1.15E 03	1.15E 03	1.15E 03	1.15E 03	1.15E 03	1.15E 03	
MC 98	1.01E 03	1.01E 03	1.01E 03	1.01E 03	1.01E 03	1.01E 03	
TC 99	1.01E 03	1.02E 03	1.02E 03	1.02E 03	1.02E 03	1.02E 03	
MO100	1.18E 03	1.18E 03	1.18E 03	1.18E 03	1.18E 03	1.18E 03	
RU100	2.73E 01	2.73E 01	2.73E 01	2.73E 01	2.73E 01	2.73E 01	
RU101	9.75E 02	9.75E 02	9.75E 02	9.75E 02	9.75E 02	9.75E 02	
RU102	1.11E 03	1.11E 03	1.11E 03	1.11E 03	1.11E 03	1.11E 03	
RH103	9.54E 02	9.86E 02	1.02E 03	1.03E 03	1.03E 03	1.03E 03	
RU104	1.02E 03	1.02E 03	1.02E 03	1.02E 03	1.02E 03	1.02E 03	
PD105	6.53E 02	6.55E 02	6.55E 02	6.55E 02	6.55E 02	6.55E 02	
RU106	4.08E 02	3.86E 02	3.45E 02	3.08E 02	2.75E 02	2.45E 02	
PD106	3.89E 02	4.12E 02	4.53E 02	4.90E 02	5.23E 02	5.53E 02	
PD107	5.79E 02	5.79E 02	5.79E 02	5.79E 02	5.79E 02	5.79E 02	
PD108	3.67E 02	3.67E 02	3.67E 02	3.67E 02	3.67E 02	3.67E 02	
AG109	4.21E 02	4.21E 02	4.21E 02	4.21E 02	4.21E 02	4.21E 02	
CD110	2.40E 01	2.41E 01	2.41E 01	2.42E 01	2.42E 01	2.42E 01	
CD111	6.56E 01	7.06E 01	7.07E 01	7.07E 01	7.07E 01	7.07E 01	
CC112	4.40E 01	4.41E 01	4.41E 01	4.41E 01	4.41E 01	4.41E 01	
SB121	1.44E 01	1.44E 01	1.44E 01	1.44E 01	1.45E 01	1.45E 01	
SB125	2.06E 01	2.06E 01	1.98E 01	1.90E 01	1.82E 01	1.74E 01	
SN126	6.98E 01	6.98E 01	6.98E 01	6.98E 01	6.98E 01	6.98E 01	
I127	1.31E 02	1.33E 02	1.35E 02	1.36E 02	1.37E 02	1.37E 02	
TE128	1.88E 02	1.88E 02	1.88E 02	1.88E 02	1.88E 02	1.88E 02	
I129	3.80E 02	3.84E 02	3.88E 02	3.89E 02	3.89E 02	3.89E 02	
TE130	4.30E 02	4.30E 02	4.30E 02	4.30E 02	4.30E 02	4.30E 02	
XE130	1.30E 01	1.30E 01	1.30E 01	1.30E 01	1.30E 01	1.30E 01	
XE131	7.81E 02	7.93E 02	7.94E 02	7.94E 02	7.94E 02	7.94E 02	
XE132	1.14E 03	1.15E 03	1.15E 03	1.15E 03	1.15E 03	1.15E 03	

Table 11-4 (continued)

AI FOLLOW-ON HOMOGENIZED DISCHARGE FROM REACTOR												
POWER= 51.12 MW/MT, BURNUP= 38528. MWD/MT, FLUX= 2.62E 15 N/CM**2-SEC												
NUCLIDE CONCENTRATIONS, GRAMS / METRIC TON (U + PU)												
	INITIAL	30. D	90. D	150. D	210. D	270. D						
CS133	1.42E C3	1.45E C3	1.45E C3	1.45E C3	1.45E C3	1.45E C3						
XE134	1.61E C3	1.61E C3	1.61E C3	1.61E C3	1.61E C3	1.61E C3						
CS134	2.86E 01	2.78E 01	2.63E 01	2.49E 01	2.36E 01	2.23E 01						
BA134	7.13E 00	7.92E 00	9.42E 00	1.08E 01	1.22E 01	1.35E 01						
CS135	1.56E C3	1.56E C3	1.56E C3	1.56E C3	1.56E C3	1.56E C3						
XE136	1.43E C3	1.43E C3	1.43E C3	1.43E C3	1.43E C3	1.43E C3						
BA136	4.75E C1	4.89E 01	4.92E 01	4.93E 01	4.93E 01	4.93E 01						
CS137	1.47E 03	1.47E 03	1.46E 03	1.46E 03	1.45E 03	1.45E 03						
BA137	3.63E 01	3.91E 01	4.47E 01	5.02E 01	5.57E 01	6.12E 01						
BA138	1.43E C3	1.43E C3	1.43E C3	1.43E C3	1.43E C3	1.43E C3						
LA139	1.34E 03	1.34E 03	1.34E 03	1.34E 03	1.34E 03	1.34E 03						
CE140	1.35E C3	1.38E 03	1.38E 03	1.38E 03	1.38E 03	1.38E 03						
PR141	1.25E C3	1.29E 03	1.32E 03	1.33E 03	1.33E 03	1.33E 03						
CE142	1.18E 03	1.18E 03	1.18E 03	1.18E 03	1.18E 03	1.18E 03						
ND143	1.21E 03	1.24E 03	1.25E 03	1.25E 03	1.25E 03	1.25E 03						
CE144	4.20E C2	3.90E C2	3.37E 02	2.91E 02	2.52E 02	2.17E 02						
ND144	5.85E C2	6.15E C2	6.68E 02	7.14E 02	7.54E 02	7.98E 02						
ND145	7.80E 02	7.81E 02	7.81E 02	7.81E 02	7.81E 02	7.81E 02						
ND146	7.36E 02	7.36E 02	7.36E 02	7.36E 02	7.36E 02	7.36E 02						
PM147	4.20E 02	4.22E 02	4.06E 02	3.89E 02	3.72E 02	3.56E 02						
SM147	1.28E 02	1.37E 02	1.55E 02	1.73E 02	1.89E 02	2.05E 02						
ND148	4.51E C2	4.51E C2	4.51E 02	4.51E 02	4.51E 02	4.51E 02						
SM148	4.81E 01	4.98E 01	5.10E 01	5.15E 01	5.16E 01	5.17E 01						
SM149	3.30E 02	3.31E 02	3.31E 02	3.31E 02	3.31E 02	3.31E 02						
ND150	2.66E C2	2.66E C2	2.66E C2	2.66E 02	2.66E 02	2.66E 02						
SM150	2.79E 01	2.79E 01	2.79E 01	2.79E 01	2.79E 01	2.79E 01						
SM151	2.01E C2	2.01E C2	2.01E 02	2.01E 02	2.00E 02	2.00E 02						
SM152	1.83E C2	1.83E C2	1.83E 02	1.83E 02	1.83E 02	1.83E 02						
EU153	8.66E 01	8.71E 01	8.71E 01	8.71E 01	8.71E 01	8.71E 01						
SM154	7.08E 01	7.08E 01	7.08E 01	7.08E 01	7.08E 01	7.08E 01						
EU155	6.87E 01	6.65E 01	6.25E 01	5.87E 01	5.51E 01	5.17E 01						
GD155	2.82E C1	2.03E 01	3.44E 01	3.82E 01	4.18E 01	4.51E 01						
GD156	5.78E C1	5.93E 01	5.97E 01	5.98E 01	5.98E 01	5.98E 01						
GD157	3.18E 01	3.18E 01	3.18E 01	3.18E 01	3.18E 01	3.18E 01						
GD158	8.81E C1	8.81E 01	8.81E 01	8.81E 01	8.81E 01	8.81E 01						
TB159	4.33E 01	4.34E 01	4.34E 01	4.34E 01	4.34E 01	4.34E 01						
SUBTOT	4.06E C4	4.08E C4	4.09E 04	4.10E 04	4.10E 04	4.10E 04						
TOTALS	4.11E C4	4.11E 04	4.11E C4	4.11E 04	4.11E 04	4.11E 04						

Table 11-5 Radioactivities of Fission Products

AI FOLLOW-CN HOMOGENIZED DISCHARGE FROM REACTOR										
POWER= 51.12 MW/MT, BURNUP= 38528. MWD/MT, FLUX= 2.62E 15 N/CM**2-SEC										
NUCLIDE RADIOACTIVITY, CURIES / METRIC TON (U + PU)										
		INITIAL	30. D	90. D	150. D	210. D	270. D			
H	3	1.08E 03	1.07E 03	1.06E 03	1.05E 03	1.04E 03	1.04E 03			
KR	85	1.19E 04	1.18E 04	1.17E 04	1.15E 04	1.14E 04	1.13E 04			
RB	86	3.05E 03	1.01E 03	1.09E 02	1.18E 01	1.27E 00	1.36E -01			
SR	89	8.10E 05	5.43E 05	2.44E 05	1.10E 05	4.93E 04	2.22E 04			
SR	90	5.10E 04	5.09E 04	5.07E 04	5.05E 04	5.03E 04	5.01E 04			
Y	90	5.24E 04	5.09E 04	5.07E 04	5.05E 04	5.03E 04	5.01E 04			
Y	91	1.12E 06	7.97E 05	3.88E 05	1.91E 05	9.43E 04	4.65E 04			
ZR	93	1.68E 00	1.68E 00	1.68E 00	1.68E 00	1.68E 00	1.68E 00			
NB	93M	8.81E -02	9.47E -02	1.08E -01	1.21E -01	1.34E -01	1.47E -01			
ZR	95	2.47E 06	1.80E 06	9.47E 05	4.99E 05	2.63E 05	1.39E 05			
NB	95M	4.94E 04	3.81E 04	2.01E 04	1.06E 04	5.59E 03	2.95E 03			
NB	95	2.46E 06	2.29E 06	1.57E 06	9.35E 05	5.26E 05	2.89E 05			
TC	99	1.73E 01	1.74E 01	1.74E 01	1.74E 01	1.74E 01	1.74E 01			
RU103		2.55E 06	1.51E 06	5.27E 05	1.84E 05	6.45E 04	2.26E 04			
RH103M		2.55E 06	1.51E 06	5.29E 05	1.85E 05	6.46E 04	2.26E 04			
RU106		1.37E 06	1.30E 06	1.16E 06	1.03E 06	9.22E 05	8.24E 05			
RH106		1.37E 06	1.30E 06	1.16E 06	1.03E 06	9.22E 05	8.24E 05			
AG110M		1.93E 03	1.78E 03	1.51E 03	1.28E 03	1.09E 03	9.23E 02			
AG110		1.11E 05	2.32E 02	1.96E 02	1.67E 02	1.41E 02	1.20E 02			
AG111		1.72E 05	1.08E 04	4.23E 01	1.64E -01	6.41E -04	2.51E -06			
CD113M		1.66E 02	1.65E 02	1.64E 02	1.62E 02	1.61E 02	1.60E 02			
IN114M		3.56E 00	2.35E 00	1.02E 00	4.46E -01	1.94E -01	8.44E -02			
IN114		5.83E 00	2.27E 00	9.88E -01	4.30E -01	1.87E -01	8.15E -02			
CD115M		3.76E 02	2.32E 02	8.81E 01	3.35E 01	1.27E 01	4.84E 00			
SN119M		2.59E 01	2.39E 01	2.02E 01	1.71E 01	1.45E 01	1.23E 01			
SN121M		6.28E 01	6.27E 01	6.26E 01	6.25E 01	6.25E 01	6.24E 01			
SN123M		7.17E 02	6.07E 02	4.35E 02	3.12E 02	2.24E 02	1.60E 02			
TE123M		4.47E 00	3.74E 00	2.62E 00	1.84E 00	1.29E 00	9.03E -01			
SB124		1.30E 02	9.23E 01	4.61E 01	2.31E 01	1.15E 01	5.77E 00			
SN125		5.23E 04	5.72E 03	6.86E 01	8.22E -01	9.84E -03	1.18E -04			
SB125		2.18E 04	2.18E 04	2.10E 04	2.01E 04	1.93E 04	1.85E 04			
TE125M		7.71E 03	7.96E 03	8.15E 03	8.07E 03	7.86E 03	7.59E 03			
SN126		1.98E 00	1.98E 00	1.98E 00	1.98E 00	1.98E 00	1.98E 00			
SB126M		2.59E 03	1.98E 00	1.98E 00	1.98E 00	1.98E 00	1.98E 00			
SB126		4.32E 03	8.21E 02	3.14E 01	3.02E 00	2.00E 00	1.96E 00			
TE127M		6.22E 04	5.33E 04	3.64E 04	2.49E 04	1.70E 04	1.16E 04			
TE127		2.85E 05	5.39E 04	3.60E 04	2.46E 04	1.68E 04	1.15E 04			
TE129M		2.83E 05	1.54E 05	4.54E 04	1.34E 04	3.93E 03	1.16E 03			
TE129		6.84E 05	9.88E 04	2.91E 04	8.56E 03	2.52E 03	7.41E 02			
I131		1.53E 06	1.19E 05	6.76E 02	3.86E 00	2.20E -02	1.26E -04			
XE131M		1.57E 04	5.28E 03	2.04E 02	6.28E 00	1.87E -01	5.51E -03			
CS134		3.73E 04	3.62E 04	3.43E 04	3.24E 04	3.07E 04	2.90E 04			
CS135		1.38E 00	1.38E 00	1.38E 00	1.38E 00	1.38E 00	1.38E 00			
CS136		1.33E 05	2.69E 04	1.10E 03	4.48E 01	1.83E 00	7.45E -02			
CS137		1.28E 05	1.28E 05	1.27E 05	1.27E 05	1.26E 05	1.26E 05			
BA137M		1.20E 05	1.19E 05	1.19E 05	1.19E 05	1.18E 05	1.18E 05			
BA140		2.25E 06	4.44E 05	1.72E 04	6.69E 02	2.60E 01	1.01E 00			

Table 11-5 (continued)

A1 FOLLOW-CN HCMCGENIZED DISCHARGE FROM REACTOR									
POWER= 51.12 MW/MT, BURNUP= 38528. MWD/MT, FLUX= 2.62E 15 N/CM**2-SEC									
NUCLIDE RADIOACTIVITY, CURIFS / METRIC TCN (U + PU)									
	INITIAL	30. D	90. D	150. D	210. D	270. D			
LA140	2.34E C6	5.11E 05	1.98E C4	7.70E 02	2.99E 01	1.16E 00			
CE141	2.38E C6	1.26E 06	3.49E 05	9.67E 04	2.68E 04	7.43E 03			
PR143	2.26E C6	5.50E 05	2.64E C4	1.27E C3	6.10E 01	2.93E 00			
CE144	1.34E C6	1.25E C6	1.08E C6	9.30E 05	8.04E 05	6.94E 05			
PR144	1.39E 06	1.25E C6	1.08E 06	9.30E 05	8.04E 05	6.94E 05			
ND147	1.03E C6	1.58E 05	3.74E C3	8.82E 01	2.08E 00	4.91E-02			
PM147	3.90E C5	3.92E C5	3.77E C5	3.61E 05	3.46E 05	3.31E 05			
PM148M	6.65E C4	4.05E C4	1.51E C4	5.59E C3	2.08E C3	7.72E 02			
PM148	7.53E C4	4.74E 03	1.21E C3	4.40E 02	1.67E 02	6.20E 01			
SM151	5.47E 03	5.48E 03	5.47E 03	5.46E C3	5.46E 03	5.45E 03			
EU152	1.81E 01	1.80E C1	1.79E 01	1.77E 01	1.75E 01	1.74E C1			
GD153	1.04E C0	9.58E-01	8.07E-01	6.79E-01	5.72E-01	4.82E-01			
EU154	1.25E C3	1.24E C3	1.23E C3	1.22E C3	1.21E C3	1.21E C3			
EU155	8.75E C4	8.48E 04	7.97E 04	7.48E 04	7.02E 04	6.60E 04			
EU156	1.04E C5	2.65E C4	1.65E 03	1.03E C2	6.46E 00	4.03E-01			
TB160	1.26E C4	9.46E 03	5.31E 03	2.98E C3	1.68E 03	9.42E 02			
GD162	4.68E C3	4.42E 03	3.94E 03	3.52E 03	3.14E 03	2.80E 03			
TB162M	4.68E 03	4.42E C3	3.94E C3	3.52E C3	3.14E 03	2.80E 03			
SUBTOT	3.23E C7	1.80E C7	1.02E 07	7.09E C6	5.44E 06	4.43E 06			
TOTALS	2.22E C8	1.81E C7	1.02E 07	7.09E C6	5.44E 06	4.43E 06			

Table 11-6 Decay Heats of Fission Products

AI FOLLOW-CN HOMOGENIZED DISCHARGE FROM REACTOR									
PCWER= 51.12 MW/MT, BURNUP= 38528. MWD/MT, FLUX= 2.62E 15 N/CM**2-SEC									
NUCLIDE THERMAL POWER, WATTS / METRIC TON (U + PU)									
	INITIAL	30. D	90. D	150. D	210. D	270. D			
KR 85	1.90E 01	1.89E 01	1.87E 01	1.85E 01	1.84E 01	1.82E 01			
SR 89	2.52E 03	1.96E 03	8.79E 02	3.95E 02	1.77E 02	7.98E 01			
SR 90	6.65E 01	6.64E 01	6.61E 01	6.59E 01	6.56E 01	6.53E 01			
Y 90	3.00E 02	2.91E 02	2.90E 02	2.89E 02	2.98E 02	2.87E 02			
Y 91	4.25E 03	2.99E 03	1.48E 03	7.28E 02	3.59E 02	1.77E 02			
ZR 95	1.29E 04	9.40E 03	4.96E 03	2.61E 03	1.38E 03	7.27E 02			
NB 95M	6.88E 01	5.31E 01	2.80E 01	1.48E 01	7.79E 00	4.11E 00			
NB 95	1.18E 04	1.10E 04	7.51E 03	4.48E 03	2.52E 03	1.39E 03			
RU103	8.32E 03	4.92E 03	1.72E 03	6.02E 02	2.11E 02	7.37E 01			
RH103M	6.04E 02	3.58E 02	1.25E 02	4.38E 01	1.53E 01	5.36E 00			
RU106	8.13E 01	7.68E 01	6.86E 01	6.12E 01	5.47E 01	4.88E 01			
RH106	1.33E 04	1.25E 04	1.12E 04	9.98E 03	8.91E 03	7.94E 03			
AG110M	3.12E 01	2.87E 01	2.44E 01	2.07E 01	1.75E 01	1.49E 01			
AG110	8.05E 02	1.68E 00	1.43E 00	1.21E 00	1.03E 00	8.71E 01			
CD113M	2.19E 01	2.18E 01	2.16E 01	2.14E 01	2.13E 01	2.11E 01			
CD115M	1.53E 00	9.44E 01	3.59E 01	1.36E 01	5.18E 02	1.97E 02			
SN123M	2.44E 00	2.07E 00	1.48E 00	1.06E 00	7.62E 01	5.46E 01			
SB124	1.76E 00	1.25E 00	6.24E 01	3.12E 01	1.56E 01	7.80E 02			
SB125	7.33E 01	7.32E 01	7.03E 01	6.74E 01	6.47E 01	6.20E 01			
TE125M	6.62E 00	6.84E 00	7.00E 00	6.93E 00	6.75E 00	6.52E 00			
TE127M	3.43E 01	2.94E 01	2.01E 01	1.37E 01	9.36E 00	6.39E 00			
TE127	4.65E 02	8.79E 01	5.87E 01	4.01E 01	2.74E 01	1.87E 01			
TF129M	5.60E 02	3.05E 02	8.98E 01	2.64E 01	7.78E 00	2.29E 00			
TE129	2.48E 03	3.59E 02	1.06E 02	3.10E 01	9.14E 00	2.69E 00			
CS134	3.91E 02	3.80E 02	3.67E 02	3.40E 02	3.22E 02	3.05E 02			
CS136	2.06E 03	4.17E 02	1.70E 01	6.93E 01	2.83E 02	1.15E 03			
CS137	2.07E 02	2.07E 02	2.06E 02	2.05E 02	2.04E 02	2.04E 02			
BA137M	4.69E 02	4.69E 02	4.67E 02	4.65E 02	4.63E 02	4.62E 02			
BA140	7.60E 03	1.50E 03	5.81E 01	2.26E 00	9.76E 02	3.40E 03			
LA140	3.88E 04	8.48E 03	3.29E 02	1.28E 01	4.96E 01	1.92E 02			
CE141	4.69E 03	2.48E 03	6.87E 02	1.90E 02	5.27E 01	1.46E 01			
PR143	4.90E 03	1.19E 03	5.73E 01	2.75E 00	1.32E 01	6.36E 03			
CE144	1.18E 03	1.09E 03	9.45E 02	8.16E 02	7.05E 02	6.09E 02			
PR144	1.04E 04	9.28E 03	8.01E 03	6.92E 03	5.98E 03	5.16E 03			
ND147	2.88E 03	4.43E 02	1.05E 01	2.47E 01	5.82E 03	1.37E 04			
PM147	2.01E 02	2.02E 02	1.94E 02	1.86E 02	1.78E 02	1.71E 02			
FM148M	8.44E 02	5.14E 02	1.91E 02	7.10E 01	2.64E 01	9.80E 00			
PM149	6.16E 02	3.98E 00	9.90E 00	3.68E 00	1.37E 00	5.07E 01			
SM151	9.53E 00	9.55E 00	9.54E 00	9.52E 00	9.51E 00	9.50E 00			
EU152	2.21E 01	2.20E 01	2.18E 01	2.16E 01	2.14E 01	2.12E 01			
EU154	1.17E 01	1.16E 01	1.15E 01	1.15E 01	1.14E 01	1.13E 01			
EU155	7.37E 01	7.14E 01	6.71E 01	6.30E 01	5.91E 01	5.55E 01			
EU156	1.09E 03	2.79E 02	1.74E 01	1.09E 00	6.91E 02	4.26E 03			
TB160	1.06E 02	7.96E 01	4.47E 01	2.51E 01	1.41E 01	7.93E 00			
GC162	1.59E 01	1.50E 01	1.34E 01	1.20E 01	1.07E 01	9.53E 00			
TB162M	3.13E 01	2.96E 01	2.64E 01	2.36E 01	2.10E 01	1.88E 01			
SUBTOT	1.36E 05	7.17E 04	4.04E 04	2.89E 04	2.22E 04	1.80E 04			
TOTALS	2.27E 06	7.25E 04	4.04E 04	2.89E 04	2.22E 04	1.80E 04			

We have also investigated the possibility of using tetra-boron carbide (B_4C) as the outermost shield zone instead of borated water. A solid layer of B_4C may be preferable to an annulus of water. The results for the AI reference design fuel indicate that ~ 16 cm of B_4C will give the same neutron + gamma attenuation as ~ 8 cm of borated water when these materials follow 55 cm of iron.

12. CRITICALITY (TASK 12) (J. P. Nichols)

The objective of this task is to develop evaluated criticality data for application to the aqueous processing of LMFBR fuels. The work involves compilation of basic data and liaison with other critical experiment groups (Task 12.1); calculation of basic parameters (Task 12.2); the performance of select critical experiments with systems containing fixed and soluble neutron absorbers (Task 12.3); and the application of the critical data to the design of process equipment (Task 12.4).

12.2 Criticality Calculations for Fuel Shipping Casks (D. W. Magnuson)

A series of calculations were made to investigate the criticality parameters of conceptual LMFBR fuel shipping casks and to compare results obtained by computer programs of varying complexity. It is concluded that relatively simple and inexpensive discrete ordinates calculations (ANISN) with the 16 group Hansen-Roach cross section set provide adequate results for survey studies of sodium-filled arrays of LMFBR elements that are reflected by steel. It is concluded that an array of 36 core elements in a cask will be appreciably subcritical but that the source neutron multiplication factor, $1/(1-k_{eff})$, may be in the range of 3 to 15 depending on the plutonium concentration of the elements. In general, the neutron multiplication factors are expected to be highest with elements from a small demonstration reactor or from an LMFBR with a modular core and lower with elements from a large commercial LMFBR that probably will have regular geometry and low Pu concentration. Techniques that are available for further reduction of the source neutron multiplication include judicious arrangement of core elements of various reactivity within the cask, interspersing of radial blanket elements, and use of lithium instead of sodium as the coolant.

A proposed shipping cask for spent LMFBR fuels will hold 36 elements (a 37-unit hexagonal array with the center one missing), will be shielded by 20 in. of iron, and will have 2-in.-sodium-filled spaces between elements to permit cooling by convection. Criticality calculations were made for a mockup of such a cask containing elements from three conceptual LMFBR's. These calculations are described and summarized in Table 12-1.

Table 12-1. Criticality Calculations for 37-Element Shipping Casks for Various LMFBR Fuel Elements

	Radius ^a (cm)		Axial Dimension (cm)		Calculated k Value			
	Region		Region		123 Group XSDRN	16-Group Hansen-Roach		
	1	2	1	2		ANISN	DOT	KENO
AI - REFERENCE DESIGN, OUTER STARTER ELEMENT ^b								
Fuel Element, White Boundary	7.268	--	∞	--		1.6856		
Fuel Element + 1 in. Sodium, White Boundary	7.268	9.935	∞	∞		1.5855		
37(Fuel Element + 1 in. Sodium), 50 cm Fe Reflector	60.432	110.432	∞	∞		1.1177		
∞(Fuel Element + 1 in. Sodium), Axial Blanket	∞	∞	60.96	91.44		1.3104		
37-Element Shipping Cask	60.432	110.432	60.96	91.44		(0.924) ^c		
AI - FOLLOW-ON DESIGN, INNER ELEMENT MID-CYCLE ^d								
Fuel Element + 1 in. Sodium, White Boundary, Homogeneous	10.225	--	∞	--		1.1031		
37(Fuel Element + 1 in. Sodium), 50.8 cm Fe Reflector	62.196	112.996	∞	∞		0.8308		
∞(Fuel Element + 1 in. Sodium), Axial Blanket	∞	∞	54.407	88.697		0.9237		
37-Element Shipping Cask	62.196	112.996	54.407	88.697		(0.696) ^c		
AI - FOLLOW-ON DESIGN, OUTER ELEMENT MID-CYCLE ^d								
Fuel Element + 1 in. Sodium, White Boundary, Homogeneous	10.225	--	∞	--		1.2528		
37(Fuel Element + 1 in. Sodium), 50.8 cm Fe Reflector	62.196	112.996	∞	∞		0.9557		0.957 ± 0.002 ^e
∞(Fuel Element + 1 in. Sodium), Axial Blanket	∞	∞	54.407	88.697		1.0559		
37-Element Shipping Cask	62.196	112.996	54.407	88.697		(0.805) ^c		
GE - FOLLOW-ON DESIGN, INNER ELEMENT DISCHARGE ^f								
Fuel Element, White Boundary	7.536	--	∞	--	1.2217	1.2394		
Fuel Element + 1 in. Sodium, White Boundary	7.536	10.163	∞	∞		1.1566		
Fuel Element + 1 in. Sodium, White Boundary, Homogeneous	10.163	--	∞	--		1.1535		
37(Fuel Element + 1 in. Sodium), 50.8 cm Fe Reflector	61.819	112.619	∞	∞		0.8782		
∞(Fuel Element + 1 in. Sodium), Axial Blanket	∞	∞	38.10	68.58		0.8989		
37-Element Shipping Cask	61.819	112.619	38.10	68.58		(0.683) ^c		
GE - FOLLOW-ON DESIGN, OUTER ELEMENT DISCHARGE ^f								
Fuel Element, White Boundary	7.536	--	∞	--		1.3544		
Fuel Element + 1 in. Sodium, White Boundary	7.536	10.163	∞	∞		1.2629		
37(Fuel Element + 1 in. Sodium), 50.8 cm Fe Reflector	61.819	112.619	∞	∞		0.9661		
∞(Fuel Element + 1 in. Sodium), Axial Blanket	∞	∞	38.10	68.58		0.9753		
37-Element Shipping Cask	61.819	112.619	38.10	68.58		(0.746) ^c	0.7630	0.764 ± 0.004

a. The hexagonal fuel element and sodium regions were approximated by circles of equal area.

b. Atom densities from Table A-18, p. A-84, NAA-SR-MEMO-12604.

c. Estimated from other calculations.

d. Atom densities calculated from the fuel element description and loadings at mid-cycle, AI-AEC-12791, Vol. 1.

e. Hexagonal geometry used in the KENO calculation.

f. Atom densities calculated from the fuel element description and loading at discharge, GEAP-5678.

Most of the calculations were made using the ANISN transport code in the S_4 approximation with Hansen-Roach 16-group cross sections. A 123-group XSDRN transport calculation, which used cross sections from the XSDRN library and which treated the ^{238}U resonances by the Nordheim Integral Method, resulted in a decrease of 1.45% in k_∞ . DOT (two-dimensional transport) and KENO (Monte Carlo) calculations, both using 16-group Hansen-Roach cross sections, of k_{eff} for the shipping cask represented by a fueled region, an axial blanket, and an iron reflector in cylindrical geometry agree with the value obtained from two one-dimensional ANISN calculations. In all calculations except for one KENO calculation the hexagonal geometry of the fuel elements was replaced by cylindrical geometry on an equal area basis; the KENO calculation indicates that the effect of the cylindrical approximation is to cause an underestimation of k_∞ by about 0.1%. Smeared atom densities, based on volume fractions, were used for the fuel element and also for the fuel element and sodium annulus. A two-region calculation for a single fuel element surrounded by an annulus of sodium, when compared to a one-region calculation using smeared densities, showed an increase in k_∞ of 0.0031.

For the AI Follow-On LMFBR, the substitution of six radial blanket elements for outer fuel elements in a 37-unit array reflected by iron reduces k_∞ (KENO calculations in hexagonal two-dimensional geometry) from 0.957 ± 0.002 to 0.848 ± 0.002 . The substitution of natural lithium for sodium (a suggestion by J. D. Rollins) changed k_∞ for the fuel element with 1 in. of alkali metal around it from 1.2528 to 0.7929 (ANISN calculations).

13. ENGINEERING STUDIES (TASK 13) (A. R. Irvine, C. D. Watson)

The purpose of this task is to examine the overall problem of LMFBR spent fuel recovery plant design and to relate the individual problems involved in fuel recovery to the overall problem and to the other individual problems. The plant design surveys which are to be made include adaptation of existing LWR reprocessing facilities for recovery of fuel from FFTF and initial demonstration reactors, possible adaptation of later LWR reprocessing facilities to accept LMFBR spent fuel from commercial reactors during the first decade (or more) of their existence, and, finally, preparation of a conceptual design for a facility that will be needed when a large number of breeder reactors are in use. Individual problems to be evaluated range from special problems in safety and containment (which are peculiar to reprocessing of LMFBR spent fuel) to equipment design and operational procedures to prevent formation of plutonium polymer.

13.1 Existing Commercial Facilities

A study of existing commercial facilities is under way to determine their suitability for reprocessing spent fuel originating in the Fast Test Reactor (FTR). The study will also determine what additional modifications would be required to accommodate fast breeder reactor type fuels from demonstration and early commercial LMFBR's. The general approach is to investigate the lowest capital investment method (consistent with reasonable and safe operating practice) and then to determine the capital investment requirement for obtaining additional development information or lower overall operating cost. The Nuclear Fuel Services, Inc., reprocessing facility is being used as the basis for this study as it is currently the only commercial facility in operation.

Receiving and Storage Facilities (A. R. Irvine)

A basic assumption of this study is that fuel will be shipped to the existing facility in a cask that employs liquid sodium to transport heat within the fuel cavity. Facilities for receipt of the normal feed material to existing commercial reprocessing facilities are not appropriate for receipt of a sodium-containing cask.

We have prepared a concept of a receiving facility for sodium-filled casks that we believe could be a prototype for subsequent facilities serving this function. It includes means for containment of radioactive particulate, exclusion of oxygen and other reactive materials from the sodium in the cask, transport of fuel subassemblies from the cask to a sodium removal station and thence to a combination canning and exit port, purification of the inert gas atmosphere, and retention of any radio-iodine released in the course of processing

the subassemblies. The cell for performing the mechanical manipulations is relatively small, having internal dimensions of only 10 ft x 20 ft x 35 ft high. The facility concept includes underwater transfer of the sodium-cleaned fuel subassemblies from the receiving facility to the existing storage canal where they would be held until processing could be accomplished. A written description and cost estimate are being prepared.

Reactor Reference Data (R. S. Lowrie, S. D. Clinton, G. A. West)

Reactor characteristics and specifications for the FTR, AI Reference Oxide, AI Follow-On and GE Follow-On fuels were compiled (see Table 13-1) for use in an initial Engineering Evaluation Study of the reprocessing of FTR fuel in a Nuclear Fuel Services-type plant. The reprocessing of the other LMFBR fuels will be included in a secondary phase of the study.

Reference Flowsheets (S. D. Clinton, D. E. Horner)

Reference flowsheets are being prepared for the Engineering Evaluation Studies of the reprocessing of 180-day decayed fuel from the FTR. Similar flowsheets are also being developed for the Atomics International Reference Oxide, Atomics International Follow-On and General Electric Follow-On reactor fuels. This month a flowsheet for the FTR fuel was prepared. The flowsheets for reprocessing Reference Oxide AI and GE Follow-On fuels will be completed next month (November).

The FTR reference flowsheet (see Fig. 13-1) presents an estimate of the amount and concentration of uranium, plutonium, fission products, and stainless steel at the various equipment locations for the processing of 180-day decayed fuel assemblies at the rate of 1000 kg of U and Pu per day (as fed to the reactor). After a burnup of 45,000 megawatt days/metric ton, 29.5 FTR fuel assemblies will be supplied daily to the reprocessing plant consisting of the following: 752 kg of U, 200 kg of Pu, 48 kg of fission products, and about 2800 kg of stainless steel. The aqueous feed from the dissolver is poisoned with 0.1 M $\text{Cd}(\text{NO}_3)_2$ for criticality control. The product concentration from the first TBP solvent extraction cycle will be 46 g of U per liter, and the product concentration from the third TBP solvent extraction cycle will be 67 g Pu per liter. Presently, it has not been decided whether the unfueled stainless steel gas plenum sections from the shearing operation should be diverted away from the dissolver or pass through it. Only 1 wt % of the stainless steel was assumed to dissolve in boiling 10-13 M HNO_3 . The first, second, and third TBP solvent extraction cycles have been expanded (see Figs. 13-2, 13-3, and 13-4) to indicate flow ratios and concentrations in the various pulse columns.

BASIS 1,000 Kg (Pu + U) AS CHARGED TO THE REACTOR PER DAY

Fig. 13-1 Reprocessing Plant Material Balance for FTR Fuel Assemblies.

Table 13-1 Reactor Reference Data for Engineering
Evaluation Studies of Reprocessing
FTR (IMFBR) Fuels

Specifications	R E A C T O R			
	FTR (A-0035-R2)	AI Ref. Oxide (NAA-SR-Memo 12604)	AI Follow-On (AI-AEC-12792)	GE Follow-On (GEAP-5678)
Mw(t)	395	2500	2400	2417
	<u>Core-Axial Blanket Fuel Assembly</u>			
Fuel Configuration	Hexagonal	Hexagonal	Hexagonal	Hexagonal
Number/Reactor - inner	28	252	133	108/19
Number/Reactor - outer	48		141	126/12
Shroud Material	304 SS	304 SS	316 SS	316 SS
thickness (in.)	0.140	0.130	0.140	0.060
distance across flats (in.)	4.615	5.4	5.617	5.621
Length				
core (in.)	36.0	48.0	42.84	30.0
upper axial blanket (in.)	0.5	12.0	12.00	15.0
lower axial blanket (in.)	0.5	12.0	15.00	15.0
plenum (in.)	37.25	60.0	37.6	21.0
fuel rod (in.)	88.75	144.0	110.5	82.25
overall (in.)	144.0	212.0	180.0	111.0
Fuel Rod Data				
number/assembly	217	217	217	240 pin-wrapped 91 pin-unwrapped
outside diam (in.)	0.230	0.250	0.300	0.248
pitch (in.)	0.286	0.341	0.359	0.300
clad thickness (in.)	0.015	0.015	0.0175	0.010
clad material	316 SS	304 SS	316 SS	316 SS
fuel density (% theo.)				
inner	93	80	88.3 core ~95% blanket	89.6
outer	93	93	88.3 core ~95% blanket	89.6
bond material	He	He	He	He
vented	No	No	No	Yes

Table 13-1 (Cont'd.)

Specifications	R E A C T O R			
	FTR (A-0035-R2)	AI Ref. Oxide (NAA-SR-Memo 12604)	AI Follow-On (AI-AEC-12792)	GE Follow-On (GEAP-5678)
fuel rod spacer	wire	wire	wire	wire
spacer material	316 SS	304 SS	316 SS	316 SS
wire wrap diam (in.)				
inner coolant channels	0.056	0.091	0.055	0.050
outer coolant channels	0.056	0.091	0.0247	0.050
pitch-diam ratio	1.243	1.364	1.197	
axial pitch of wire-wrap helix (in.)	12	12	12	6
Core Composition	oxide	oxide	oxide	oxide
inner	19.1% Pu-81.9% U		13.32% Pu-86.68% U	15.01% Pu-84.99% U
outer	25.4% Pu-74.6% U		18.38% Pu-81.62% U	20.97% Pu-79.03% U
Fuel loading per Assembly (kg)				
{ 239-Pu	5.81		6.22	
{ 240-Pu	0.74		2.42	
{ 241-Pu	—		0.47	
{ 242-Pu	—		0.18	
{ 235-U	0.20		0.12	
{ 238-U	27.69		60.3	
{ 239-Pu	7.72		8.60	
{ 240-Pu	1.06		3.30	
{ 241-Pu	—		0.64	
{ 242-Pu	—		0.26	
{ 235-U	0.18		0.11	
{ 238-U	25.62		56.82	
Nuclear Data--Core				
avg. sp. pow. Mw/MT (+ Pu)	153	175	114	157
burnup, avg., Mwd/MT	45,000	80,000	67,000	100,167
reactor charge (U + Pu) MT	2.6	12.6	21.65	
(Pu fissile) MT	0.5	1.96	2.56	
(Pu/U + Pu)	0.215	0.22	0.19	
rod, avg. lin. power, kw/ft	7.0	10.0	10.15	9.63
rod, max. lin. power, kw/ft	12.2	16.0	16.2	
max. clad temp., °F	1200	1230	1184	1150
max. fuel temp., °F	4200	4748		5200

Table 13-1 (Cont'd.)

Specifications		R E A C T O R			
		FTR (A-0035-R2)	AI Ref. Oxide (NAA-SR-Memo 12604)	AI Follow-On (AI-AEC-12792)	GE Follow-On (GEAP-5678)
Nuclear Data--Axial Blanket					
avg. sp. pow. Mw/MT (U)			5.5	5.7	12.18
burnup, avg., Mwd/MT (U)			2500	3340	
charge, MT (U)			7.32	14.66	
<u>Radial Blanket Assembly</u>					
Fuel Configuration		Hexagonal	Hexagonal	Hexagonal	Hexagonal
Number/Reactor - inner		None	147	66	126
outer			87	72	126
Shroud Material			304 SS	316 SS	316 SS
thickness			0.130	0.100	0.120
distance across flat (in.)			5.4	5.617	5.621
Length					
blanket material, (in.)	inner row		60	56.34	60
	outer row		48	42.84	60
plenum, (in.)	inner row			10.0	19.5
	outer row			10.0	19.5
fuel rod, (in.)	inner row		84.0	71.0	82.25
	outer row		72.0	57.875	82.25
overall assembly length, (in.)			212.0	180.0	111.0
Fuel Rod Data					
rods/assembly	inner row		169	91	127
	outer row		91	61	127
rod diam, (in.)	inner row		0.35	0.50	0.418
	outer row		0.51	0.617	0.418
pitch, (in.)	inner row		0.36	0.5547	0.468
	outer row		0.49	0.6758	0.468
pitch/diam ratio	inner row		1.11	1.109	1.119
	outer row		1.047	1.095	1.119
cladding thickness, (in.)			0.015	0.175	0.015
cladding material			304 SS	316 SS	316 SS
fuel composition			UO ₂	UO ₂	UO ₂

Table 13-1 (Cont'd.)

Specifications	R E A C T O R			
	FTR (A-0035-R2)	AI Ref. Oxide (NAA-SR-Memo 12604)	AI Follow-On (AI-AEC-12792)	GE Follow-On (GEAP-5678)
fuel density, (% theo.)	None		94	95
bond material		He	He	He
vented		No	No	Yes
fuel rod spacer		Wire	Wire	Wire
wire diam (in.)		0.040	0.053	0.050
	inner row			
	outer row	0.040, 0.023	0.057	0.050
axial pitch of wire wrap				
helix, (in.)				
	inner row	8	12.0	2.0
	outer row	8	12.0	2.0
Nuclear Data				
avg. spec. pow., Mw/MT, inner		10.0	10.2	7.0
	outer		5.06	
burnup, avg., Mwd/MT, inner		8,100	15,000	
	outer		10,340	
charge, MT (U)		26.7	17.78	
	inner		17.78	
	outer

Fig. 13-2 First Solvent Extraction Flow Sheet for FTR Fuel.

Fig. 13-3 Second Solvent Extraction Flow Sheet for FTR Fuel.

Fig. 13-4 Third Cycle Solvent Extraction Flow Sheet for FTR Fuel.

Head-End Processing (C. D. Watson, R. S. Lowrie, R. L. Cox, W. S. Groenier, G. A. West)

A Head-End Engineering Evaluation Study has begun of the reprocessing of 180-day decayed FTR fuel in a Nuclear Fuel Services-type plant. Later, AI Reference Oxide, AI Follow-On and GE Follow-On fuels will be studied and compared to FTR reprocessing. An analysis of the dismantling, shearing, and dissolution processing steps is presently being made. This will include the mechanical processing, sodium deactivation, heat dissipation, and criticality aspects. Initial emphasis has been placed on a study of the mechanical processing equipment at NFS to reveal problem areas, if any, brought about by the physical dimensions of FTR fuel and the possible presence of sodium. Some very preliminary head-end observations follow:

1. Wet abrasive sawing (cropping) of end hardware in the presence of unexpected quantities of sodium may necessitate the addition of nitrogen blanketing equipment to control the hydrogen content of the off-gas.
2. Shroud (flow duct) removal equipment, such as plasma-arc cutting, may be required if shearing of shrouded intact assemblies fails to produce sheared pieces sufficiently small to pass through the discharge chute from the shear. Presently, NFS does not have equipment for shroud removal.
3. The length of stroke (~ 9 in.) of the present NFS shear is considered to be too short to consistently produce discrete sheared pieces of product from an intact shrouded FTR sub-assembly that would pass easily through the discharge chute of the shear.
4. The dissolver will probably require the addition of a steam-nitrogen purge system to accommodate unexpected quantities of sodium.

The study will be continued through the next report period (November).

II. FUEL MATERIAL PREPARATION

R. G. Wymer, R. E. Brooksbank, R. E. Leuze,
J. P. McBride, W. D. Bond, P. A. Haas

1. CONVERSION STUDIES (R. E. Brooksbank)

In the LMFBR fuel cycle, it would be highly advantageous to convert the product solutions from the fuel reprocessing plants to solid fuel materials that are suitable both for shipping and for direct use in re-fabrication of fuel elements. Earlier fuel cycle studies have not necessarily optimized the conversion to meet both of these requirements. In order to meet both requirements, the solids must be in the desired chemical form with the necessary purity. In this study, we hope to determine the best method for accomplishing the desired conversion. Although the major emphasis will be placed on conversions to oxides, the conversion to advanced fuel forms (such as carbides, nitrides, and carbonitrides) will be included in our studies.

We are studying processes that are currently in use or under development; an economic evaluation is being made prior to undertaking extensive laboratory and engineering process studies. The conversion processes being evaluated include a variety of approaches, such as fluidized-bed conversion, flame denitration, sol-gel, and numerous precipitation processes.

1.1 Sinterable Powder Preparation (W. D. Bond, P. A. Haas, R. E. Brooksbank)

Laboratory and engineering studies of the conversion of aqueous solutions of uranium and plutonium to powders will be initiated after our process evaluation studies (Sect. 1.2) have indicated which of the possible methods are the more promising.

1.2 Process Evaluation

Currently plutonium is shipped from chemical processing plants as a nitrate solution. While this is satisfactory at present, safety considerations will probably dictate that the increased quantities of plutonium that will result from LMFBR fuel processing be shipped in a solid form. The objectives of this program are to define the most reasonable form for shipment to fabrication plants and to develop or adapt processes for this purpose. Processes currently used or being developed will be studied, and an economic evaluation will be made to establish the most promising approach. This work will, in turn, provide guidance for further experimental work. As a need for further developmental work is recognized, it will be reported.

The existing methods under study include those that are most likely to be used in production of first generation fast reactor fuel elements.

They are: (1) dry powder blending of oxide powders for pelletizing, (2) coprecipitation of plutonium and uranium to provide a mixed oxide for pelletizing, and (3) use of the sol-gel method to form either separate or blended UO_2 and PuO_2 microspheres or pelletizing stock. These are the most directly useful conversions, but if it appears desirable to convert to an intermediate solely for shipping purposes, this can also be considered.

1.2.1 Chemical Conversion (F. E. Harrington, J. D. Sease, R. B. Pratt)

We continued the study of the costs involved in four alternative routes to the fuel preparation and fuel element fabrication for a specific LMFBR, as described earlier.¹ The first draft of the report of this cost study has been completed.

Additional detailed cost studies will not be started until the conversion process has been more clearly defined.

1.2.2 Shipping and Storage (R. W. Horton)

The uranium compounds commonly stored and shipped, and hence most available as feed material for LMFBR fuel fabricators, are UF_6 and UO_3 . Of these, the one currently available in larger supply is UF_6 that has been depleted in ^{235}U by passage through the gaseous diffusion cascade. The trioxide is the common intermediate product in the uranium ore refining process (UO_3 is converted to UF_6 in preparing feed material for gaseous diffusion).²

In the case of LMFBR fuel reprocessing plants, large amounts of ^{238}U will be separated from plutonium in the solvent extraction system and will be available as uranyl nitrate. This solution can be denitrated to UO_3 at the reprocessing plant, and the dry UO_3 can then be shipped and stored more economically than the uranyl nitrate solution. The trioxide can also be converted to a dioxide powder with the desired physical properties by a controlled oxidation-reduction step, and therefore would serve as a useful source material to the fuel fabricator.

Plutonium is a product of spent reactor fuel processing plants, and is usually available as a nitrate solution. The isotopic composition is a function of neutron exposure; the major fissile isotope, ^{239}Pu , may constitute 60 to 95% of the total mass. (Low ^{239}Pu is associated with an increased ^{241}Pu content; the minimum fissile content is about 70%.) Considering the need for blending plutonium from several sources, the nitrate solution would appear to be an attractive form for plutonium. However, the cost of shipping solution is very high.¹

¹LMFBR Fuel Cycle Studies Progress Report No. 5, ORNL-TM-2671 (August 1969).

²C. D. Harrington and A. E. Ruehle, Uranium Production Technology, Chap. 2, "Chemistry of Uranium Processing," D. Van Nostrand Co., Inc., Princeton, 1959.

Further studies of the factors involved in optimizing the forms of uranium and plutonium to be shipped from the reprocessing plant to the fuel fabricators are in progress.

1.3 Advanced Fuel Studies (W. D. Bond)

Increasing demands on reactor fuels make the development of new or improved fuels important. Among the more promising advanced fuel materials for the LMFBR are the monocarbides, carbonitrides, and nitrides of plutonium and uranium. Sol-gel processes for the preparation of these materials are being developed on a laboratory scale.

Most of our work has been concerned with the preparation of carbides. We had previously demonstrated the preparation of dense ThC_2 and $(\text{Th,U})\text{C}_2$ microspheres by sol-gel processes. In more recent work, laboratory-scale preparation of dense uranium carbide microspheres was demonstrated, and plutonium carbide microspheres were prepared. Work is now in progress on the preparation of uranium-plutonium carbide microspheres and on uranium-plutonium carbonitride.

2- SOL-GEL PROCESSES FOR PREPARATION OF LMFBR RECYCLE FUEL (R. E. Leuze)

Sol-gel processes are ideally suited to the preparation of LMFBR recycle fuel since they can be readily adapted to continuous and remote operation for processing materials behind shielding. The processes under development prepare sols by controlled solvent extraction of nitrate from plutonium and uranium nitrate solutions, the products from fuel reprocessing plants. The sols are then converted to the desired ceramic forms, usually high density oxide microspheres, required for subsequent fabrication into recycle fuel elements. This section reports progress for all of the process development and demonstration, equipment design, and instrument development being carried out to obtain sufficient information for design of a full scale facility. Also included is the preparation of sol-gel materials required for testing and for use in other parts of the LMFBR fuel cycle program.

2.1 Urania Sol Process and Equipment Development (J. P. McBride)

Although the LMFBR recycle fuel under consideration is mixed urania-plutonia, study of urania sol preparation is important because urania is used as a fertile material in an LMFBR and is the major component in the mixed oxide. Urania sols are also useful for studying other phases of the program such as sphere forming and tray drying.

The CUSP process (Concentration Uranium Sol Preparation) developed at ORNL has been selected as the method for preparation of urania sols since it is most amenable to scale-up and consistently produces good sols with desirable characteristics.^{1,2} This section of the report covers all aspects of CUSP sol preparation including uranous nitrate feed preparation, laboratory- and engineering-scale process development, sol preparation, and equipment design and instrumentation.

Urania Sol Production (B. C. Finney)

The engineering-scale development and evaluation of the CUSP process for the preparation of UO₂ sol was continued. A new amine extractant regeneration method was evaluated, and the effect of using U(IV) nitrate-formate feeds that had been aged for times ranging from one to four days was studied.

In preparing UO₂ sol by the CUSP process using a water-carbonate solvent regeneration system, there is a small loss of amine from the extractant (2 to 3% per run; 15 liters aqueous phase and 30 liters solvent

¹J. P. McBride, K. H. McCorkle, W. L. Pattison, and B. C. Finney, "Preparation of Concentrated, Crystalline Urania Sols by Solvent Extraction," presented at 15th Annual Meeting of the American Nuclear Society, Seattle, Washington, June 15-19, 1969; summary published in Trans. Am. Nucl. Soc. 12, 206 (1969).

²J. P. McBride, K. H. McCorkle, and W. L. Pattison, LMFBR Fuel Cycle Studies Progress Report No. 5, August (1969), p. 52, ORNL-TM-2671.

phase), and the extractant becomes very dark due to entrained UO_2 . There is no evidence that the use of the discolored extractant has a deleterious effect on the sol products, but interfacial levels are obscured, and there is an accumulation of black, interfacial crud at the organic-aqueous interface in the regeneration contactors. A series of five runs was made to evaluate the use of a NaNO_2 - HNO_3 scrub as a method of cleaning the extractant. It was found that a scrub consisting of 0.2 M NaNO_2 -0.5 M HNO_3 at an extractant-scrub flow ratio of 10 did clean the solvent by oxidizing the entrained U(IV) to U(VI) and extracting it into the aqueous phase. The NaNO_2 and HNO_3 were mixed just prior to use. Loss of amine, however, increased to 14-24% per run. There was no noticeable change in the amine loss when the NaNO_2 - HNO_3 scrub contactor was operated at 35-40°C as compared to 55-60°C. It was concluded that the NaNO_2 - HNO_3 clean-up of the solvent was undesirable because of operating inconvenience occasioned by the rapid loss of amine.

It has been the general practice to use the 1 M U(IV) nitrate-formate feed solution ($\text{NO}_3^-/\text{U} = 2$; $\text{HCOOH}/\text{U} = 0.5$) to prepare a UO_2 sol the day after the feed was prepared. Four CUSP UO_2 sol preparation runs were made using feed solutions that had been aged for 21, 46, 70, and 94 hr. Chemical tests on the feed indicated no self-oxidation of the feeds as a result of the aging under an argon atmosphere. In all cases, a high quality, crystalline UO_2 sol with an $87 \pm 1\%$ U(IV) content was prepared.

Equipment Development (F. G. Kitts, F. L. Daley, J. R. Parrott)

Development of equipment for the CUSP flowsheet for forming urania sols by solvent extraction with an amine is in progress in the Pilot Plant. This equipment will be used in connection with UO_2 - PuO_2 sol preparation. A uranium reductor (U^{6+} to U^{4+} using PtO_2 catalyst and elemental H_2) and equipment for urania sol and plutonia sol forming and mixing will be added to the existing Solex facility³ and used to prepare mixed sols containing 80% UO_2 -20% PuO_2 .

Our development work has been restricted mainly to testing mixing devices which might be used as the feed-extractant contactor in the CUSP process. CUSP sols have been produced using a vertical 1 in. ID glass column packed with 1/4 in. Berl saddles and co-current flow either upward or downward; flow rates of both phases may be varied independently. The contactors we have been using consist of a series of stirred mixing regions in a vertical pipe. The packed columns have the advantage of having no mechanical moving parts.

Three more runs were attempted in modified equipment but all were unsatisfactory. Although a portion of the product sol may be useable, the accumulation of emulsions in both the phase separator and the solvent regenerator makes the operation unsatisfactory and the yield unacceptable.

³J. W. Snider, The Design of Engineering-Scale Solex Equipment, ORNL-4256 (April 1969).

To check the possibility that solvent clean-up was inadequate in the batch system used in Bldg. 3019, the contactor and phase separator were moved to Bldg. 3503 and coupled with the continuous solvent regeneration system of the engineering-scale sol preparation equipment located there. A run was made using co-current upflow in the 1 in. ID contactor and again emulsion accumulated at the interface in the phase separator and carried over into the regeneration system. A very fluid sol with proper conductivity resulted, but upon standing a few hours a stable emulsion separated and rested on top of the sol.

Further attempts will be made using this set-up. We will try running organic continuous, which necessitates downflow and may present problems with venting during the gassing phase. Equipment modifications will be made as indicated by operating experience.

2.2 - Urania-Plutonia Sol Process and Equipment Development (R. E. Leuze)

This section of the report covers all aspects of mixed urania-plutonia sol preparation including preparation of high-nitrate plutonia sol, special treatment of CUSP sol prior to mixing with plutonia, techniques for mixing plutonia and urania sols, and treatments required to produce satisfactory mixed sol ready for subsequent processing into the desired ceramic form. Progress on equipment development and engineering-scale demonstration is also reported here.

Efforts at the present time are directed at setting process conditions and preparing equipment in order to demonstrate engineering-scale preparation of mixed UO_2 - PuO_2 and to produce several kilograms of irradiation test material during the first half of FY-1970. In the process selected for this demonstration, urania sol is pretreated to remove excess formate and is then mixed with a high-nitrate plutonia sol prepared by an alcohol extraction process (APEX). The mixed sol is contacted with a secondary amine to extract excess nitrate and is subsequently formed into microspheres or shards which can be dried and calcined to dense oxide product.

Mixed Plutonia-Urania Sol Compatibility Studies (O. K. Tallent)

The occurrence of thixotropic gelling is the principal indication of incompatibility in mixed CUSP-APEX sols. In past work it was found that mixed sol stability was improved by partial extraction of formate (formic acid) from urania sol; several experiments indicated that a nitrate deficiency in mixed sols also promoted difficulties at the microsphere forming step. These effects have now been further evaluated by determining the effects of nitric acid and formic acid additions on sol stability. As expected, the addition of formic acid decreased the time to gelation; nitric acid additions improved stability and increased the time to gelation.

The mixed sol studied contained 0.72 M UO_2 -0.18 M PuO_2 . The NO_3^-/Pu mole ratio of the APEX plutonia sol was 0.69. The (CUSP) urania sol was

extracted with n-hexanol prior to mixing and had NO_3^-/U and HCOO^-/U mole ratios of 0.11 and 0.4, respectively. The HNO_3 and HCOOH additions were made to portions of the sol, and the times to gelation were determined at 24°C .

The time to gelation increased almost linearly with increasing HNO_3 concentration: time to gelation increased from the initial value of 35 minutes with no HNO_3 added to 4-1/3 hrs with an added HNO_3 concentration of 0.33 M. The time to gelation decreased linearly as the logarithm of the added HCOOH concentration: gelation occurred after 5 min and 20 min, respectively, at added formic acid concentrations of 0.33 and 0.1 M.

These experiments indicate that anion balance in mixed sol systems is an important variable; further studies in this area will increase our understanding and ability to control the factors responsible for gelation in CUSP-APEX sols.

Mixed Plutonia-Urania Sol Flowsheet Development (O. K. Tallent and M. H. Lloyd)

Recent laboratory studies have indicated several methods for improving mixed sol stability and microsphere forming behavior (see above). Based on this work, several process changes appear desirable. Although it can be anticipated that additional changes will be required, a flowsheet is presented below which has produced satisfactory results on a laboratory scale.

Plutonia Sol Preparation (APEX) - This sol is prepared in the continuous solvent extraction equipment which was described last month.

1. The plutonium nitrate feed solution [$0.1\text{-}0.15\text{ M Pu}(\text{NO}_3)_4$; $1.0\text{-}1.5\text{ M HNO}_3$] is extracted with n-hexanol until the NO_3^-/Pu mole ratio is reduced to about 2.5.
2. The sol is digested at 100°C for about 1 hr.
3. The sol is cooled, and extraction with n-hexanol is continued until the NO_3^-/Pu mole ratio is reduced to ~ 1.0 (0.8-1.2).
4. The sol is concentrated by evaporation at 100°C to a plutonium concentration of $\sim 0.5\text{ M}$.
5. The sol can then be extracted to a NO_3^-/Pu mole ratio of ~ 0.6 (this can be accomplished by batch extraction, $\text{O/A} = 10$). The sol can then be concentrated to 1.0 M if desired.

Pretreatment of Urania Sol

1. A 1 M CUSP sol ($\sim 0.45\text{ M HCOOH}$; 0.1 M HNO_3) is extracted with water-saturated n-hexanol ($\text{O/A} = 5$) to remove formic acid. (All operations involving urania sol should be performed in an argon atmosphere to prevent oxidation of UO_2 .)

2. If the extracted sol is not used immediately, it should be stored with the alcohol over it, and the two phases should be equilibrated just prior to use.

Sol Blending

1. A sol batch size is selected which can be formed into microspheres the same day the sols are mixed.
2. The CUSP and APEX sols to be blended are cooled to ice bath temperature.
3. A measured volume of CUSP sol is put into an argon-blanketed container, and sufficient APEX sol is added to provide a mixed sol which contains 80% UO_2 -20% PuO_2 . The mixed sol is stored in an ice bath until microsphere forming is completed.

Microsphere Forming

1. Microspheres are formed in 2-ethyl-1-hexanol which contains 0.2 vol % Span 80, 0.3 vol % Pluronic 92, and 0.5 to 1.5 vol % water. The apparent pH of the alcohol is adjusted to 2.5-3.0 with HNO_3 . The alcohol is sparged with argon prior to and during microsphere forming.
2. Sphere residence time in the column should not exceed 35 minutes, and product spheres should be separated from residual solvent as soon as possible.
3. The gel spheres can then be steam dried and calcined to 1150°C in 4% H_2 -Ar.

2.3 - Sphere Preparation (P. A. Haas)

Information concerning the conversion of urania and urania-plutonia sols into microspheres of the desired size and with the desired properties is reported here. This includes studies on sphere-forming, chemistry of the sphere-forming column, recycle of the drying alcohol, drying and firing of gel spheres, and classification of the fired particles. The latest information on adapting these process steps and equipment to remote operation is also included.

Microsphere Preparation Development (P. A. Haas)

The effects on sphere forming of the NaOH scrub to remove formic and nitric acids from the 2-ethyl-1-hexanol were studied in a series of tests with a batch of concentrated CUSP sol ($\sim 2 \text{ M } \text{UO}_2$, batch no. EV-40) and a new charge of 40 liters of 2EH containing 0.4 v/o Span 80. Sol feed rate was held constant at 7.6 cc/min. In the first test sol

was fed for 0.9 hr. The apparent 2EH pH was 4.0. There was no sticking or clustering, but there was severe raisin distortion of product.

In the second test 0.1 v/o Ethomeen S/15 was added, and sol feed was resumed for 3.2 hr with no signs of sticking, clustering, or raisin distortion. The pH of the 2EH remained at about 5 without acid additions. After this time, sticking and clustering appeared and became excessive. A 2EH sample showed 260 mg/liter of HCOO^- .

Sol feed was cut off, and the 2EH was put through a caustic scrubber (0.5 M NaOH); the HCOO^- analyses were 67 mg/liter after 2-1/2 passes and 63 mg/liter after 5 passes through the scrubber. The 2EH pH was 6.0. The 6 liters of caustic scrub analyzed 1400 mg/liter HCOO^- .

After scrubbing the 2EH, sol feed was resumed for 2.5 hr without signs of sticking or clustering.

The presence of 260 mg/liter of HCOO^- (0.006 M HCOO^-) in the 2EH at the time clustering became excessive agreed with previous values of formic acid concentration in the 2EH at the time of appearance of clustering. The analyses show 8 g HCOO^- in the NaOH solution in agreement with the 7.5 g lost by the 2EH. The significance of the apparent leveling off of the HCOO^- concentration at 65 mg/liter in the scrubbed 2EH is not clear. The accumulation of HCOO^- in the 2EH was about 10% of the formic acid in the sol as compared to 20 to 30% transfer when the 2EH is scrubbed continuously. The cause of this difference is not known. It may be due to such things as the shorter holdup time for the 2 M UO_2 sol feed (the 20 to 30% values were for 1 M sols), or partial decomposition of formic acid in the still. With continuous caustic scrub of the 2EH, all the 2EH is scrubbed before it enters the still; thus part of the formate and nitrate are removed before they are subjected to the high temperature in the still.

Use of caustic scrubbers for the 2EH entering the still interferes with the operation of the microsphere forming column by causing entrainment of drops of caustic in the column. A membrane-type liquid-liquid separator was purchased from the Selas Corporation and was tested for preventing this entrainment. The Selas Separator appeared to prevent the entrainment, but the coalescing membrane plugged after 6 hr of use. The aqueous interface was maintained above this membrane for a second test (the interface was near the bottom for the first test). This change slowed down, but did not stop, the increase in pressure drop (indicating plugging) during operation. Even after the amount of entrainment was reduced by using a packed column with the organic phase continuous as the scrub column, the Selas Separator plugged. We do not plan any further tests of the separator for this application.

The contactor initially used for scrubbing the 2EH with 0.5 M NaOH was two aqueous continuous spray columns. This was changed to one 4-ft.-length of 1/2 in. ceramic Berl Saddle packing with the organic phase continuous. This packed column scrubber generally gives much less entrainment of caustic solution than the spray columns. The 2EH is generally a clear yellow color, with unobstructed visibility through the 6-in. ID glass column above the packed section.

However, there are short periods of high entrainment during which the aqueous flow distributor sometimes becomes obscured by the entrainment in a 1-1/2 in. thickness of 2EH. This high entrainment commonly occurs at startup or after additions of surfactants, but sometimes occurs without apparent cause.

In sphere forming studies, three CUSP UO_2 sols of high NO_3^-/U ratios have been better forming gel spheres than corresponding sols of the standard NO_3^-/U ratios. The results of these studies are as follows:

1. Sol EV-40, which had a NO_3^-/U ratio of about 0.18 gave better sphere formation than EV-41 of about 0.12 NO_3^-/U . These sols were similar, differing only in that EV-40 had a high conductivity and high NO_3^-/U after evaporation to concentrate it-- apparently some nitric acid wash solution which had been trapped in the evaporator system ran into it during evaporation.
2. The last liter of EV-41 was giving considerable clustering and some sticking in spite of the use of the NaOH scrub of the 2EH. Ten volume percent of 5% HNO_3 (about 0.04 HNO_3/U) was added and the sol was allowed to stand overnight. The sol then gave much better sphere formation.
3. About 0.05 HNO_3/U was added to 1 M UO_2 sol CUSP-ES-45. This sol was about 12 weeks old and clustered very badly when last run (at that time it was ten weeks old). The sol with added HNO_3 clustered much less than when last tested before HNO_3 addition.

CUSP sols with initial NO_3^-/U ratios above 0.11 usually gel at progressively lower U concentrations as the NO_3^-/U ratio is increased. Sols EV-41 and CUSP-ES-45 were both fluid at the outset, and the additions of nitric acid would be expected to have the effect of bringing the sols closer to gelation. Although thick sols are poor for forming spheres, sols at concentrations close to gelation generally exhibit better sphere forming behavior than sols farther from gelation as long as the sols do not thicken to the point that they are difficult to handle. Thus, an explanation for the observed improved sphere forming behavior upon addition of HNO_3 to the sols is that it brought them close enough to gelation to profit from this observed effect of approach to gelation.

2.4 Preparation of Sol-Gel Test Materials (W. T. McDuffee, P. A. Haas)

Preparation of sol-gel materials for use in other parts of the LMFBR fuel cycle program is reported here. This includes materials for irradiation capsules. Development of fuel fabrication techniques such as Sphere-Pac or pellet formation is also included.

Natural Urania Materials

Current needs for UO_2 sols, UO_2 -gel spheres, or dense, fired UO_2 spheres are supplied using CUSP sols. All outstanding requests for these materials either have been filled or the material has been prepared and is available for delivery. This includes the UO_2 -gel spheres, 1.5 kg of which are to be in the 400- to 600- μ diam range and 0.5 kg to be in the less than 44- μ range.

Enriched Urania Sol

We have devoted our efforts to the preparation of 2 kg (HM) of 93% enriched UO_2 sol this month. Because of criticality requirements, the sol is prepared by the precipitation-peptization method where batch size control can be used. This material is scheduled for use in fabricating pellet fuel for the EBR-II, Series II irradiations. A high quality, sediment-free $^{235}\text{UO}_2$ sol is required in order to reproducibly blend it with PuO_2 sol to the narrow range of Pu/U ratio required. To ensure that the most stable sol is obtained, we settle each one overnight and decant the supernate from any deposited solids. We currently have produced approximately 0.5 kg (HM) of sol meeting these requirements. Low yields from early batches in the series resulted from faulty instrumentation used in the reduction step. A careful study of the reduction step revealed that the reduction was complete long before the instruments indicated it was. When the reduction is continued much (~1- to 2 hr) beyond the point where U(VI) is absent, back oxidation of the U(IV) to U(VI) takes place along with reduction of the nitrate. Thus, if a test is made for U(VI) after the solution has been over-reduced, a positive indication of U(VI) is obtained. This makes the operators believe that reduction is incomplete, and, as a result, several runs were continued until there was so much reduction of the nitrate that U(IV) precipitation as the hydrous oxide occurred. During the course of uncovering and correcting the instrumental problems, it was demonstrated that the reduction of 0.55 M $\text{UO}_2(\text{NO}_3)_2$ is best conducted below 30°C, and that with 2 g of PtO_2 catalyst/liter the reaction is complete in 1 to 2 hr.

Once the reduction step was under control, we improved the techniques used for precipitation and washing the hydrous UO_2 cake. These improvements were better exclusion of air from the system, less vigorous agitation while reslurrying the UO_2 cake during the washing step, and a careful control of the wash solution velocity through the settled cake. It was necessary also to alter the technique of peptizing the washed UO_2 cake somewhat, thus permitting us to take into account the ability of the present equipment to produce a drier UO_2 cake than was formerly obtained. With the volume of peptizing acid formerly used, the cake was difficult to resuspend completely; this resulted in low yields. Once this deficiency was corrected, UO_2 sols of high quality were obtained from which little if any solids ("heel") deposited on standing overnight.

2.5 - Plutonium Storage Facility

(J. R. Parrott, R. G. Nicol, W. A. Shannon)

A facility for safely storing and handling plutonium is being built in Building 3019 to meet the needs for developmental work involving the sol-gel flowsheet for fast reactor fuels. This facility is located in the basement (room 501) adjacent to the Metals and Ceramics Division alpha laboratory. It will have a capacity of 100 kg of plutonium in a solid (fluoride) or a liquid (nitrate) form.

A preliminary criticality review was made of the system. Questionable points in the original analysis are being reexamined. The glass Raschig rings to be placed in the storage tank have been received and are being tested and analyzed to determine that they meet specifications as a fixed neutron absorber in solutions of fissile material.

III. FUEL FABRICATION AND EVALUATION

(A. L. Lotts, C. M. Cox, J. D. Sease, and
T. N. Washburn)

The primary emphasis of fuel fabrication and evaluation studies related to the fuel cycle for the liquid-metal fast breeder reactor is on oxide fuels. The objective of our program is to obtain an economically optimized (U,Pu)O₂ fuel cycle for a liquid-metal fast breeder reactor by extending the performance capability and advancing the fabrication technology of oxide fuels. These fuels have the most advanced technology and greatest potential for reliable operation in first-generation LMFBR's. They have been tested in fast-flux environments but as yet have not been exposed under actual prototypic conditions. Currently, the burnup and heat rate are limited to about 50,000 Mwd/metric ton and 16 kw/ft, respectively, based on irradiation experiments with fuels that are not necessarily optimized for thermal, chemical, and mechanical performance.

The capability of oxide fuels can possibly be improved by adjusting structures or void distribution in the fuels. We emphasize irradiating fuels derived from the sol-gel process with thoroughly characterized structures and void distributions different from those of the oxide fuels irradiated heretofore. These include fuels fabricated by Sphere-Pac, vibratory compaction, extrusion, and pelletization. We compare the performance of these with the performance of reference fuels such as pellets derived from mechanically blended powders and coprecipitated material. The development of computer programs to assist in the analysis of test results and the development of a mathematical model to predict the performance of a fuel rod are integrated with the test program.

1. FABRICATION DEVELOPMENT

(J. D. Sease)

The purpose of our fabrication development work is to provide suitable fabrication processes for sol-gel-derived materials and to fabricate irradiation test specimens and capsules for a variety of irradiation tests.

1.1 Process Development

This work is currently concerned with the engineering-scale development of the Sphere-Pac process and the development of sol-gel pelletization techniques for the fabrication of irradiation test specimens.

1.1.1 Sphere-Pac (C. R. Reese, W. L. Moore)

Sphere-Pac is a process for loading fuel pins with microspheres using vibratory energy. The objective of the Sphere-Pac development effort is to investigate the material and processing parameters that will allow utilization of the Sphere-Pac process in a production facility.

During the previous month an effort was made to predict and control the coarse bed density of the loaded bed. Using micromesh screens and U.S. standard sieves, shape-separated thorium microspheres were divided into various size fractions ranging from 100 to 700 μ . Different batches with size frequency distribution curves of various shapes were mixed by weight. Table 1 lists the mathematical formulae describing the distributions considered. Each distribution was loaded into a stainless steel pin 16 in. long $\times$ 0.245-in. ID, and the coarse bed density was measured. As seen in Table 1, the shape of the distribution exhibits no appreciable effect on the coarse bed density.

Table 1. Effect of Particle Size Distribution Shape
Upon Coarse Bed Density

Distribution	Coarse Bed Density (% T.D.)
$n(D) = \text{constant (420 to 600 } \mu)$	60.6
$n(D) = -50 + 0.12 D$	60.3
$n(D) = 77.1 - 0.12 D$	60.8
520- to 540- μ cut	61.2
50% 520-540 μ	60.6
50% 420-440 μ	
$\frac{1}{\sqrt{2\pi}} e^{-1/2 Z^2}$	60.6
where $Z = \frac{\text{diameter} - \text{avg. diameter}}{\text{standard deviation}}$	

$n(D)$ = frequency per unit diameter

D = particle diameter in microns

Batches were mixed employing normal size frequency distribution curves with varying mean particle sizes. Each batch was loaded by three different operators. To determine the effect of operator loading techniques on the coarse bed density, each operator loaded the pin to the point that was considered to be the equilibrium density. However, as can be seen in Table 2, the coarse bed densities obtained by each operator varied. The variance in densities obtained with different operators is attributed to the lack of reproducibility of the loading technique employed to obtain maximum coarse bed densities. Work is commencing to develop a uniform loading technique. An electronic variable frequency vibration system is being evaluated in an effort to minimize loading technique variance.

The preliminary data depicted in Table 2 indicate that the coarse bed density increases as the mean particle size decreases. Each coarse bed was infiltrated with fine thoria microspheres of the size range 5 to 45 μ . However, the infiltrated density does not follow the same trend as the coarse bed density. As shown by the infiltrated densities listed in Table 2, the ability to infiltrate the coarse bed with fine microspheres of the size 5 to 45 μ decreases as the mean size of the coarse bed distribution increases. Further work is being done to determine the effect of the mean particle size of the coarse bed distribution upon the density of the coarse bed and infiltrated density. The effect of the spread of the coarse bed distribution will also be investigated. Preliminary data shown in Table 2 indicate that the coarse bed density increases as the ratio of the maximum particle size to the minimum particle size of the coarse bed distribution increases. Experiments will be conducted in which the mean particle size of the normal distribution will be held constant while the spread of the distribution is varied to determine the effect of the distribution spread upon the coarse bed density.

During the month we made photographs of the coarse fraction of Sphere-Pac loaded beds. The beds were prepared by soldering screens to the bottom of 0.186-in. OD $\times$ 0.245-in. ID, 6-in. long tubes. Thoria microspheres having a normal distribution in the 530- to 590- μ range were Sphere-Pac loaded into the pins and a screen was soldered to the pin tops to prevent bed shifting. After being impregnated with resin, the pins were sectioned longitudinally and transversely and polished. Photographs shown in Figs. 1 and 2 were made of the sections utilizing bright field and side lighting. Only short range ordering of the microspheres was observed in both sized tubes. Due to the lack of ordering, it is not presently possible to apply mathematical formulæ to analyze Sphere-Pac behavior of distributed particle sizes. It is felt that the lack of ordering results in inability to obtain a true equilibrium bed density, i.e., loading technique will affect bed density.

Table 2. Packing Density of Normally Distributed Coarse Beds
Before and After Infiltration

Mean Size (μ)	Particle Size (Max/Min)	Coarse Bed Density (% T.D.)			Infiltrated Density (% T.D.) Operator 3
		Operator 1	Operator 2	Operator 3	
250	3.4	64.6	61.9	62.4	66.9
325	5.2	63.4	62.6	61.8	66.0
435	2.9	61.1	60.8	60.9	81.7
535	2.3	60.9	60.5	60.6	82.4

Fig. 1 Transverse section of 530-590 μ
normally distributed 0.186-in. ID thoria
Sphere-Pac bed. 11.4X

Fig. 2 Longitudinal section of 530-590 μ
normally distributed 0.186-in. ID thoria
Sphere-Pac bed. 11.4X

1.1.2 Pelletization of Sol-Gel Urania-Plutonia (R. A. Bradley)

Introduction. - ETR irradiation capsules 43-120 and 43-121 will each contain two pins loaded with $(\text{Pu}_{0.20}, \text{U}_{0.80})\text{O}_2$ microspheres by the Sphere-Pac process and two pins loaded with 84% dense sol-gel $(\text{Pu}_{0.20}, \text{U}_{0.80})\text{O}_2$ pellets. For both the pellets and microspheres, the O/M must be 1.98 ± 0.01 and the gas release less than 0.10 cc/g. During the past month our work has been concerned with fabricating the pellets for the capsules, adjusting the O/M of the microspheres, and characterizing the fuel.

Experimental Procedure. - We prepared a 1300-g batch (ET3) of urania-plutonia powder by blending UO_2 and PuO_2 sols and drying to a gel at 100°C with an argon purge. After grinding in a fluid energy jet-mill, sieving with a 325-mesh screen and blending, the powder was subdivided into 100-g batches (designated ET3A and ET3M) for calcination. Samples of the blended sol, dried gel shards, jet-milled powder, and the fines collected in the dust bag of the jet-mill exhaust were analyzed for U and Pu. The bulk density of the dried gel shards and the surface area of the shards, the jet-milled powder, and the fines were also determined. The results of all these analyses are summarized in Table 3.

A 100-g batch (ET3A) was calcined by heating in argon to 520°C , holding for 4 hr, and then cooling in CO_2 . Another batch (ET3B) was calcined in the same manner except that Ar-4% H_2 was used instead of argon. Sinterability tests were performed on each batch of calcined powder. Pellets were pressed at 20,000 to 60,000 psi and sintered in Ar-4% H_2 by heating at 300°C/hr to 1450°C and holding for 10 hr. The forming pressures, densities, and shrinkages of the pellets produced in these tests are summarized in Table 4. After analyzing the results of these tests, we decided to calcine five additional batches (ET3C-ET3G) in Ar-4% H_2 to produce enough powder for the ETR pellets. The calcined powder was characterized by visual appearance (primarily color), surface area, oxygen-to-metal ratio, and the green density of pellets formed at an arbitrarily selected pressure. The results of this characterization are summarized in Table 5.

A sinterability test was performed on each of the seven batches of calcined powder by pressing three pellets from each batch at 55,000 psi and sintering according to the schedule described previously. The results of this test are given in Table 6. All seven batches were then blended to form a uniform batch of powder (ET3N) for fabricating the ETR pellets. After the powder was blended, a final test sinter was made to determine the pressure required to produce 84% dense pellets. The forming pressures, green densities, and sintered densities of these pellets are given in Table 7. This test indicated that a forming pressure of 41,500 psi would yield 84% dense pellets; therefore, four batches of 40 pellets each were pressed at that pressure and sintered by the previously described schedule. To aid in reducing the O/M, a roll of Ta foil was placed upstream and adjacent to the pellets to serve as a getter. Six control pellets, two each at the left end, center, and right end of

Table 3. Summary of Analyses on Blended Sol, Dried Gel Shards,
and Jet-Milled Powder for Batch ET3

Material Form	Uranium	Plutonium	$\frac{\text{Pu}}{\text{U} + \text{Pu}}$	Bulk Density (g/cc)	Surface Area (m ² /g)
Blended sol	199.4 $\frac{\text{mg}}{\text{ml}}$	47.7 $\frac{\text{mg}}{\text{ml}}$	0.193	—	—
Dried gel shards	68.56%	15.33%	0.183	4.40 ^a 4.48 ^b	36.2
Jet-milled powder	67.32%	16.13%	0.193	—	44.2
Jet-milled fines	64.45%	14.76%	0.186	—	51.3

^a Determined by mercury porosimetry at atmospheric pressure.

^b Determined by mercury porosimetry at 10,000 psi.

Table 4. Summary of Densities and Shrinkages for Sinterability Tests
on Powder Batches ET3AH95 and ET3BH96

Sintering Run	Powder Batch	Pressure $\times 10^{-3}$ (psi)	Avg. Gr. Dens. (% T.D.)	Avg. Sin. Dens. (% T.D.)	Length Shrinkage (%)	Diameter Shrinkage (%)
99	ET3AH95	20	46.6	73.0	12.9 $\begin{smallmatrix} + 1.0 \\ - 0.7 \end{smallmatrix}$	15.4 $\begin{smallmatrix} + 0.0 \\ - 0.0 \end{smallmatrix}$
99	ET3AH95	30	48.8	78.0	14.1 $\begin{smallmatrix} + 0.6 \\ - 0.7 \end{smallmatrix}$	15.4 $\begin{smallmatrix} + 0.0 \\ - 0.0 \end{smallmatrix}$
99	ET3AH95	40	50.5	81.0	14.3 $\begin{smallmatrix} + 0.1 \\ - 0.0 \end{smallmatrix}$	15.4 $\begin{smallmatrix} + 0.0 \\ - 0.0 \end{smallmatrix}$
99	ET3AH95	50	52.2	83.6	14.3 $\begin{smallmatrix} + 0.2 \\ - 0.1 \end{smallmatrix}$	15.4 $\begin{smallmatrix} + 0.0 \\ - 0.0 \end{smallmatrix}$
100	ET3AH95	45	51.3	82.4	13.8 $\begin{smallmatrix} + 0.9 \\ - 1.3 \end{smallmatrix}$	15.8 $\begin{smallmatrix} + 0.0 \\ - 0.0 \end{smallmatrix}$
100	ET3AH95	50	51.8	83.9	14.1 $\begin{smallmatrix} + 0.3 \\ - 0.3 \end{smallmatrix}$	15.8 $\begin{smallmatrix} + 0.0 \\ - 0.1 \end{smallmatrix}$
100	ET3AH95	55	53.2	85.9	13.9 $\begin{smallmatrix} + 0.3 \\ - 0.3 \end{smallmatrix}$	15.6 $\begin{smallmatrix} + 0.0 \\ - 0.0 \end{smallmatrix}$
100	ET3AH95	60	53.3	85.5	14.1 $\begin{smallmatrix} + 0.2 \\ - 0.4 \end{smallmatrix}$	15.4 $\begin{smallmatrix} + 0.1 \\ - 0.0 \end{smallmatrix}$
100	ET3BH96	30	43.3	79.2	18.1 $\begin{smallmatrix} + 0.2 \\ - 0.2 \end{smallmatrix}$	19.1 $\begin{smallmatrix} + 0.1 \\ - 0.1 \end{smallmatrix}$
100	ET3BH96	40	44.6	81.5	17.9 $\begin{smallmatrix} + 0.1 \\ - 0.1 \end{smallmatrix}$	19.2 $\begin{smallmatrix} + 0.0 \\ - 0.0 \end{smallmatrix}$
100	ET3BH96	50	46.5	83.0	17.5 $\begin{smallmatrix} + 0.1 \\ - 0.1 \end{smallmatrix}$	18.8 $\begin{smallmatrix} + 0.2 \\ - 0.2 \end{smallmatrix}$
100	ET3BH96	60	49.7	89.4	17.2 $\begin{smallmatrix} + 0.1 \\ - 0.2 \end{smallmatrix}$	18.5 $\begin{smallmatrix} + 0.0 \\ - 0.0 \end{smallmatrix}$

Table 5. Characterization of Urania-Plutonia Powder
Calcined for ETR Pellets

Batch No.	Calcining Conditions			Atm	Surface Area (m ² /g)	O/M	Gr. Dens. @ 55,000 psi (% T.D.)	Appearance
	Heating Rate (°C/hr)	Max Temp (°C)	Time at Temp (hr)					
ET3AH95	300 Cool	500 —	4 —	Argon CO ₂	7.63	2.226	53.9	Black
ET3BH96	400 Cool	520 —	4.1 —	Ar-4% H ₂ CO ₂	26.7	2.365	49.3	Brown
ET3CH97	360 Cool	520 —	4.1 —	Ar-4% H ₂ Ar-4% H ₂	35.5	2.441	47.4	Yellowish brown
ET3DH98	360	600	0	Ar-4% H ₂	—	—	47.7	Color between ET3B-ET3C
	Cooling	600-520	0.5	Ar-4% H ₂	—	—	—	
	Hold	520	3.25	Ar-4% H ₂	—	—	—	
	Cool	—	—	CO ₂	—	—	—	
ET3EH99	380	520	4	Ar-4% H ₂	—	—	46.0	Color between ET3B-ET3C
	Cool	—	—	CO ₂	—	—	—	
ET3FH100	380	520	4	Ar-4% H ₂	—	—	46.1	Color between ET3B-ET3C
	Cool	—	—	CO ₂	—	—	—	
ET3GH101	360	520	4	Ar-4% H ₂	—	—	45.9	Color between ET3B-ET3C
	Cool	—	—	CO ₂	—	—	—	
ET3N H95-H101	Blend of batches ET3AH95-ET3GH101				(a)	2.404	47.3	Color between ET3B-ET3C

^a Results not yet received.

Table 6. Summary of Densities and Shrinkages for Sinterability
Test of Seven Calcine Batches

Powder Batch No.	Pellet No.	Green Density (% T.D.) ^a	Avg. Density (% T.D.) ^a	Sintered Density (% T.D.) ^b	Avg. Sin. Dens. (% T.D.) ^b	Length Shrinkage (%)	Diameter Shrinkage (%)
ET3AH95	1105	53.7	53.9	85.1	85.3	13.7	15.5
	1106	53.5		85.2		13.9	15.5
	1107	54.4		85.5		13.6	15.4
ET3BH96	1108	49.3	49.3	81.5	81.4	17.5	15.4
	1109	49.3		81.5		17.2	15.5
	1110	49.2		81.1		16.9	15.4
ET3CH97	1111	47.4	47.4	90.2	89.4	18.7	20.3
	1112	47.2		89.1		19.1	19.9
	1113	47.7		89.0		18.6	19.9
ET3DH98	1114	45.9	47.7	86.2	86.2	22.7	17.9
	1115	(c)		(c)		(c)	(c)
	1116	49.6		86.1		16.5	17.8
ET3EH99	1117	45.5	46.0	81.7	84.8	17.8	19.0
	1118	46.0		84.8		18.0	19.4
	1119	46.4		84.9		17.9	19.3
ET3FH100	1120	46.7	46.1	86.5	84.7	18.3	19.5
	1121	45.0		82.5		17.5	19.7
	1122	46.6		85.1		17.6	19.3
ET3GH101	1123	45.4	45.9	82.4	84.8	19.4	20.0
	1124	45.9		85.2		18.3	19.7
	1125	46.3		86.7		18.5	19.8

^a Pressed at 55,000 psi.

^b Sintered in Ar-4% H₂ at 1450°C for 10.5 hr.

^c Broke pellet before sintering.

Table 7. Results of Sinterability Test on Batch ET3N^a

Forming Pressure $\times 10^{-3}$ (psi)	Green Density (% T.D.) ^b	Sintered Density (% T.D.) ^b
40	45.1	83.4
45	45.9	85.2
50	46.9	86.1
55	47.3	87.7

^a Batch ET3N obtained by blending batches ET3AH95-ET3GH101.

^b Based on theoretical density of 11.06 g/cc.

the boat were dimensionally inspected to determine green and sintered density. The moisture level in the furnace effluent gas was monitored to aid in controlling O/M. One pellet from each sintering run was analyzed for O/M. The pellets were ground to the required diameter on a centerless grinder and then the O/M and gas release were determined for each sintering run. The green and sintered densities of the control pellets, H₂O in effluent gas, the O/M before and after grinding, and the gas release for each of the four sintering runs are summarized in Table 8.

Results and Discussion. - The ratio of Pu to heavy metal (U + Pu) in the blended sol was 0.193; however, in the dried gel shard, the ratio was 0.183 (see Table 3). Only one analysis was run on the shards so the results are not conclusive, but there is a possibility of some segregation of Pu and U during drying. The jet-milled powder was found to contain 0.193 wt fraction plutonium, indicating that the effects of any segregation which might occur during drying are no longer evident after the material has been blended in the grinding operation. The fines collected in the dust bag of the jet-mill exhaust contained 0.186 wt fraction plutonium. This analysis was performed to see if there was any gross segregation of Pu and U during grinding. The results indicate that the amount of separation, if any, is very small. Mercury porosimetry measurements on the dried gel shards show the bulk density, measured at atmospheric pressure, to be 4.40 g/cc and the density measured at

Table 8. Densities, Oxygen-to-Metal Ratio, and Gas Release of Pellets
Sintered in Four Production Runs for ETR

Sintering Run	Position in Boat	Green Density (% T.D.) ^a	Sintered Density		Average Density (% T.D.) ^a	H ₂ O in Effluent Gas (ppm)	O/M		Gas Release (cc/g)
			(% T.D.) ^a	(% T.D.) ^b			Before Grinding	After Grinding	
103	Left end	44.3	83.4	84.6	84.5	26	1.991	1.946	0.91
	Center	44.5	84.1	85.3					
	Right end	45.1	81.4	82.6					
104	Left end	45.0	82.2	83.4	84.3	27	1.974	1.940	1.00
	Center	45.0	86.7	87.9					
	Right end	45.2	84.6	85.8					
105	Left end	45.3	83.1	84.2	84.1	15	1.967	1.952	(c)
	Center	45.4	85.5	86.7					
	Right end	45.5	84.3	85.5					
106	Left end	45.3	83.9	85.1	83.0	23	1.963	1.943	(c)
	Center	45.3	85.2	86.4					
	Right end	45.4	83.3	84.5					

^a Based on theoretical density of 11.06 g/cc. (O/M = 2.00)

^b Based on theoretical density of 10.91 g/cc. (O/M = 1.945)

^c Results not yet available.

10,000 psi to be only 4.48 g/cc. Since a pressure of 10,000 psi is sufficient to force mercury into 170Å openings, the measurement at 10,000 psi indicates that either

1. the porosity is closed,
2. the pore size is less than 170Å or at least the passages connecting the pores are less than 170Å, or
3. the crystal density is 4.48 g/cc.

With mercury porosimetry alone, one cannot determine which of the above is correct; however, in conjunction with BET surface area measurements, this can be resolved. The surface area of the dried gel shards is about 36 m²/g, which corresponds to a particle size in the range of 100 to 200Å. This indicates the dried gel shards are made up of agglomerates of 100 to 200Å crystallites with interconnected porosity of less than 170Å.

The surface area of the jet-milled powder was about 44 m²/g, indicating the basic character of the particles was not changed during grinding; the particles were only reduced in size. This is the first time that we have observed an increase in surface area during jet-milling; normally it decreases slightly. The higher surface area of the fines (51.3 m²/g) is consistent with previous observations.

The sinterability tests on the two batches of powder calcined in argon (ET3AH95) and in Ar-4% H₂ (ET3BH96) indicated that either type powder could be used for making 84% dense powder. The green densities were lower and shrinkages higher for pellets pressed from the powder calcined in Ar-4% H₂; however, this powder appeared to press slightly better so it was selected for further work.

The high surface areas, 26.7 and 35.5 m²/g, obtained on calcined batches ET3BH96 and ET3CH97, respectively, were surprising (Table 5). We expected surface areas less than 10 m²/g. The surface area analysis on batch ET3N, obtained by blending seven calcine batches, has not been received. The O/M of the powder, ranging from 2.22 to 2.44 is a little higher than the 2.15 to 2.25 we were attempting to achieve. The higher surface area would account for this because a high surface area powder would oxidize more readily when cooled in CO₂. One batch (ET3CH97) was inadvertently cooled in Ar-4% H₂ instead of CO₂. After a small amount of handling in the argon-purged (but not high purity atmosphere) glove-box, this material had a yellowish brown color, indicating considerable surface oxidation. All of the other powder calcined in Ar-4% H₂ was brown, while that calcined in argon was black.

The results of the sinterability tests on the seven calcine batches prior to blending indicated that five of the seven batches would be suitable for making 84% dense pellets (Table 6). The pellets from batch ET3BH96 were only 81.4% dense. No explanation could be found for the lower density of these pellets. The pellets from batch ET3CH97 were 89.4% dense; however, this was probably due to the increased surface

oxidation of this powder because it was not stabilized in CO_2 . We decided to blend all seven batches, expecting that the low density obtained with batch ET3BH96 would compensate for the high density in batch ET3CH97.

The test sinter on the blended batch of powder (ET3N) produced sintered pellet densities (Table 7) about 1 to 2% higher than we expected, based on the results of sintering individual batches. A different die was used in pressing these pellets, but this does not appear to be the cause of the density variation since the green densities of the pellets corresponded closely with those of pellets pressed from the individual batches. The source of variation might be additional oxidation of the powder as it was handled. If this was the case, our stabilization treatment was not effective. We will continue to investigate this.

The average density of the six control pellets in the four production sintering runs was 84.5, 84.3, 84.1, and 83.0% (Table 8). In each run there was a variation in density due to position in the boat, with the density being 2 to 3% lower at each end of the boat than it was in the center. This indicates that the temperature profile is not constant over the 4-in. length of the boat. We have not calculated the densities of the remaining thirty-four pellets from each run so we do not know what the yield will be of pellets meeting the $84 \pm 1\%$ specification. One pellet from each sintering run was sent for O/M analysis before centerless grinding. The O/M's of the four runs were 1.991, 1.974, 1.967, and 1.963; however, after removing about 0.008 in. from the surface on the grinder, the O/M's were 1.946, 1.940, 1.952, and 1.943. Based on the thermodynamic data of Rand and Markin,¹ one would expect an O/M of 1.94 with 15 ppm H_2O and 1.955 with 26 ppm H_2O ; therefore, the lower O/M's are fairly consistent with the moisture observed in the effluent gas (Table 8). Although the pellets were stored in a closed container that was evacuated and backfilled with dry argon, the gas release was extremely high (~ 1.0 cc/g). We do not know the reason for the high gas release yet, but we believe it to be associated with an increased affinity for moisture in the low O/M pellets. We are looking further into this problem now. We intend to heat treat all of these pellets to remove the gas and then readjust the O/M to 1.98. The gas release is usually less of a problem when the O/M is not so low.

Future Work. - During the next month we expect to load the ETR fuel pins and continue preparation of a large batch of powder for the EBR-II pellets.

¹ M. H. Rand and T. L. Markin, Some Thermodynamic Aspects of (U,Pu) O_2 Solid Solutions and Their Use as Nuclear Fuels, AERE-R5560, August 1967.

1.2 Capsule Fabrication

The two instrumented TREAT capsules (TREAT I and II) were shipped to the TREAT reactor. Planning for the fabrication of the EBR-II, Series II fuel rods continued.

1.2.1 TREAT Capsule Fabrication (E. J. Manthos, W. L. Moore)

The two completed TREAT capsules were shipped to the Reactor site on October 14, 1969.

1.2.2 EBR-II, Series II Unencapsulated Fuel Rods (R. B. Pratt, M. K. Preston)

We are currently analyzing requirements for fabricating 19 unencapsulated fuel rods which will be irradiated in a shared 37-rod subassembly in the EBR-II Reactor.

During the month a sequence diagram showing interactions of 1000 activities necessary for fabricating 19 EBR-II fuel pins was completed. The unlevelled computer output shows a predicted completion date of May 1, 1970. We are currently examining means for reducing the job duration in order to meet the proposed schedule. The critical path is through activities associated with the fabrication of UO_2 blanket pellets. This occurs because blanket pellets must be available prior to loading the first fuel rod, and 43 days are estimated for obtaining the UO_2 powder. We are currently looking into the possibility of obtaining powder from other sources in order that these activities might proceed at an earlier time. If as many as 24 days can be cut from this schedule, the critical path will most probably shift to the Sphere-Pac preparation activities. To date, we are currently awaiting AEC-RDT approval to fabricate the 19 fuel pins. This approval is expected by early November. We are also awaiting information concerning detailed design of the fuel element hardware. Argonne National Laboratory has been requested to send us the required design information early in November.

We have received 700 ft of 0.250-in. OD $\times$ 0.218-in. ID 316 stainless steel seamless tubing in 7 ft lengths from Battelle Northwest to fabricate the EBR-II, Series II fuel pins. We have started inspecting this tubing to obtain tubes suitable for the ETR fuel pins which require tube lengths of 7 9/16 ft and 33 3/4 in., while EBR-II will require tubing in lengths of approximately 40 in. Longitudinal and transverse ultrasonic inspections have been made on 11 tubes with a 0.001-in. deep $\times$ 0.004-in. wide $\times$ 0.030-in. long match standard. Tubing areas with flaws less than the standard are acceptable for EBR-II use, while tubing area with flaws equal to but not greater than the reference standard are acceptable for ETR use. The following table gives the tubes obtained for the three different fuel pins we are now fabricating. (Table 9).

Table 9. Tubes Obtained From Ultrasonic Inspection of EBR-II Tubing

Tube No.	EBR-II	33 3/4-in. ETR	7 9/16-ft ETR
19-1 ^a		1	
19-2		1	3
19-3	1 (51 1/2 in.)		3
19-4			9
19-5		2	
19-6	1 (73 1/2 in.)		1
19-7	1 (40 1/2 in.)	1	
19-8	1 (49 in.)		3
19-9	1 (44 1/16 in.)		4
19-10	1 (60 1/4 in.)		1
19-11			7
TOTAL	6	5	31

^a 36 in. of 19-1 used to fabricate two inspection standards.

1.2.3 ETR Instrumented Capsules (M. K. Preston)

As our next thermal flux irradiation test of mixed oxide fuels, we are planning a series of four relatively simple instrumented capsules for the ETR. The capsules, which will be double encapsulated with NaK heat transfer bond, will contain thermocouples for monitoring the clad surface temperatures. In two of the capsules, the fuel column will consist of four separate 3-in. fuel sections with a total fuel pins stack height of 34 in. while the other two fuel columns will consist of a single fueled section of 20 in. The cladding for the fuel pins will be the same as that we plan to use in our Series II, EBR-II experiments, 316 stainless steel 0.250-in. OD $\times$ 0.015-in. wall thickness. Fabrication of the two capsules with the 3-in. fueled sections has begun.

Capsule components for weld development, for dummy fabrication to check out procedures, and for fabrication of the two 3-in. fueled stacked units have been completed. Weld development is 90% complete with only two welds still in development to comply with the ETR standards.

Loading equipment has been assembled for loading NaK-44 (44% K) into the capsules. The equipment is presently being leak checked and practice runs should be made on dummy assemblies the first week in November.

Assembly procedures are being prepared, along with data sheets, for the capsules with the stacked 3-in. fueled sections. These are being reviewed and checked out on dummy hardware.

Fuel pin subassemblies consisting of the tubing and bottom end plugs have been welded and inspected and are ready to be loaded with fuel.

2. FUEL EVALUATION

(C. M. Cox, T. N. Washburn,
J. M. Leitnaker)

The fuel evaluation work involves both out-of-reactor and in-reactor tests to characterize the fuels of interest and to determine performance limits of the fuels.

2.1 Characterization of Fuels

The development of sol-gel fuel fabrication requires characterization of the material to control the process and to determine which properties are important to irradiation behavior. Characterization requires determining both the chemical composition and the physical properties. Thermodynamic studies contribute to the development of the process for the fuel and aid in predicting fuel performance for both irradiation testing and model studies.

2.1.1 Analytical Chemistry (W. H. Pechin)

Several gas release measurements were made on (U,Pu)O₂ microspheres which suggest a new facet to the problem of gas evolution by this fuel at elevated temperature. The microspheres prepared for ETR capsules were found to have gas release values of 0.71 cc/g for the fines and 0.19 cc/g for the coarse. As this material has a very low oxygen-to-metal ratio (1.968) and is, therefore, presumably quite susceptible to moisture pickup and as some time had elapsed between the sampling and the analysis, new samples were submitted for analysis. The second analysis resulted in values of 0.48 cc/g for the fines and 0.17 cc/g for the coarse, indicating that the initial high values could not be completely accounted for by degradation of those samples.

As a guide to salvaging this material, a third sample of fines was analyzed to determine the temperature at which the major portion of the gas was being evolved. The results of this analysis are shown in Table 10. The largest apparent gas evolution occurred at or below 900°C - the lowest temperature routinely measured with the apparatus. Above 1200°C the pressure decreased, indicating that some of the gas was being readsorbed. This decrease continued until the 1600°C measurement, at which temperature the trend was reversed. A third sample of coarse was then analyzed in the usual manner resulting in a value of 0.19 cc/g which is in excellent agreement with the two previous samples of this material, and indicates that the equipment was functioning normally.

The final gas volume listed in Table 10 would normally be considered in good agreement with the results on other samples of this material. The behavior of pressure with temperature is not what one would expect however. The increase on heating from 900 to 1200°C is expected, but a decrease of almost that same magnitude on continued heating to 1500°C is completely unexpected. While these variations are small compared to the total gas release from this particular sample, they are quite significant compared to the desired gas release of 0.10 cc/g or less.

Table 10. Gas Release From (U,Pu)O₂ Fine Microspheres
as a Function of Temperature

Temp (°C)	Pressure (mm Hg)	Cumulative Volume (std cc/g)
900	0.52	0.384
1000	0.56	0.414
1100	0.57	0.422
1200	0.57	0.422
1300	0.56	0.414
1400	0.54	0.399
1500	0.53	0.392
1600	0.55	0.409

A possible explanation of this behavior is suggested by the occurrence of PuH_2 and its similarity to PuO_2 and UO_2 as illustrated in Table 11. It is possible that hydrogen could occupy some of the oxygen lattice sites of the hypostoichiometric oxide with a decreasing equilibrium concentration at increasing temperatures. The explanation for the gas release could be explained as follows: At low temperatures adsorbed or chemisorbed water is released as vapor; as the temperature increases, this water reacts with the oxide to form H_2 which then is picked up by the solid. Upon further heating, the hydrogen is reevolved as gas. In the rapid heating test, water would still react with the oxide to form H_2 , but most of the H_2 would be formed at temperatures not suitable for its retention by the lattice.

If such a mechanism occurs, it has serious implications in the processing of the material since the unoccupied lattice sites in a hypostoichiometric oxide may be extremely reactive to a number of elements.

Table 11. Similarities Between PuH_2 , PuO_2 , and UO_2

Compound	Crystal Structure Type	Lattice Parameter Å
UO_2^a	CaF_2	5.4705
PuO_2^b	CaF_2	5.396
$(\text{U}_{0.8}, \text{Pu}_{0.2})\text{O}_{1.95}^c$	CaF_2	5.479
PuH_2^b	CaF_2	5.395

^a W. A. Yound et al., NAA-SR-6765, 1962.

^b R. P. Elliot, Constitution of Binary Alloys, First Supplement, pp. 503, 509, McGraw Hill, New York, 1965.

^c B. J. Seddon, Physical Properties of Some Plutonium Ceramic Compounds. A Data Manual, TGR 1601, 1968, p. 11.

2.1.2 Alkali Metals-Uranium-Oxygen Investigations (J. M. Leitnaker, K. E. Spear)

Crystals have been recently observed growing from irradiated mixed oxide fuel material by hot-cell personnel. The location of these crystals, on the fuel periphery, suggested that they grew from a fission product. The general appearance of the crystals, white to clear, was suggestive of growth of crystals recently observed on sodium uranate samples. These uranate samples had been deliberately vibratorily polished with water as

the polishing medium and set aside for several days. A white growth was observed forming on the surface and, after a period of several weeks, covering the exposed portion of the sample. X-ray diffraction measurements indicated primarily $\text{Na}_2\text{CO}_3 \cdot \text{H}_2\text{O}$ and traces of other phases which were tentatively identified as $\text{Na}_6\text{U}_7\text{O}_{24}$ and $\text{Na}_4(\text{UO}_2)(\text{CO}_3)_3$. Chemical analysis of the white material indicated about 2.6% U.

Hot-cell personnel have identified Cs in the white crystals growing from the fuel sample. This, in our opinion, further confirms identification as an alkali metal hydrous carbonate. In any case, this appears to be a new and potentially useful technique for pinpointing the location of Cs, the way various treatments affect its concentration and location, and will possibly give further information on fuel-fission product-cladding interaction.

2.1.3 Rare Earth Solution in Mixed Oxides (J. M. Leitnaker, K. E. Spear)

We discussed previously² the importance of the solution of these fission products with relation to oxygen content and fuel swelling. During the present period we have started a more thorough evaluation of the oxide literature.

Based on work by Brauer and Gradinger,³ Ferguson and Fogg⁴ have reported a general relationship between lattice parameter of a fluorite structure solid solution extrapolated to pure solute when the solute is a cubic sesquioxide and the solvent is a dioxide of fluorite structure. This relationship is

$$a_f = 0.3943 a_c + 1.2285 \quad (1)$$

where a_f is the extrapolated lattice parameter in angstroms and a_c is the lattice parameter of the bcc sesquioxide. Lattice parameter data for the rare earth sesquioxides of the D_{5d} type are available for all but Ce_2O_3 , Pr_2O_3 , and Pm_2O_3 ; however, reasonable estimates can be made for these by interpolation. Thus, in the fluorite phase [which includes all fission product rare earth concentrations of interest in UO_2 , PuO_2 , and $(\text{U,Pu})\text{O}_2$] a systematic approximation to $\text{MO}_2\text{-(RE)}\text{O}_{1.5}$ solid solution lattice parameters can be made.

² J. M. Leitnaker, K. E. Spear, "Fuel-Cladding Compatibility Studies for Fast Breeder Reactors," Fuel Cycle Technology Monthly Progr. Rept. August 1969, September 1969.

³ G. Bauer and H. Gradinger, "(Über anomale Mischkristalle im Bereiche des Fluoritgittertyps," Naturwiss. 38, 559 (1951).

⁴ I. F. Ferguson and P.G.T. Fogg, "The Oxides of Uranium. Part VIII. The System Uranium Dioxide-Yttria," J. Chem. Soc., 3675 (1957).

Ferguson and Fogg⁵ show that Equation (1) is closely obeyed in the system $\text{UO}_2\text{-Y}\text{O}_{1.5}$. The work of Lynch⁶ shows that, within the experimental error, the $\text{UO}_2\text{-LaO}_{1.5}$ system also obeys the above general relationship. We therefore conclude that the relationship is generally applicable and will use it until other data are available. In Table 12 we tabulate the lattice constants of the imaginary fluorite $\text{REO}_{1.5}$ phase from which a straight line interpolation to UO_2 , PuO_2 , and $(\text{U,Pu})\text{O}_2$ is deemed reasonable.

Our own work, as well as previous work by a number of investigators, indicates that the solutions of $\text{UO}_2\text{-REO}_{1.5}$ are easily oxidized, although quantitative numbers have not been determined. Anderson *et al.*⁷ indicate $\text{Y}\text{O}_{1.5}\text{-UO}_2$ solid solutions "oxidize vigorously at temperatures of -20 to $+20^\circ\text{C}$." Therefore, at this juncture, it is difficult to fix with any confidence the lattice constant versus composition of $(\text{U,RE})\text{O}_{2-x}$ compositions; most of the literature data appear to be badly scattered and not determined as a function of oxygen potential. The data of Aitken and Joseph,⁸ however, appear to be good in this regard. Their experiments were performed at a constant oxygen potential ($P_{\text{O}_2} = 10^{-5.5}$) over the low " YO_2 " position of the range. Extrapolation of their linear data between $\text{U}_{0.81}\text{Y}_{0.19}\text{O}_2$ and $\text{U}_{0.54}\text{Y}_{0.46}\text{O}_2$ to pure, imaginary YO_2 yields a lattice parameter of $5.398\text{\AA}$. Thus, oxidation of $\text{Y}\text{O}_{1.5}$ in the imaginary fluorite phase to the imaginary YO_2 fluorite phase decreases the lattice parameter by $0.186\text{\AA}$. Our data with $\text{U}_{0.826}\text{Ce}_{0.173}\text{O}_{2-x}$

⁵ I. F. Ferguson and P.G.T. Fogg, "The Oxides of Uranium. Part VIII. The System Uranium Dioxide-Yttria," J. Chem. Soc. 3675 (1957).

⁶ E. D. Lynch, "Oxidation and Phase Stability in Solid Solution Systems Containing Uranium Oxide," in Characterization of Uranium Dioxide, TID-7637, 1962.

⁷ J. S. Anderson, I. F. Ferguson, L.E.J. Roberts, "Anionic Vacancies in Fluorite-type Oxides," J. Inorg. Nucl. Chem. 1, 340-1 (1955).

⁸ E. A. Aitken and R. A. Joseph, "Thermodynamic Study of Solid Solutions of Uranium Oxide. II Uranium Oxide-Yttrium Oxide, J. Phys. Chem. 70, 1090-7 (1966).

Table 12. Calculated^a Fluorite Lattice Constants of Rare Earth Sesquioxides

Rare Earth	a_o (Å)	Rare Earth	a_o (Å)
Y (b)	5.398	Gd	5.495
La	5.724	Tb	5.452
Ce (c)	5.686	Dy	5.436
Pr (c)	5.647	Ho	5.408
Nd	5.594	Er	5.381
Pm (c)	5.580	Tm	5.361
Sm	5.538	Yb	5.341
Eu	5.514	Lu	5.329

^a Calculated from the equation

$$a_o = 0.3943 a_c + 1.2285$$

where a_c is the lattice constant of the $D5_3$ sesquioxide.

^b Yttria is often treated as a rare earth, not only because of its size, but also because of its position in the periodic table.

^c The sesquioxide lattice parameter is an interpolated value.

equilibrated in 5 parts CO_2 to 1 part CO yield an extrapolated lattice parameter of 5.464 Å for " CeO_2 " versus a literature value of CeO_2 of 5.410 Å, equilibrated in air.⁹ The significance of our observation seems to be that a significant difference is obtained in lattice parameters of samples equilibrated at different oxygen potentials.

A number of experiments have been performed to try to determine the actual composition of cerium dioxide. The purpose is, of course, to establish a base to which weight change measurements can be related. Thus far, our measurements have not been conclusive. CeO_2 cannot be heated in air in either a Pt or an Al_2O_3 boat at 1000°C without reaction. Quartz, however, seems to be a satisfactory crucible material; further checks are under way.

⁹ Von F. Hund, R. Wagner, and U. Pertz, "Anomale Mischkristalle im System Cerdioxyde Uranoxyd," Z. Elektrochem. 56, 61-5 (1952).

Experiments are proceeding to coprecipitate U^{+6} and Ce^{+4} at 5, 10, 20, and 40 mole fraction. These samples are being heated at $400^{\circ}C$ in H_2 , using precalibrated frits. We hope to provide in this way definitive composition-lattice parameter-oxygen potential data. A similar study is planned for the U-Eu-O system. In this way we hope to fix these relationships for the entire U-RE-O, Pu-RE-O, and U,Pu-RE-O fission product systems.

2.2 Irradiation of Fuels

The final evaluation of $(U,Pu)O_2$ fuels will be based upon their irradiation performance. The irradiation testing program is concentrating on comparative tests of three fabrication forms: Sphere-Pac, pellets, and extrusions. The program includes thermal-flux irradiations that permit use of instrumented capsules and achievement of high burnup levels in relatively short times. These tests will provide supplemental information essential to the analysis of the fast-flux irradiation tests, in which the radial fission-rate distribution and cladding damage are more typical of anticipated LMFBR operating conditions. The test program also includes power transient tests to investigate fuel performance under abnormal operating conditions. The development of mathematical models to predict fuel behavior and of computer programs for analyzing fuel performance are integral parts of the irradiation test program.

2.2.1 Uninstrumented ETR Tests (A. R. Olsen, D. R. Cuneo, M. K. Preston)

The series of uninstrumented tests being irradiated in the X-basket facilities of the Engineering Test Reactor (ETR) since 1966 were originally started as screening tests of Sphere-Pac fuels. The objective of these tests has since been expanded to investigate the effects of fabrication form with extended burnup on fission-gas release, fission product migration, and fuel swelling. More recently these capsules have been used to provide short-cooled irradiated fuel for LMFBR fuel reprocessing studies.

The status of these tests is given in Table 13. At the present time only experiment 43-113 is under irradiation. This test is scheduled to be moved to a higher flux position during the next ETR shutdown, and Idaho Nuclear personnel are currently analyzing the flux monitors irradiated in the new position during ETR cycle 104-B. This flux measurement is being made to reduce the probability of overpowering this experiment, a problem which we have encountered with some of the previous experiments.

The metallographic examination of capsule 43-103 is approximately 90% complete, with alpha and beta-gamma autoradiographs still to be obtained. Two additional burnup samples from experiment 43-115 have been submitted for analysis, and the metallographic examination is approximately 60% complete and continuing. With the possible exception of the UO_2 fuel pin which shows restructuring essentially out to the

Table 13. Noninstrumented Thermal Flux Tests of (U,Pu)O₂ Fuels

Experiment Number	Fuel		Number of Rods	Peak Burnup (% FIMA) ^a	Peak Linear Heat Rate (w/cm)	Peak Cladding Inner Surface Temperature (C°)	Status September 1969
	Form	Composition					
43-99	Sphere-Pac	(²³⁵ U _{0.80} ,Pu _{0.20})O _{2.00}	2	1.5 ^b	1640 ^b	1000	Examined
43-100	Sphere-Pac	(²³⁵ U _{0.80} ,Pu _{0.20})O _{2.00}	2	1.4 ^b	1470 ^b	900	Examined
43-103	Sphere-Pac Pellet	UO _{2.02} (20% ²³⁵ U)	3	5	690	530	Being examined
		UO _{2.00} (20% ²³⁵ U)	1				
43-112	Sphere-Pac	(²³⁸ U _{0.85} ,Pu _{0.15})O _{1.97}	3	0.7	500	360	Being examined
		UO _{2.02} (20% ²³⁵ U)	1				
43-113	Sphere-Pac	(²³⁸ U _{0.85} ,Pu _{0.15})O _{1.97}	3	10 ^c	500 ^c	380 ^c	In-reactor, ≅ 8% FIMA
		UO _{2.02} (20% ²³⁵ U)	1				
43-115	Sphere-Pac	(²³⁸ U _{0.85} ,Pu _{0.15})O _{1.97}	3	5 ^c	600 ^c	460 ^c	Being examined
		UO _{2.02} (20% ²³⁵ U)	1				
43-116	Sphere-Pac	(²³⁸ U _{0.85} ,Pu _{0.15})O _{1.97}	4	1.5 ^c	600 ^c	460 ^c	Being examined
43-117 } 43-118 } 43-119 }	Ref. FTR Pellets	(²³⁸ U,Pu)O _{1.98}	4	1.5 ^d	600 ^c	460 ^c	In preparation

^aFIMA is fissions per initial actinide metal atom.^bRods failed in reactor from overpowering.^cThese are target design values.^dThis is an approximate level. Test will be irradiated for two ETR cycles to produce fuel for reprocessing studies.

fuel-cladding interface, the microstructures for these pins appear to be as expected. Final evaluation will be done when the metallographic examination is complete.

Fuel for capsules 43-117 through 43-119 is on order but has not yet been delivered. Capsule hardware machining and inspection is in progress.

2.2.2 Instrumented ETR Tests (C. F. Sanders)

Two instrumented ETR capsules are being constructed to investigate the swelling of Sphere-Pac fuels under conditions of high fission-gas retention.

During the past month most of our effort was concentrated on fabrication of the hardware, inspection, and development of the welding and brazing procedures. The checkout and installation of the instrumentation at Idaho Falls was completed, and flux monitors are being inserted to determine the unperturbed neutron flux. The fuel pins will be loaded within a week, and the assembly of the capsules should be completed by the second week of November.

2.2.3 ORR Instrumented Tests (R. B. Fitts, D. R. Cuneo, V. A. DeCarlo, K. R. Thoms)

The ORR instrumented tests are designed to study the thermal performance of experimental (U,Pu)O₂ fuel rods. The first series of tests is studying sol-gel-derived fuels.

The postirradiation examination of capsule SG-2 has continued with the sectioning of the upper fuel rod for metallography. This effort required impregnation of the fuel rod with epoxy after each cut in order to stabilize the Sphere-Pac fuel bed. The impregnation was apparently successful, and the first sample is awaiting metallographic preparation.

Fabrication of the fuel rods for capsule SG-3 was completed last month. The upper rod contains Sphere-Pac (U_{0.8},Pu_{0.2})O_{1.99} at a smear density of 82.1% of theoretical density around a W-25% Re, 1/8-in.-diam central thermocouple well. The lower rod contains (U_{0.8},Pu_{0.2})O_{1.99} sol-gel pellet fuel at a smear density of 82.2% of theoretical. The annular pellets average 83.5% dense with a 0.0025-in. diametral gap between them and the cladding. The central hole in these pellets is 1/8-in. diam, and the thermocouple well is 0.070-in. diam.

Fabrication of capsule SG-3 is continuing after encountering some difficulties. Two Zr-2 clad thermocouples were installed in the calorimeter. Unfortunately, one failed soon after final brazing into the calorimeter. A stainless steel-clad thermocouple was installed to replace it. The remaining good Zr-2 couple was joined to a stainless

steel leadout by means of a 1/16-in. to 1/16-in. Swagelok fitting. A 1/16-in. OD $\times$ 0.042-in. ID stainless steel tube was slipped over the 0.040-in. Zr-2 and stainless steel thermocouples. The thermocouple wires were coated with polyurethane, and the joint was filled with polyurethane prior to tightening of the swagelok. This fitting will be exposed to a neutron flux of 10^{10} neutrons/cm²-sec, and it is hoped that the polyurethane will survive. The test of this type of connector could be an important step in our thermocouple development program.

The central thermocouple in the lower fuel element was broken in two places after all gas tubes and thermocouples were brazed into the bulkhead. The thermocouple was drilled out, and since at this point in the assembly it was impossible to braze, a new one was silver soldered into the bulkhead. In the past the use of silver solder in a NaK environment has been avoided; however, it will probably be acceptable in this case.

Plans now call for the capsule to be inserted into the ORR during the December 7 shutdown.

2.2.4 TREAT Reactor Tests (C. M. Cox, E. J. Manthos)

The Series I TREAT experiments will compare the transient behavior of unirradiated sol-gel-derived Sphere-Pac and pelletized (U_{0.8},Pu_{0.2})O₂.

The first two TREAT capsules, TR-1 and TR-2, were shipped to the reactor on October 14, 1969. Testing of the two capsules should commence about October 27, 1969. Arrangements have been made to ship the irradiated capsules to Oak Ridge in the GE T-2 shipping cask. The cask will be loaned to us for the month of November.

A preliminary procedure for the disassembly and examination of the irradiated capsules has been submitted to the personnel involved in the examinations for their comments and approval. The first irradiated capsule should be returned to ORNL by the middle of November.

2.2.5 Fast Flux Irradiation Tests (A. R. Olsen)

The EBR-II tests are designed to approximate the conditions to be encountered in an operating LMFBR. The objective of these tests is to establish the effects of fuel fabrication form, Sphere-Pac versus pellet, and void distribution on fuel swelling, fuel-cladding mechanical interaction, fission-gas release, and fission product redistribution in a (U,Pu)O₂ fuel operating at typical heat rates to design burnup levels required for a power producing LMFBR.

The Series I encapsulated tests all contain Sphere-Pac (U,Pu)O₂ fuel. They have been exposed to approximately 6400 Mwd of EBR-II operation. This is equivalent to a calculated peak burnup of 3.4% FIMA or 85% of the currently scheduled irradiation. A request for Approval in Principle to extend the burnup for three of these tests from 5% FIMA to 10% FIMA was submitted to the AEC this month. Continued irradiation of these capsules will provide information on the effects of high burnup on Sphere-Pac fuels approximately 18 months sooner than the tests in Series II.

The Series II experiments involve unencapsulated fuel pin irradiation in a 37-pin subassembly to be shared with the Babcock and Wilcox Company. The proposed tests were described previously.¹⁰

Although Approval in Principle has not yet been received from the AEC, we have been informed that the tests will be approved. Consequently, the critical path schedule described in Section III, 1.2.2 of this report was developed.

¹⁰ A. R. Olsen, "Fast Flux Irradiation Tests," LMFBR Fuel Cycle Studies Progr. Rept. for September 1969, ORNL-TM-2748. (in press)

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