27
KAPL-1757

AEC Research and
Development Report

Knolls
Atomic Power
Laboratory

MASTER

The Fast Oxide Breeder— The Fuel Cycle

J.K. Davidson, W.O. Haas, Jr.

J.L. Mewherter, R.S. Miller, D.J. Smith

July 1, 1957

Operated for the
United States Atomic
Energy Commission by
GENERAL ELECTRIC

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

43-41-001

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

UNCLASSIFIED

KAPL-1757

UC-81, Reactors-Power
(TID-4500, 13th ed, Rev.)

THE FAST OXIDE BREEDER - THE FUEL CYCLE

J. K. Davidson W. O. Haas, Jr.
J. L. Mewherter R. S. Miller D. J. Smith

July 1, 1957

W. M. Cashin

Authorized Classifier

7/30/58

Date

General Electric Company
KNOLLS ATOMIC POWER LABORATORY
Schenectady, New York
Operated for the
United States Atomic Energy Commission
Contract No. W-31-109 Eng-52

UNCLASSIFIED

UNCLASSIFIED

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

- A. Makes any warranty or representation, express or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or
- B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission to the extent that such employee or contractor prepares, handles or distributes, or provides access to, any information pursuant to his employment or contract with the Commission.

Price \$2.25. Available from the Office of Technical Services, Department of Commerce, Washington 25, D. C.

CONFIDENTIAL

UNCLASSIFIED

KAPL-1757

KAPL-1757

UC-81, Reactors - Power
(TID-4500, 13th ed, Rev.)

Internal DistributionNo. of Copies

AEC, Schenectady Operations Office	1
Anderson, JD	1
Cashin, WM	10
Davidson, JK	1
Document Library	4
Haas, WO, Jr.	1
Longo, J. Jr.	1
Mewherter, JL	10
Miller, RS	1
Molino, DF	1
Sears, MJ	1
Smith, DJ	1

External Distribution

Aberdeen Proving Ground	3
AEC Scientific Representative, Argentina	1
AEC Scientific Representative, France	1
AEC Scientific Representative, Japan	1
Alco Products, Inc.	1
Argonne National Laboratory	10
Armed Forces Special Weapon Project, Washington	1
Armed Services Technical Information Agency	5
Army Ballistic Missile Agency	1
Army Chemical Center	2
Atomic Energy Commission, Washington	3
Atomics International	2
Babcock & Wilcox Company	4
Battelle Memorial Institute	2
Bettis Plant	4
Brookhaven National Laboratory	4
Brush Beryllium Company	1
Bureau of Medicine and Surgery	1
Bureau of Ships (Code 1500)	1
Canel Operations	3
Chicago Operations Office	1
Chicago Patent Group	1
Combustion Engineering, Inc.	1
Consolidated Vultee Aircraft Corporation	2
Convair-General Dynamics	1
Curtiss-Wright Corporation	1

PHOTOSTAT

KAPL-1757External Distribution (continued)No. of Copies

Defence Research Member	1
Department of Food Technology (MIT)	1
Department of Navy (Code 422)	2
Department of the Army, G-2	2
Division of Raw Materials, Denver	1
Dow Chemical Company (Rocky Flats)	1
Du Pont de Nemours and Company, Aiken	4
Du Pont de Nemours and Company, Wilmington	1
Evans Signal Laboratory	1
Frankford Arsenal	1
General Electric Company (ANPD)	2
General Electric Company, Richland	12
General Nuclear Engineering Corporation	1
Iowa State College	2
Jet Propulsion Laboratory	1
Kirtland Air Force Base	1
Lockheed Aircraft Corporation (Bauer)	2
Los Alamos Scientific Laboratory	4
Mallinckrodt Chemical Works	1
Maritime Administration	1
Massachusetts Institute of Technology (Dr. Hardy)	1
Mound Laboratory	1
National Advisory Committee for Aeronautics, Cleveland	1
National Bureau of Standards, Atomic Energy Project	1
National Bureau of Standards (Library)	1
National Lead Company, Inc., Winchester	1
Naval Research Laboratory	3
New York Operations Office	2
New York University (Dr. Richtmyer)	1
Nuclear Development Corporation of America	1
Nuclear Metals, Inc.	1
Oak Ridge Institute of Nuclear Studies	1
Oak Ridge National Laboratory	4
Office of Naval Research	10
Office of Ordnance Research	1
Ordnance Materials Research Office	1
Ordnance Tank-Automotive Command	1
Patent Branch, Washington	1
Phillips Petroleum Company	6
Picatinny Arsenal	1
Public Health Service	2

DO NOT PHOTOSTAT

KAPL-1757External Distribution (continued)No. of Copies

RAND Corporation	1
Sandia Corporation, Albuquerque	1
Sylvania Electric Products, Inc.	1
Tennessee Valley Authority	1
Texas Nuclear Corporation	1
The Martin Company	1
Union Carbide Nuclear Company (ORGDP)	2
U. S. Geological Survey, Denver	1
U. S. Naval Ordnance Laboratory	2
U. S. Naval Postgraduate School	1
U. S. Naval Radiological Defense Laboratory	1
U. S. Patent Office	1
University of California Radiation Laboratory, Berkeley	2
University of California Radiation Laboratory, Livermore	2
University of Puerto Rico	1
University of Rochester, Atomic Energy Project	1
University of Rochester (Dr. Marshak)	2
Vitro Engineering Division	1
Walter Reed Army Medical Center	1
Watertown Arsenal	1
Weil, Dr. George L.	1
Westinghouse Electric Corporation (Schafer)	2
Wright Air Development Center	6
Yankee Atomic Electric Company	1
Technical Information Service Extension, Oak Ridge	325
Office of Technical Services, Washington	<u>75</u>
Total	610

DO NOT PHOTOSTAT

ACKNOWLEDGMENT

The authors wish to acknowledge the following contributors to this development program:

Dr. W. M. Cashin for his aid in the fuel element fabrication development and irradiation testing.

F. Clark and A. Andrako for the laboratory extrusion experiments.

H. W. Krake for the laboratory solvent extraction experiments.

J. N. Frank for assistance in the extrusion process development.

A. C. Schafer for aid in preparing the cost estimate.

L. Stein and H. Weidberg for the precipitation and filtration studies.

Dr. H. W. Alter of the Atomic Power Equipment Department, General Electric Company, for his aid in the process chemistry development.

G. J. Hahn of the General Engineering Laboratory, General Electric Company, for the statistical experimental design and analysis.

C. Ladd of Atomic Power Development Associates for the helpful discussion on the fuel subassembly fabrication problem.

P. Onderchain and J. Russell of the Manufacturing Services Division, General Electric Company, for a cost analysis of the fuel element fabrication process.

Dr. H. G. Sowman of the Atomic Power Equipment Department, General Electric Company, for his helpful discussions on the ceramic fabrication problems.

KAPL-1757

CONTENTS

	<u>Page</u>
ABSTRACT	11
INTRODUCTION	13
PLANT AND PROCESS DESIGN AND COST ESTIMATE	14
Design Basis	14
Process and Operation	16
Flowsheet	16
Material Balance	23
Process Schedule	29
Process Equipment	30
Plant Layout	30
Equipment Description	30
Cost Estimate	40
PLANT AND FLOWSHEET DEMONSTRATIONS	49
Fuel Dejacketing and Dissolution	49
Fuel Shear	49
Fuel Dissolution	51
Nonirradiated Mixed Oxide Dissolution Studies	52
Irradiated Mixed Oxide Dissolution	53
Plant-Scale Dissolver Installation	55
Operation and Testing of the Pilot Plant	62
Solvent Extraction	62
Mixer-Settler Laboratory System	62
Flowsheet Considerations	63
Flowsheet Design	66
Equipment and Procedures	66
Run Results	67
Alternate Flowsheets	77
Mixed Oxide Preparation	77
Equipment	77
Experimental Procedure	80
Laboratory Experiments	83
Results and Conclusions	83

KAPL-1757

	<u>Page</u>
Fuel Element Fabrication	89
Equipment	89
Experimental Procedure	89
Results and Conclusions	91
REFERENCES	92
APPENDIX: Calculation of Normal Core and Blanket Processing Rates	93

ILLUSTRATIONS

<u>No.</u>	<u>Title</u>	<u>Page</u>
1	Fast Oxide Breeder Fuel Cycle	13
2	Section of a Typical Fast Oxide Breeder Fuel Element . . .	15
3	Process Flowsheet - Fuel Dejacketing and Dissolution . . .	17
4	Process Flowsheet - Solvent Extraction	18
5	Process Flowsheet - Mixed Oxide Preparation	18
6	Process Flowsheet - Fuel Element Fabrication	19
7	Process Flowsheet - Oxide Scrap Recovery	19
8	Process Flowsheet - Waste Disposal	20
9	Process Flowsheet - Solvent Recovery	20
10	Oxide Breeder Reprocessing Plant	31
11	Fuel Element Shear System	32
12	Dissolver System	34
13	Hot-Cell Equipment	35
14	Cell Rack	36
15	Remote Disconnect Flange	38
16	Extrusion System	39
17	Shear Feed Rollers	50

KAPL-1757

<u>No.</u>	<u>Title</u>	<u>Page</u>
18	Fuel Rod Shear	50
19	Equipment Rack	56
20	Rack Alignment and Locking Device	57
21	Remote Disconnect Flange	58
22	Disconnect Flange Assembly	59
23	Dissolving Pilot Plant - Side View	60
24	Dissolving Pilot Plant - End View	61
25	Mixer-Settler Feed System	64
26	Mixer-Settler Cascade	64
27	Solvent Extraction Product Receiver	65
28	Theoretical X-Y Diagrams for 1A Bank, FOB Flowsheet	68
29	Stage Profiles - Run FOB-102	68
30	Activity in Product Stream - Run FOB-105	73
31	Uranium Concentration - Run FOB-105	74
32	Total Alpha Activity - Run FOB-105	75
33	Material Balance - Run FOB-105	76
34	Fast Oxide Breeder Precipitator and Off-Gas System - Schematic Diagram	78
35	Precipitator and Assembly	79
36	Fast Oxide Breeder Precipitation and Filtration Equipment - Schematic Diagram	80
37	Filter Unit	81
38	Laboratory Precipitation Apparatus	84
39	Sintered Density vs Green Density - Plant-Scale Tests	88
40	Sintered Density vs Green Density - Laboratory Tests	88
41	Hydraulic Extrusion Equipment	90

7

7

7

7

7

ABSTRACT

The development of a fast breeder fuel cycle based on a core fuel consisting of mixed dioxides of uranium and plutonium is described. Process flowsheets, material balances, operating schedules, cost schedules, and a plant layout are presented, and the laboratory- and plant-scale process and equipment demonstrations are described. A cost estimate based on a demonstration of the process including the unique plant equipment gives a total fuel cycle cost of 2.04 mils/kw-hr and a net fuel cycle cost of 1.53 mils/kw-hr (0.3 breeding gain assumed).

4

4

4

4

THE FAST OXIDE BREEDER - THE FUEL CYCLE

J. K. Davidson, W. O. Haas, Jr.,
J. L. Mewherter, R. S. Miller, D. J. Smith

INTRODUCTION

A development program for investigating the feasibility of a fast reactor with mixed oxides as the fuel material has been pursued at the Knolls Atomic Power Laboratory for the past two years.¹ This report indicates the status of the development of a fuel cycle for the Fast Oxide Breeder (FOB). As shown schematically in Figure 1, the fuel cycle includes mechanical dejacketing and leaching of the discharged core fuel, fission-product decontamination by liquid-liquid solvent extraction, oxide preparation, and refabrication of the oxide to the required fuel element form plus off-site processing of the blanket material. The work conducted to date has resulted in a demonstration of the process, a number of unique equipment developments, and an economic evaluation of the fuel cycle.

FIGURE 1
FAST OXIDE BREEDER FUEL CYCLE

PLANT AND PROCESS DESIGN AND COST ESTIMATE

Design Basis

One of the basic objectives in the development of a fuel cycle for the FOB project has been the design of a reprocessing scheme that will make use of the economic advantages of oxide fuel elements. These important advantages are: (1) fewer reprocessing steps (elimination of metal reduction), (2) relatively simple fabrication, and (3) a fuel element capable of extremely high burnup, which will result in a low processing rate. One can also add the advantage of being able to leach the oxide from the cladding material, since this will permit a reduction in size and complexity of the plant equipment through the elimination of high concentrations of cladding inerts in the fuel stream.

In the plant required for reprocessing the core material, the equipment in high level radiation areas can be small, relatively inexpensive, and expendable. Consequently, a "throw-away" maintenance concept can be substituted for the more conventional, and expensive, direct or remote maintenance methods.

The FOB mixed oxide fuel element is, in concept, a stainless steel tube 42 in. long with a 0.236-in. OD and a 0.206-in. ID filled with a solid solution of uranium and plutonium dioxide in an atomic ratio of about 5 to 1. The oxide is loaded at a bulk density equal to or possibly greater than 65% of the theoretical density. The core of the element consists of seven 0.200-in.-diam rods of the oxide, each 6 in. long. These fuel rods are formed by extrusion and then sintered. No thermal bond is used between the core and cladding, and a 0.003-in. maximum radial clearance between the core and cladding is specified. After loading, the tube is purged with helium, and leak-tight stainless steel plugs are welded to each end of the assembly. A view of a typical sectioned element is shown in Figure 2. Quality control requirements for this element specify no more than $\pm 1\%$ variation in fuel length, density, and loading per unit length.

The amount of material to be processed is based on the calculated time (~ 1 yr) to accomplish the desired 50% depletion of the plutonium originally charged. The gross oxide requirement shown in Table 1 applies only to the oxide preparation and fabrication section of the plant, since the requirement was obtained by assuming for this section 50% scrap oxide, which must be recycled. The capacity of the chemical processing section of the plant was calculated from the estimated core fuel discharge rate and the required fuel make-up rate.

FIGURE 2

SECTION OF A TYPICAL FAST OXIDE
BREEDER FUEL ELEMENT

KS-5545
Unclassified

TABLE 1. Process Capacity

Number of acceptable elements per day	80
Number of 6-in.-long units in eight 42-in.-long elements	560
Oxide required for 80 elements	13.1 kg
Gross powder required - 50% yield	26.2 kg
Number of 6-in.-long units in 160 elements	1120
Operation basis	2 shifts, 5-day week

Process and Operation

A process flowsheet involving the following steps has been developed. The fuel rods are sheared into sections, and the contents are dissolved in nitric acid. The nitrate solution is used as feed in a Purex-type solvent extraction system without partition. Plutonium and uranyl nitrates are added to the aqueous product, which is precipitated with ammonia and then filtered. The precipitate is converted to the mixed oxide in a hydrogen atmosphere at 700°C. Water and organic binder are blended with the oxide to form a clay, which is extruded in rod form and dried. The rods are sintered in a hydrogen atmosphere to a predetermined density, and the fired rods are loaded into stainless steel cladding tubes, which are then welded shut. The elements are then fabricated into subassemblies for reloading into the reactor. The first three process steps require heavy shielding because of fission-product activity, while the operations following fission-product decontamination require intermediate shielding because U^{237} and other radioactive transuranic elements build up as the fuel is recycled.

Flowsheet

The detailed flowsheet for the fast oxide breeder fuel cycle is presented in Figures 3 through 9. The fundamental chemical reactions are given in Table 2.

Fuel De-jacketing and Dissolution. Before the spent fuel is delivered to the reprocessing plant, it is allowed to cool in a sodium tank for 30 days. Following sodium removal and disassembly of the bundle, the single fuel rods are sheared to 1/2-in. lengths. This operation exposes the fuel surface for subsequent processing. The cut sections are charged to a dissolving vessel where 60% nitric acid is used to leach the fuel core material. The extremely radioactive stainless steel rings are retained in a basket within the dissolver for later removal and underground burial in a radioactive waste disposal area.

Solvent Extraction. The dissolved spent fuel is then partially decontaminated from fission products by a Purex-type liquid-liquid extraction process. The flowsheet specifications are given in Table 3. This process separates the uranium and plutonium from the major fraction of fission products. Additional uranium and plutonium are added after this processing to achieve the required fuel atomic ratio.

Mixed Oxide Preparation. Precipitation from the solvent extraction product is accomplished by bubbling ammonia gas through the solution. A rotary vacuum filter is used to separate the uranium and plutonium precipitate from the mother liquor. The dried cake is calcined in a hydrogen atmosphere at 700°C to reduce the precipitate to the solid solution of uranium and plutonium dioxides.

Fuel Element Fabrication. The coarse, friable powder of mixed oxide is broken to <20-mesh particle size and then blended with a binder of 2 wt % methyl cellulose and 10 wt % water to form a homogeneous clay. The clay is extruded in the form of rods onto trays and dried. The drying operation requires 16 hr at 70°F in an atmosphere of 75% relative humidity. The dried rods are cut into 6-in. lengths, and the pieces of acceptable quality are loaded into a cast alumina holder for sintering.

FIGURE 3
PROCESS FLOWSHEET
FUEL DEJACKETING AND DISSOLUTION

FIGURE 4
PROCESS FLOWSHEET SOLVENT EXTRACTION

FIGURE 5
PROCESS FLOWSHEET MIXED OXIDE PREPARATION

FIGURE 6
PROCESS FLOWSHEET
FUEL ELEMENT FABRICATION

FIGURE 7
PROCESS FLOWSHEET oxide scrap recovery

FIGURE 8
PROCESS FLOW SHEET
WASTE DISPOSAL

FIGURE 9
PROCESS FLOWSHEET
SOLVENT RECOVERY

TABLE 2. Chemical Reactions of the Fast Oxide Breeder Fuel Cycle

Fuel Dissolution

or

Precipitation Supernate Treatment Precipitate Decomposition

TABLE 3. Solvent Extraction Conditions at 25°C

<u>A Bank</u>	<u>Nominal</u>
1AF (Aqueous Feed)	
Relative flow rate	100
$\text{UO}_2(\text{NO}_3)_2$ molarity	1.05
$\text{Pu}(\text{NO}_3)_4$ molarity	0.16
HNO_3 molarity	2.00
1AS (Aqueous Scrub)	
Relative flow rate	66.7
HNO_3 molarity	3.00
1AX (Organic Extractant)	
Relative flow rate	333.3
Tributyl phosphate, vol %	30.0
<u>C Bank</u>	
1CX (Aqueous Strip)	
Relative flow rate	500.0
HNO_3 molarity	0.02

These fuel units are sintered for 1 hr at 1700°C. Each sintered unit is checked for tolerance specifications and, if acceptable, is loaded into a precut length of Type 347 stainless steel cladding. The tube is purged with helium, and the end plugs are welded in place. Each completed element is inspected for completeness of seal and X-rayed to check the fuel loading and welds. Acceptable elements are assembled into bundles for insertion into the reactor.

Scrap Oxide Recycle. The anticipated 50% rejected oxide in the fabrication of the rods will be recycled to the process. Separate dissolving facilities are required to avoid recontamination of the already decontaminated fuel material. The recycle stream is introduced into the process just before the precipitation step.

Waste Disposal. The fission-product waste from the solvent extraction process is neutralized, concentrated to 30 wt % solids, and stored in cooled underground storage tanks. The ammonium nitrate from the precipitation step is converted to sodium nitrate by the addition of sodium hydroxide and, after concentration to 30 wt % solids, is placed in drums for shipment off site. Other liquid wastes are either combined with these major solutions or directly discharged after analysis.

Solvent Recovery. The contaminated solvent from the extraction step is recovered for reuse. The procedure for washing the solvent is given in Table 4. It is necessary to provide for 2.5% solvent loss per cycle because of the solubility of the solvent in the aqueous streams and the difficulty in separating phases during washing.

TABLE 4. Solvent Wash Procedure

	<u>Time, min</u>
Agitation with 0.1 vol % water	15
Standing (then discard wash)	15
Agitation with 0.1 vol % 0.1-M NaOH	60
Standing (then discard wash)	15
Agitation with 0.1 vol % water	15
Standing (then discard wash)	15
Agitation with 0.1 vol % 0.05-M HNO ₃	15
Standing (then discard wash)	15
Agitation with 0.1 vol % water	15
Standing (then discard wash)	15

Material Balance

Steady-state operation of the FOB reactor is achieved by an incremental refueling scheme whereby at any particular time the reactor core will consist of bundles of fuel elements varying from zero to maximum burnup. These bundles are distributed in such a way that replacement of a fully burned-up bundle by a new bundle will not greatly alter the nuclear characteristics of the reactor.²

The normal plant processing rates are given in Table 5. The detailed calculation of these rates is presented in the Appendix. The design basis for the flowsheet material balance was obtained from these rates by including a safety factor for continuous reactor operation, maintenance shutdowns of the reprocessing plant, and possible lower burnup feed material. The material balance for the processing plant is given in Tables 6 through 12 and is based on a daily feed rate of 10.1 kg of UO_2 and 1.7 kg of PuO_2 .

TABLE 5. Normal Core and Blanket Processing Rates

Basis:

50% burnup of the plutonium originally charged to the core
 Internal breeding ratio of 0.5
 External breeding ratio of 0.9
 Blanket material removed at 5000 gm Pu/ton U
 Plant operation for 250 days/year
 Reactor operation for 300 days/year

<u>Core Fuel</u>	<u>kg/day</u>
PuO_2	1.36
UO_2	7.88
Fission-product oxides	0.90
Stainless steel	2.24
 <u>Blanket Fuel</u>	
Uranium	171
Plutonium	0.855
Stainless steel	11.45

TABLE 6. Material Balance - Fuel De jacketing and Dissolution

Basis: 10.1 kg UO_2 and 1.7 kg PuO_2

Flowsheet Streams*	UO_2 , kg	PuO_2 , kg	$\text{UO}_2(\text{NO}_3)_2$, kg	$\text{Pu}(\text{NO}_3)_4$, kg	HNO_3 , kg	NO_2 , kg	NaOH , kg	H_2O , kg	Stainless Steel, kg	Total Weight, kg	Specific Gravity	Total Volume, liters
101 (sheared elements)	10.1	1.7	-	-	-	-	-	-	2.88	14.7	-	-
102 (cladding)**	-	-	-	-	-	-	-	-	2.88	2.88	-	-
103 (acid)	-	-	-	-	15.6	-	-	9.4	-	25.0	1.378	18.1
104 (dilution)	-	-	-	-	-	-	-	15.9	-	15.9	1.000	15.9
105 (IAF)	-	-	14.74	3.05	4.35	-	-	25.35	-	47.4	1.3941	34.0
106 (off-gas)	-	-	-	-	-	3.44	-	-	-	3.44	-	-
107 (scrub H_2O)	-	-	-	-	-	-	-	7.08	-	7.08	1.000	7.08
108 (scrub waste)**	-	-	-	-	-	4.7	-	7.08	-	11.78	1.2463	9.46
109 (scrub caustic)**	-	-	-	-	-	-	1.0	10.0	-	11.0	1.1089	9.92

*The numbers refer to the flowsheet streams
in Figures 3 through 9.

**Waste stream.

TABLE 7. Material Balance - Solvent Extraction

Basis: 10.1 kg UO₂ and 1.7 kg PuO₂

Flowsheet Streams*	Relative Flow	UO ₂ (NO ₃) ₂ , M	Pu(NO ₃) ₄ , M	HNO ₃ , M	TBP in Kerosene, vol %	UO ₂ (NO ₃) ₂ , kg	Pu(NO ₃) ₄ , kg	HNO ₃ , kg	TBP, kg	H ₂ O, kg	Total Weight, kg	Specific Gravity	Total Volume, liters
201 (LAF)	100	1.10	0.16	2.00	-	14.74	3.05	4.35	-	28.42	50.56	1.487	34.0
202 (LAX)	333.3	-	-	-	30	-	-	-	32.85	-	93.91	0.837	113.3
203 (LAS)	66.7	-	-	3.00	-	-	-	4.29	-	20.6	24.9	1.097	22.7
204 (LAW)**	166.7	-	-	2.05	-	-	-	7.32	-	53.1	60.4	1.066	56.7
205 (LAP)	340	0.320	0.054	0.180	30	14.74	3.05	1.32	32.85	-	113.02	0.966	117.0
206 (LCX)	500	-	-	0.02	-	-	-	0.21	-	170.0	170.0	1.001	170.0
207 (LCP)	510	0.215	0.036	0.14	-	14.74	3.05	1.53	-	169.7	189.0	1.088	173.7
208 (LCW)	333.3	-	-	-	30	-	-	-	32.85	-	93.91	0.837	113.3

*See Figures 3 through 9.

**Waste stream.

TABLE 8. Material Balance - Mixed Oxide Preparation

Basis: 10.1 kg UO₂ and 1.7 kg PuO₂

Flowsheet Streams*	UO ₂ (NO ₃) ₂ , kg	(NH ₄) ₂ U ₂ O ₇ , kg	UO ₂ , kg	Pu(NO ₃) ₄ , kg	Pu(OH) ₄ , kg	PuO ₂ , kg	HNO ₃ , kg	NH ₃ , kg	NH ₄ NO ₃ , kg	H ₂ , kg	H ₂ O, kg	Total Weight, kg	Specific Gravity	Total Volume, liters
301 (LCP)	14.74	-	-	3.05	-	-	1.53	-	-	-	169.7	189.0	1.088	173.7
302 (scrap oxide recycle)	15.90	-	-	3.95	-	-	2.83	-	-	-	175.4	198.1	1.108	178.7
303 (make-up)	1.20	-	-	0.90	-	-	1.30	-	-	-	5.7	9.1	1.804	5
304 (NH ₃)	-	-	-	-	-	-	-	8.4	-	-	-	8.4	-	-
305 (off-gas)	-	-	-	-	-	-	-	1.6	-	-	-	1.6	-	-
306 (H ₂ O scrub)	-	-	-	-	-	-	-	-	-	-	3.76	3.76	1.000	3.76
307 (NH ₄ OH scrub waste)**	-	-	-	-	-	-	-	1.6	-	-	3.76	5.36	0.892	6.0
308 (slurry)	-	25.2	-	-	5.0	-	-	-	25.4	-	350.8	406.4	-	-
309 (filter wash)	-	-	-	-	-	-	-	-	-	-	40.0	40.0	1.000	40.0
310 (filtrate)	-	-	-	-	-	-	-	-	25.4	-	390.8	416.2	1.023	401.2
311 (precipitate)	-	25.2	-	-	5.0	-	-	-	-	-	-	30.2	-	-
312 (H ₂)	-	-	-	-	-	-	-	-	-	0.2	-	0.2	-	-
313 (H ₂ off-gas)	-	-	-	-	-	-	-	1.4	-	-	2.8	4.2	-	-
314 (oxide)	-	-	21.8	-	-	4.4	-	-	-	-	-	26.2	-	-

*See Figures 3 through 9.

**Waste streams.

TABLE 9. Material Balance - Fuel Element Fabrication

Basis: 10.1 kg UO₂ and 1.7 kg PuO₂

Flowsheet Streams*	UO ₂ , kg	PuO ₂ , kg	Methyl Cellulose, kg	H ₂ O, kg	H ₂ , kg	Stainless Steel, kg	Total Weight, kg
401 (oxide)	21.8	4.4	-	-	-	-	26.2
402 (binder)	-	-	0.26	-	-	-	0.26
403 (H ₂ O)	-	-	-	5.24	-	-	5.24
404 (clay)	21.8	4.4	0.26	5.24	-	-	31.70
405 (water vapor)	-	-	-	5.24	-	-	5.24
406 (green rod)	21.8	4.4	0.26	-	-	-	26.46
407 (H ₂)	-	-	-	-	0.1	-	0.1
408 (H ₂ off-gas)	-	-	0.26	-	0.1	-	0.36
409 (scrap oxide recycle)	10.9	2.2	-	-	-	-	13.1
410 (sintered rod)	10.9	2.2	-	-	-	-	13.1
411 (cladding)	-	-	-	-	-	2.88	2.88
412 (fuel rod)	10.9	2.2	-	-	-	2.88	16.0

*See Figures 3 through 9.

TABLE 10. Material Balance - Oxide Scrap Recovery

Basis: 10.1 kg UO₂ and 1.7 kg PuO₂

Flowsheet Streams*	UO ₂ , kg	PuO ₂ , kg	UO ₂ (NO ₃) ₂ , kg	Pu(NO ₃) ₄ , kg	HNO ₃ , kg	NO ₂ , kg	H ₂ O, kg	Total Weight, kg	Specific Gravity	Total Volume, liters
501 (oxide)	10.9	2.2	-	-	-	-	-	13.1	-	-
502 (acid)	-	-	-	-	16.3	-	9.9	26.2	1.378	19.0
503 (dilution)	-	-	-	-	-	-	165.5	165.5	1.000	165.5
504 (feed)	-	-	15.90	3.95	2.83	-	175.4	198.08	1.1084	178.7
505 (off-gas)	-	-	-	-	-	3.50	-	3.50	-	-
506 (scrub H ₂ O)	-	-	-	-	-	-	7.10	7.10	1.000	7.10
507 (scrub waste)	-	-	-	-	4.72	-	7.10	11.82	1.246	9.49

*See Figures 3 through 9.

TABLE 11. Material Balance - Waste Disposal

Basis: 10.1 kg UO₂ and 1.7 kg PuO₂

Flowsheet Streams*	Stainless Steel, kg	HNO ₃ , kg	NaOH, kg	NH ₃ , kg	NH ₄ NO ₃ , kg	H ₂ O, kg	NaNO ₃ , kg	Total Weight, kg	Specific Gravity	Total Volume, liters
601 (cladding)	2.88	-	-	-	-	-	-	2.88	-	-
602 (1AW)	-	7.32	-	-	-	53.1	-	60.4	1.066	56.7
603 (dissolver wash)	-	-	-	-	-	40.0	-	40.0	1.000	40.0
604 (feed make- up condensate)	-	-	-	-	-	-	-	-	-	-
605 (filtrate waste)	-	-	-	-	25.4	390.8	-	416.2	1.023	401.2
606 (caustic sol- vent wash)	-	-	0.06	-	-	14	-	14	1.0	14
607 (caustic)	-	-	10.6	-	-	-	-	10.6	-	-
608 (acid scrub)	-	4.70	-	-	-	7.08	-	11.78	1.2463	9.46
609 (acid scrub)	-	4.72	-	-	-	7.10	-	11.82	1.246	9.49
610 (HNO ₃ solvent wash)	-	0.04	-	-	-	14	-	14	1.0	14
611 (bottoms low level neutralizer)	-	-	-	-	-	63.0	27.0	90.0	-	-
612 (bottoms high level neutralizer)	-	-	-	-	-	52.50	22.50	75.00	1.336	56.1
613 (condensate high level neutralizer)	-	-	-	-	-	73.6	-	73.6	1.000	73.6
615 (condensate low level neutralizer)	-	-	-	-	-	407.1	-	407.1	1.000	407.1
617 (caustic scrub)	-	-	1.0	-	-	10.0	-	11.0	1.1089	9.92
618 (NH ₃ scrub)	-	-	-	1.6	-	3.76	-	5.36	0.892	6.0
619 (water solvent wash)	-	-	-	-	-	42	-	42	1.000	42
621 (waste to direct discharge)	-	-	1.0	1.6	-	477	-	480	1.001	480

*See Figures 3 through 9.

TABLE 12. Material Balance - Solvent Recovery

Basis: 10.1 kg UO_2 and 1.7 kg PuO_2

Flowsheet Streams*	HNO_3 , kg	NaOH , kg	TBP, kg	Kerosene, kg	H_2O , kg	Total Weight, kg	Specific Gravity	Total Volume, liters
701 (solvent addition)	-	-	8.21	15.26	-	23.47	0.837	28.04
702 (used sol- vent - 1CW)	-	-	32.85	61.06	-	93.91	0.837	113.3
703 (H_2O wash)	-	-	-	-	14	14	1.000	14
704 (NaOH wash)	-	0.06	-	-	14	-	1.0	14
705 (H_2O wash)	-	-	-	-	14	14	1.000	14
706 (HNO_3 wash)	0.04	-	-	-	14	14	1.0	14
707 (H_2O wash)	-	-	-	-	14	14	1.000	14
708 (NaOH waste)	-	0.06	-	-	14	14	1.0	14
709 (HNO_3 waste)	0.04	-	-	-	14	14	1.0	14
710 (H_2O waste)	-	-	-	-	42	42	1.000	42
711 (washed solvent)	-	-	32.85	61.06	-	93.91	0.837	113.3

*See Figures 3 through 9.

Process Schedule

The plant is designed to operate on a two-shift, 40-hr-week schedule except for the solvent extraction equipment, which requires three-shift, 40-hr-week operation. The first two shifts require sixteen operators and one shift engineer, while the third shift requires only two operators to maintain continuous operation of the solvent extraction equipment. Details of operation are as follows.

Fuel Dejacketing and Dissolution - two men. The first shift has responsibility for the sectioning and dissolution of one day's output of fuel from the reactor; that is, one dissolver batch. The second shift is responsible for the dissolution of the recycled oxide from the fuel refabrication operation.

Solvent Extraction - one man, except two men on third shift. One man operates the solvent extraction equipment on a continuous basis and, in addition, operates the solvent recycle equipment through one cycle of batch washes each day. An extra man on the third shift is provided for safety reasons and is required to operate the waste handling equipment.

Oxide Preparation - two men. Two men operate the three precipitators and the rotary drum filter. It is necessary to put one batch through each precipitator during each of the two shifts in a staggered manner. The over-all time schedule of each precipitator involves 6 hr, 2 hr of which are for filtration. The precipitation schedule is such that the filter operates continuously on both shifts. The cake from the filter is split into 500-gm batches to ensure that subsequent operations are critically safe. Every 20 min a 500-gm batch is fed to the furnace for firing.

Blending - one man. One man operates the six blenders. As the batches are received from the furnace at 20-min intervals, they are placed in a blender and the cycle is started. Each blender has a time cycle of 1.5 hr, after which the completed batch is transferred to the extruder.

Extrusion and Drying - two men. The extruder operates on a 20-min cycle per batch, so that it handles the material from the blender without holdup. Each cycle results in one tray of fuel, which is inserted into the dryer where it is left for 16 hr.

Sintering and Inspection - two men. After drying, the rods are broken into 6-in. sections and inserted into an alumina boat which holds 30 sections. One boat is inserted into the sintering furnace every 20 min. The rods are inspected at this point, and those not meeting specifications are recycled.

Loading, Welding, and Inspection - three men. Three men insert the rods into the cladding, purge the assembly with helium, weld the caps, and test the completed element for leaks. They are expected to turn out one element every 10 min.

Material Handling - three men. The additional three men per shift assist in handling the material in the plant and allow for vacations and sickness.

Process Equipment

Plant Layout

The building details for the FOB fuel cycle plant are shown in Figure 10. The building is 91 by 65 ft and contains 10,830 ft² of floor space. It has structural steel framing, corrugated transite walls, and a steel deck roof. The process area consists of both a hot and a warm cell, which are surrounded by an operating area, cold feed area, and offices. A sampling gallery above the cells connects directly to an analytical laboratory located on the second floor.

A 5-ton jib crane is provided for handling the fuel cask and cladding disposal casks on the top of the hot cell. The warm cell is serviced throughout its entire length by a 2-ton traveling crane and a manipulator similar to a General Mills type and, in addition, has a pair of servo-manipulators at each of nine windows. The hot-cell equipment is mounted on traveling racks coupled to the wall by remote connectors. The rack and coupling details are discussed in the section on Plant and Flowsheet Demonstrations. These racks are partially decontaminated, removed, and buried in the event that equipment failure occurs.

The shielding on the hot cell is 3.5 ft thick, on the warm cell it is 2 ft thick, and in both cells it is of 4.5 density concrete. This reduces the radiation levels in the operating areas to < 6 mr/hr.

To protect the operating area from plutonium contamination, the cells are lined with stainless steel and sealed during use. The hot-cell operating pressure is lower than that of the warm cell, which in turn, is lower than the pressure in the operating area, so that all air flow is from less contaminated to more contaminated areas. The ventilation provides one air change per minute to the cell area and twenty air changes per hour to the operating area.

Equipment Description

Fuel Rod Shear. A design for the fuel rod shear is shown in Figure 11. In this design the shear consists of a magazine containing fixed and rotating die pieces and 33 full-length tubes, which are loaded at the reactor canyon with fuel elements disassembled from the fuel bundles. The magazine

OXIDE BREEDER
REPROCESSING
PLANT
FIGURE 10

KS 16992
UNCLASSIFIED

KS-16605
UNCLASSIFIED

FUEL ELEMENT SHEAR SYSTEM

FIGURE 11

is loaded from a transfer cask into the ceiling of the hot cell, where it engages a motor drive unit. A shaft extending through the center of the magazine turns the rotating shear blade at the lower end of the magazine. The shear cuts one 1/2-in. section off each fuel rod in one complete revolution of the shear blade.

Dissolver. The sheared sections fall by gravity into a critically safe dissolver provided with a basket for removing the leached cladding sections. This arrangement is shown in Figure 12. Provision for easy removal of the dissolver basket is made by constructing the dissolver from two vertical 8-ft sections of standard 6-in. pipe located on 2-ft centers. These two legs are interconnected by horizontal sections of 1-1/2-in. pipe at the bottom, middle, and a point above the operating liquid level. A down-draft condenser is located in the top portion of one vertical leg; the other leg is left clear for removal and replacement of the basket. A cladding basket disposal cask, constructed so that it can be lowered over the leg containing the basket, is used also to seal the open top of the dissolver. The sectioned fuel is charged to the dissolver through a vertical tube entering the dissolver leg at a point just above the top of the basket.

Mixer Settlers. The contactors used in the solvent extraction process are pump-mix mixer settlers^{11,12} constructed of stainless steel. The impellers are driven through a gear system by a dc motor mounted on the top of each bank. The "A" bank has 16 stages, each 3 in. high, 15 in. wide, and 4.1 in. long including a 1.5-in. mixing section. The "C" bank has 12 stages, each 3 in. high, 1.5 in. wide, and 7.3 in. long including a 1.5-in. mixing section. The mixer settlers are shown in Figure 13 above the receiver tanks on the traveling rack. This figure also illustrates the position of the remote connectors on the rack and the location of the remote head diaphragm pump used to pump the feed solution to the mixer settlers.

Disposable Equipment Rack. The traveling racks containing the dissolving, solvent extraction, and waste handling equipment are constructed from 1-in. angle iron. A typical rack is shown in Figure 14 before equipment installation. It is 10 ft high, 10 ft long, and 27 in. wide. Four wheels located at the top corners of the rack travel on overhead rails to allow rack removal. A locking rod and alignment pins are mounted at the top front of the rack to ensure correct positioning of the rack with respect to the fixed block of the remote disconnect flange.

DISSOLVER SYSTEM

FIGURE 12

FIGURE 13. HOT-CELL EQUIPMENT

FIGURE 14 CELL RACK

KS-20272
UNCLASSIFIED

Remote Disconnect Flange. The remote disconnect flange which has been developed for connecting process lines through the cell wall to equipment located on the disposal rack has a number of standard Crawford Swagelok fittings compressed between two stainless steel mounting blocks by a clamping bolt. The flange assembly, which can be operated remotely from the outer cell face, replaces the regular fitting nuts of the individual connectors. This method of construction provides a gasketless, leakproof connection entirely of stainless steel construction, a feature which is particularly advantageous in handling corrosive radioactive solutions. As many as eight 3/8-in. lines can be coupled by a single unit. A typical unit is shown in Figure 15.

Rotary Drum Filter. The filter is a standard laboratory unit with a drum filter area of 4 ft². The material is removed from the surface of the stainless steel filter screen by a knife blade.

Oxide Conversion Furnace. The conversion furnace is a standard electric, hydrogen atmosphere unit with a 6-in.-diam muffle about 48 in. long. The 24-in. hot zone must be capable of operating continuously at 1800°C. The boats for use in the furnace can be of stainless steel, 4 in. in diameter by 8 in. long with a 2-in. longitudinal slot for charging and removing the oxide.

Blenders. The Hobart 2-liter mixer is satisfactory for blending. The paddle is spring-loaded so that it will press against the bottom of the bowl to produce a grinding as well as a blending action.

Extruder. A design for a remote extruder is shown in Figure 16. The oxide clay is pressed from the extruder barrel through the die onto the take-up table by the hydraulic ram. The table is synchronized with the ram to receive the rod in the tray grooves. Automatic drive motors move the table both back and forth and sideways to receive the extruded rod. Provision is made to clean the die, barrel, and ram remotely after each extrusion by rotating the assembly into a cleaning position.

Drying Cabinet. The drying cabinet, an enclosure 3 ft wide, 6 ft high, and 24 ft long, is maintained at 75°C and 75% relative humidity. At one end of the cabinet, trays holding the extruded rod are placed on chain-driven shelves, which are moved through the dryer in a period of 16 hr. The dryer is designed so that each batch will be contained in an area 1.5 by 1.5 by 3 ft for criticality control.

Sintering Furnace. For sintering, use is made of a standard electric furnace with molybdenum wire windings on an alumina tube muffle. A protective atmosphere of hydrogen is required to prevent oxidation of both the oxide fuel and the molybdenum windings during firing. The muffle is 6 in. in diameter by 60 in. long with a 24-in. hot zone. The furnace is capable of operating at a temperature of 1800°C.

FIGURE 15. REMOTE DISCONNECT FLANGE

KS-8644
Unclassified

FIGURE 16

Other Equipment. The equipment not discussed is standard in chemical plants, and details other than the required size have not been determined.

Cost Estimate.

The cost estimate has been based on the Lang factors, which are commonly used in the process industries.⁵ These are of sufficient accuracy for preliminary evaluation and depend on the best-known information about the plant: namely, the delivered cost of the major equipment required for the recycle process proper. The factors depend on the general type of operation involved (solids or liquids) and the scope of the installation (new plant or plant addition). The original factors have been adjusted from experience and recent data to correspond to a radiochemical plant capable of handling part liquids and part solids and constructed as a major plant addition at an already developed reactor site. The factors used in the evaluation are given in Table 13 for the plant investment and in Table 14 for the annual operating cost. Most of the costs for the delivered process equipment were determined from the correlation of C. C. Chilton⁶ by application of a cost index ratio factor. However, the costs of equipment actually constructed at KAPL were used wherever possible. Details of the building cost are presented in Table 15, of equipment costs in Tables 16 through 22 of equipment spares in Table 23, and of direct labor charges in Table 24. The total plant investment is given in Table 25. The annual cost of operation is calculated as shown in Table 26. By adding to the operating cost the inventory charges on the uranium and plutonium, the expense of blanket processing, and an allowance of \$400,000 for subassembly fabrication the total cost of operation is found to be \$3,536,030 per year.

When this figure is divided by the plant power output of 1.73×10^9 kw-hr/yr, a cost for the fuel cycle of 2.04 mils/kw-hr is obtained. These data are summarized in Table 27. If the possible credit allowance is made for the additional plutonium recovered as a result of breeding, the cost of power is 1.53 mils/kw-hr (assumed breeding gain of 0.3).

TABLE 13. Plant Investment Cost Factors

Major process equipment, delivered	A	
Major process equipment, installed	1.4A	1.4A
Instruments, delivered	B	
Instruments, installed	1.65B	1.65B
Process piping	0.6A	0.6A
Nonmanufacturing capital investments		
Building estimate	C	
Auxiliary estimate	A	
Cost of delivered spares	D	
	<u>(C + A + D)</u>	<u>(C + A + D)</u>
Total physical cost		3.0A + 1.65B + C + D
Plant construction overhead (including design, construction overhead, profit, and contingencies)		<u>0.4(3.0A + 1.65B + C + D)</u>
Total plant investment before startup		1.4(3.0A + 1.65B + C + D)
Startup		<u>0.14(3.0A + 1.65B + C + D)</u>
Total Plant Investment	E =	1.54(3.0A + 1.65B + C + D)

TABLE 14. Annual Operating Cost Factors

Raw materials	F	F
Direct labor	G	G
Indirect manufacturing expenses		
Process utilities	H	
Maintenance	$I = 0.06[E - 1.4(C + D)] + 0.01(1.4C)$	
Supplies	$J = 0.005[E - 1.4(C + D)] + 0.001(1.4C)$	
Supervision and clerical	0.25G	
Property taxes and insurance	0.03E	
Laboratory	0.25G	
Employee benefits and continuity of service	$0.15(1.25G) + 0.075I$	
Miscellaneous plant overhead	<u>$0.60(1.25G + I + J)$</u>	
Sum of indirect manufacturing expenses	= K	K
Depreciation	0.10E	<u>0.10E</u>
Shop cost		$(F + G + K + 0.10E)$
Manufacturing cost addend	$0.05(F + G + K + 0.10E)$	<u>$0.05(F + G + K + 0.10E)$</u>
Manufacturing cost		$1.05(F + G + K + 0.10E)$
Waste storage	L	L
Profit before taxes	$M = 0.20[0.5E + (1/12)N]$	<u>M</u>
Annual Operating Cost	N =	$1.05(F + G + K + 0.10E) + L + M$

TABLE 15. Building Cost

Building, cells, and stack	\$590,830	\$ 590,830
Manipulators, delivered	180,000	
Manipulators, installed	252,000	<u>252,000</u>
Building estimate		\$ 842,830
Plant construction overhead (design, construction overhead, profit, contingencies)		<u>337,130</u>
Total Building Cost		\$1,179,960

TABLE 16. Fuel Dejacketing and Dissolution - Costs*

<u>Equipment</u>		
<u>Number</u>	<u>Description</u>	<u>Cost</u>
1	Rack	\$ 470
3	Connectors	1,770
1	Rod shear	10,400
1	Dissolver (jacketed), 50 liters, 2 legs - 6-in. diam x 8 ft long	11,750
2	Scrubber columns, 22 liters, 1 leg - 6-in. diam x 4 ft long	700
1	Tank, (agitated), 50 gal.	1,500
2	Tanks, 5 gal.	600
2	Tanks (one jacketed), 90 liters 2 legs - 6-in. diam x 8 ft long	3,100
1	Tank, 11 liters, 1 leg - 6-in. diam x 2 ft long	200
1	Iodine reactor and cooler	~2,000
1	Ejector, 6-in. water	500
1	Ejector condenser	4,000
1	Evaporator condenser, 10 gph	600
1	Pump (Lapp-Pulsafeeder - remote head), 50 cc/min, max	1,300
8	Jets, 2 gpm	2,400
50	Dissolver baskets at \$70	<u>3,500</u>
	Sub-total	\$44,790
<u>Instrumentation</u>		
1	Dissolving temperature recorder- controller	950
1	12-point temperature recorder	<u>3,000</u>
	Sub-total	\$ 3,950

*Grand totals are given in Table 22.

KAPL-1757

TABLE 17. Solvent Extraction - Costs*

<u>Equipment</u>		
<u>Number</u>	<u>Description</u>	<u>Cost</u>
1	Mixer settler, 16 stages	\$ 7,000
1	Mixer settler, 12 stages	6,000
3	Tanks (agitated), 50 gal.	4,500
1	Tank, 220 liters, 5 legs - 6-in. diam. x 8 ft long	4,100
1	Tank, 135 liters, 3 legs - 6-in. diam x 8 ft long	2,400
1	Tank, 90 liters, 2 legs - 6-in. diam x 8 ft long	1,600
1	Lapp-Pulsafeeder pump, no remote head, 50 cc/in., max	1,100
2	Lapp-Pulsafeeder pumps, no remote head, 150 cc/in., max	2,200
6	Jets, ~2 gpm	1,800
2	Bank motors, 1/3 hp dc	100
2	Motor rectifiers and controllers (motor generator set)	2,000
1	Rack	470
4	Connectors	<u>2,360</u>
	Sub-total	\$35,630
<u>Instrumentation</u>		
2	Single-point interface level recorders	900
2	Mixer speed indicators (ammeter and voltmeter)	<u>100</u>
	Sub-total	\$ 1,000

*Grand totals are given in Table 22.

TABLE 18. Mixed Oxide Preparation - Costs*

<u>Equipment</u>		
<u>Number</u>	<u>Description</u>	<u>Cost</u>
2	Precipitators (jacketed), 90 liters, 1 leg - 6-in. diam x 16 ft long	\$ 6,000
1	Rotary drum vacuum filter unit	7,000
1	Electric furnace, H ₂ atmosphere, muffle - 6-in. diam x 4 ft long	35,000
1	Scrubber column, 22 liters, 1 leg - 6-in. diam x 4 ft long	350
1	Tank, 5 gal.	300
2	Tanks, 90 liters, 1 leg - 6-in. diam x 16 ft long	2,500
1	Tank, 17 liters, 1 leg - 6-in. diam x 3 ft long	300
1	Pump, canned rotor, 600 cc/in., max	150
1	Pump, centrifugal, 2 gpm	250
1	Jet, 2 gpm	300
1	Sampling system	<u>10,000</u>
	Sub-total	\$62,150
<u>Instrumentation</u>		
1	Single-point temperature recorder	250
1	pH meter (remote indicating - in line)	~10,000
1	Liquid level, specific gravity indicator	450
4	Flow indicator gas rotameters, 0 to 1 cfm	<u>400</u>
	Sub-total	\$11,100

*Grand totals are given in Table 22.

TABLE 19. Fuel Element Fabrication - Costs*

<u>Equipment</u>		
<u>Number</u>	<u>Description</u>	<u>Cost</u>
6	Blenders, 500-gm capacity	\$ 2,400
1	Extruder and take-up system	10,000
1	Drying rack (humidity controlled), 6-ft x 6-ft x 12-ft cabinet - 36 shelves	10,000
1	Cutting and loading machine	5,000
1	Electric furnace, H ₂ atmosphere, muffle - 6-in. diam x 5 ft long	35,000
1	Inspecting equipment	5,000
1	Heliarc welder and welding station	15,000
1	Loading unit	13,000
1	Leak test unit	7,000
8	In-process storage units	9,000
4	Carriers	9,000
Sub-total		\$119,400

*Grand totals are given in Table 22.

TABLE 20. Oxide Waste Recovery - Costs*

<u>Equipment</u>		
<u>Number</u>	<u>Description</u>	<u>Cost</u>
1	Dissolver (jacketed), 50 liters, 2 legs - 6-in. diam x 8 ft long	\$11,750
1	Scrubber column, 22 liters, 1 leg - 6-in. diam x 4 ft long	350
1	Tank, 90 liters, 2 legs - 6-in. diam x 8 ft long	1,600
1	Ejector	500
1	Ejector condenser	4,000
4	Jets	1,200
25	Dissolver baskets at \$70	1,750
Sub-total		\$21,150
<u>Instrumentation</u>		
1	Dissolver temperature controller	950
1	8-point temperature recorder	2,000
Sub-total		\$ 2,950

*Grand totals are given in Table 22.

KAPL-1757

TABLE 21. Waste Disposal - Costs*

<u>Equipment</u>		
<u>Number</u>	<u>Description</u>	<u>Cost</u>
1	Tank (jacketed), 200 gal. 24 in. x 96 in.	\$ 2,250
1	Tank, 100 gal., 24 in. x 48 in.	650
2	Tanks (jacketed), 50 gal.	1,400
2	Tanks, 25 gal.	300
1	Tank, 5 gal.	200
1	Hold basin, 1000 gal.	5,800
6	Condensers	3,600
11	Jets	3,300
1	Rack	470
3	Connectors	<u>1,770</u>
Sub-total		\$19,740

<u>Instrumentation</u>		
3	Strip chart, single-point temperature recorders, 0 → 300°C	750
1	12-point temperature recorder	3,000
8	Liquid level - specific gravity, volume systems	<u>4,000</u>
Sub-total		\$ 7,750

*Grand totals are given in Table 22.

TABLE 22. Solvent Recovery - Costs

<u>Equipment</u>		
<u>Number</u>	<u>Description</u>	<u>Cost</u>
1	Tank, 500 gal.	\$ 3,900
2	Tanks, 100 gal.	3,200
1	Tank (agitated), 100 gal.	2,300
1	Tank, 50 gal.	450
5	Jets	<u>1,500</u>
Sub-total		\$11,350

<u>Instrumentation</u>		
4	Liquid level - specific gravity indicators	<u>2,000</u>
Sub-total		\$ 2,000

Equipment Grand Total		314,210
Instrumentation Grand Total		<u>28,750</u>
Equipment and Instrumentation Grand Total		\$342,960

TABLE 23. Cost of Equipment Spares

<u>Number</u>	<u>Description</u>	<u>Cost</u>
3	Manipulators at \$20,000	\$ 60,000
1	Dissolver at \$11,750	11,750
3	Shears at \$7,200	21,600
1	Extruder at \$10,000	10,000
3	Racks at \$470	1,410
6	Connectors at \$590	<u>3,540</u>
Total Cost of Delivered Spares		\$108,300

TABLE 24. Direct Labor Charges

<u>Number</u>	<u>Description</u>	<u>Cost</u>
9	Operators for chemical plant at \$5,500 per year	\$ 49,500
25	Operators for refabrication plant at \$5,500 per year	137,500
2	Shift engineers at \$8,000 per year	<u>16,000</u>
Total Direct Labor Cost		\$203,000

TABLE 25. Plant Investment

Major process equipment, delivered	\$ 314,210	
Major process equipment, installed	439,890	\$ 439,890
Instruments, delivered	28,750	
Instruments, installed	47,440	47,440
Process piping	188,530	188,530
Nonmanufacturing capital investments		
Building estimate	842,830	
Auxiliary estimate	314,210	
Cost of delivered spares	<u>108,300</u>	
	\$1,265,340	<u>1,265,340</u>
Total physical cost		\$1,941,200
Plant construction overhead		<u>776,480</u>
Total plant investment before startup		2,717,680
Startup		<u>271,770</u>
Total Plant Investment		\$2,989,450

TABLE 26. Annual Operating Cost

Raw materials	\$ 50,000	\$ 50,000
Direct labor	203,000	203,000
Indirect manufacturing expenses		
Process utilities	63,000	
Maintenance	111,270	
Supplies	9,470	
Supervision and clerical	50,750	
Property taxes and insurance	89,680	
Laboratory	50,750	
Employee benefits and continuity of service	46,410	
Miscellaneous plant overhead	<u>224,690</u>	
	646,020	646,020
Depreciation	298,950	<u>298,950</u>
Shop cost		1,197,970
Manufacturing cost addend	59,900	<u>59,900</u>
Manufacturing cost		1,257,870
Waste storage	50,000	50,000
Profit before taxes	326,160	<u>326,160</u>
Annual Operating Cost		\$1,634,030

TABLE 27. Total Fuel Cycle Cost

Inventory charge (at 6%)		
Uranium - 50 tons natural U at \$40/kg	}	\$ 552,000
Plutonium - 600 kg Pu at \$12/gm		
Core fuel cost		
\$400,000 included for subassembly fabrication		2,034,030
Blanket processing		
Shipping	at \$ 5/kg	
Processing	at \$ 25/kg	
Fabrication	at \$ <u>10/kg</u>	
23.75 tons natural U	at \$ 40/kg	<u>950,000</u>
	COST, \$/year	3,536,030
	mils/kw-hr	2.04
Credit for plutonium breeding gain		
(assume breeding gain = 0.3)		
(assume \$12/gm for Pu)		<u>890,000</u>
		0.51 mils/kw-hr
Net Fuel Cycle Cost		2,646,030
		1.53 mils/kw-hr

NOTE: $\frac{\text{kw-hr}}{\text{year}} = 800,000(0.3)(300)(24) = 1.73 \times 10^9$

KAPL-1757

Fuel cycle cost, $\frac{\text{mils}}{\text{kw-hr}} = \frac{\text{cost, \$ / year}}{1.73 \times 10^6}$

PLANT AND FLOWSHEET DEMONSTRATIONS

The major emphasis of work on the FOB fuel cycle has been on the development and demonstration of the essential features of the core re-processing cycle in order to establish a firm basis for the fuel cycle cost estimate. A large number of experiments and engineering demonstrations have been carried out both with actual fuel material containing plutonium and, when the quantities of plutonium became too large to handle easily, with natural uranium or a mixture of uranium and thorium (where thorium was a substitute for plutonium). These experiments and demonstrations are described in the following sections.

Fuel Dejacketing and Dissolution

Fuel Shear

Spent reactor fuels which can be leached from their cladding material permit a reduction in the size and complexity of the processing plant equipment through the elimination of high concentrations of cladding inerts in the feed stream. Mechanical dejacketing of the fuel element to expose the fuel for leaching can be accomplished in a number of ways, depending upon the nature of the core-to-cladding interface and the geometry of the fuel element.

Initial irradiation experiments have shown that the FOB fuel sometimes develops a void along the central axis and sinters radially toward the cladding wall. After irradiation, the mixed oxides are friable but not powdery.⁷ Although no metallurgical bond is formed, the condition of the fuel eliminates the possibility of dumping or pushing the core from the cladding and, in fact, requires some sort of shearing or crushing action to expose the fuel for chemical dissolution. Because of the pin-type geometry of the fuel element, a shearing technique has been developed for the dejacketing operation.

The fuel rod shear and general arrangement used in the demonstration are shown in Figures 17 and 18. This unit will shear rods of 0.25-in. diameter or smaller into 1/2-in. sections with a minimum of crimping of the cladding. The shear consists of a die that moves with a reciprocating motion across a circular hole in the face of the die plate. Powered feed rollers, shown in Figure 17, move the rod uniformly from a collection trough through the die hole and against a stop on the moving die that determines the length of the sections. A pickup finger is located in the feed trough to force the fuel rod into the roller system.

A number of simulated fuel elements made by swaging stainless steel tubing over sintered alumina rod have been used for testing the equipment. These tests show that the deformation of the cladding and chipping of the fuel at the point of sectioning were not excessive at operating rates up to 75 cpm. In addition to testing the shear with alumina rod, the

FIGURE 17. SHEAR FEED ROLLERS

FIGURE 18. FUEL ROD SHEAR

KS-8645
Unclassified

equipment has been used to shear a dissolver loading of fuel consisting of 0.180-in.-diam uranium and thorium dioxide rods clad with 0.015-in.-wall stainless steel tubing.

Fuel Dissolution

One of the main economic advantages of the UO_2 - PuO_2 fuel lies in the chemical simplicity of oxide preparation compared with metal preparation and fabrication. At the same time the oxide was expected to give difficulty in the aqueous dissolution of fuel for chemical processing. It is well known that ignited PuO_2 goes into solution only with difficulty, even in HNO_3 - HF mixtures, whereas UO_2 is readily soluble in HNO_3 . It was hoped at the outset that the PuO_2 dissolution problem would not be severe if the oxides were formed as a solid solution of PuO_2 in UO_2 .

The chemical approach to fuel preparation was to find conditions for the formation of a solid solution of the oxides. This was thought to be possible because the crystal structures are isomorphous and the difference in lattice constants is not large enough to preclude mixed crystal formation. The fact that ThO_2 - UO_2 mixed crystals are known supports this hypothesis.

The simplest sequence of chemical reactions was found to lead to oxide solid solution. Uranyl nitrate and plutonium(IV) nitrate in atomic ratios varying from 6 to 1 to 4 to 1 uranium to plutonium were mixed in 0.1- to 0.3-M acid solution and precipitated with gaseous ammonia. The yellow-green precipitate appeared to be homogeneous, although no study was made of its chemical composition or structure. The cake filtered well, and less than 0.1% of either metal ion was found in the supernate. After being washed with one cake volume of water and partially dried, the material was decomposed to the mixed dioxides by heating in a stream of dry hydrogen from 1 to 2 hr at temperatures of 600, 800, and 1000°C. The products were cooled in hydrogen and were found to take up oxygen slowly at room temperature on exposure to air. The mixed dioxides were of small particle size, in general, with any agglomerates easily friable. No fines or dusts were observed.

X-ray diffraction patterns of these products showed complete solid solution formation. After 5 hr of heating in 60% nitric acid, all products had dissolved. It should be noted that in some of these studies a minute amount of fine black precipitate remained after 5 hr of dissolution. This precipitate also occurred in cold studies in which no plutonium was present. The precipitate did not dissolve in the presence of hydrofluoric acid and is believed to have been platinum from the filter disk used.

To test the feasibility of using reprocessed fuel, the product end stream from the irradiated solvent portion of run FOB-102 (see Solvent Extraction, p. 62) was used in making mixed oxide. The end stream was washed with Ultrasene to remove tributyl phosphate (TBP) and hydrolysis

products and was then adjusted to a plutonium-to-uranium atomic ratio of 1 to 5. This adjusted product was precipitated with ammonium hydroxide, centrifuged, and washed with water. The cake was dried and fired in a stream of hydrogen at $\sim 800^{\circ}\text{C}$ for 1 hr and then cooled in hydrogen. X-ray diffraction patterns showed complete solid solution formation with no indication of phosphate. This fuel completely dissolved as readily as other mixed $\text{PuO}_2\text{-UO}_2$ fuels.

Pure plutonium dioxide prepared in the same way as the mixed dioxides did not dissolve in nitric acid and required long heating with nitric acid - hydrofluoric acid for partial dissolution. A mixture of plutonium dioxide and higher uranium oxide, formed from the ammonia precipitate by heating at 600°C in air, exhibited only partial dissolution of the plutonium in concentrated nitric acid.

An attempt was made to produce a solid solution of the dioxides directly from the mixture of plutonium(IV) and uranyl nitrates in an atomic ratio of 1 to 5 plutonium to uranium in acid solution. In a stream of dry hydrogen the solution was evaporated to dryness, and the resulting solid was heated at 800°C for 1 hr. X-ray diffraction pictures of this product showed separate uranium dioxide and plutonium dioxide patterns. Only partial dissolution of the plutonium dioxide was effected in hot concentrated nitric acid.

The peroxide precipitation of a solution containing plutonium(IV) and uranyl nitrate in an atomic ratio of 1 to 7 plutonium to uranium in 2-M acid gave an excellent product with $<1\%$ loss of plutonium ion to the supernate. However, the peroxide precipitates did not form a solid solution when calcined at 800°C in dry hydrogen.

No attempts were made to form mixed dioxides from the oxalate precipitates or the precipitates of any other anions.

Nonirradiated Mixed Oxide Dissolution Studies

Dissolutions of nonirradiated pellets of mixed $\text{PuO}_2\text{-UO}_2$ were carried out to determine conditions which give the maximum rate of dissolution in nitric acid and which lead to the minimum concentration of acid in the dissolver solution.

Three pellets of mixed oxides in which the plutonium-to-uranium ratio was 1 to 5.4 were dissolved. In each dissolution the acid was added at 25°C ; then the dissolver vessel was heated to 99°C and maintained at that temperature throughout the dissolution. Pellets 1 and 2 were dissolved in different volumes of 13.4-M nitric acid. Pellet 3 was dissolved in a mixture of 10.7-M nitric acid and 5×10^{-3} -M hydrofluoric acid. In each dissolution the initial reaction was vigorous and accompanied by the

evolution of nitrogen dioxide, which ceased after 30 min of reaction time. At this time the undissolved portion of each pellet was in the form of a black suspension. The dissolution of this black suspension required from 1.5 to 4.5 hr of further heating at 99°C.

The results of these dissolutions, shown in Table 28, indicate that in the absence of fluoride ions relatively long periods of heating will be necessary if the final acidity of the dissolver solution is to be low enough for solvent extraction separations.

TABLE 28. Data on the Dissolution of Nonirradiated Mixed Oxide Pellets

<u>Pellet</u>	<u>Initial HNO₃, M</u>	<u>Initial HF, M</u>	<u>Dissolution Time, hr</u>	<u>Final HNO₃, M</u>	<u>Final Uranium, M</u>	<u>Final Plutonium, M</u>
1	13.4	-	5.1	4 to 5	2.01	0.39
2	13.4	-	3.2	7 to 8	1.31	0.25
3	10.7	0.005	2.1	4 to 5	1.40	0.27

Irradiated Mixed Oxide Dissolution

Dissolution of irradiated mixed oxide from the inside of one-half of the irradiation capsule described in KAPL-1446⁷ was accomplished smoothly and rapidly with hot concentrated nitric acid. Although the inside bore of the capsule was only 0.188 in. and the half-capsule was positioned vertically with the bottom end closed, the evolution of nitric oxides provided sufficient agitation to complete the dissolving within 2 hr. The resulting solution was clear and without evidence of the colloidal suspension noted in previous cold dissolutions.

The sectioning of the capsule and dissolution of the oxides were carried out in the Radioactive Materials Laboratory (RML). An ordinary tubing cutter was used to cut the capsule at the center, and one-half of the capsule was then dropped through an air condenser into a small quartz flask mounted on a clamp stand. The flask and stand were designed to fit into the 3-1/2-in. bore of a 14-in.-diam cask so that convenient transfer from the RML cave to the Engineering High Level Laboratory (EHLL) could be made. To facilitate transfer of the feed solution, the flask was constructed with a ball and socket joint so that the condenser could be replaced by a transfer joint containing polyethylene vent and dip legs. These vent and dip legs extended through the 3/8-in. stainless steel vertical member of the clamp stand, and this, in turn, extended through

a hole drilled in the top plug of the cask. After assembly, shielding was sufficient to permit direct access to the polyethylene leads for final transfer to the EHLL feed system.

Dissolution of the mixed oxide was carried out in a three-fold excess of 15.1-M nitric acid in order to cover the upper, open end of the half-capsule. After addition of the acid at room temperature, the water bath surrounding the vessel was heated to boiling. Reaction started immediately upon the addition of cold acid and remained vigorous for 1 hr but subsided in the next hour until there was no further evolution of nitrogen dioxide. At the end of this time, the source of heat was removed and the solution was allowed to cool for 1 hr. There was no evidence of black suspension, and upon examination of the capsule no oxide was found to be present. After the solution was mixed by bubbling air through the sampling pipet, a sample was taken which was analyzed as follows:

$H^+ = 6.73 \text{ M}$
 $\beta = 1.70 \times 10^{10} \text{ c/m/ml}$
 $\gamma = 2.07 \times 10^{10} \text{ c/m/ml}$
 $\alpha = 7.93 \times 10^8 \text{ c/m/ml}$, equivalent to 0.042-M plutonium
 (assuming no isotope ratio change)
 $U = 0.26 \text{ M}$
 $Mg = 0.92 \text{ M}$, calculated from the weight of the MgO pellet at
 the end of the capsule. (There was no evidence
 of MgO dissolution.)

Dissolution of irradiated mixed oxide from inside the long-burnup fuel capsule, KAPL-26-2,⁸ was accomplished smoothly with hot 13.5-M nitric acid in 5 hr. The resulting solution was clear and without evidence of undissolved oxide. This capsule, which contained plutonium dioxide and uranium dioxide in a ratio of 1 to 5.4, had been irradiated in the MTR to an estimated 35% burnup of the plutonium.

The sectioning of the capsule and dissolution of the oxides were carried out in RML facilities. A cutting wheel was used to remove the ends of the capsule and to section the capsule near the center. After removal of the MgO end plugs, both sections of the capsule were dropped into a quartz flask similar to one used in previous dissolvings.

Dissolution of the mixed oxide was carried out in sufficient 13.5-M nitric acid to meet feed specifications after dissolution. After addition of the acid at room temperature, the water bath was heated to boiling. Definite reaction was evident 10 min after acid addition and remained

vigorous with nitrogen dioxide evolution for the next 20 min. The solution was dark green and nearly opaque with a black suspension, which slowly dissolved and completely disappeared after 5 hr. After the solution was cooled and diluted with water, a sample was taken which was analyzed as follows:

$\text{HNO}_3 = 5.1 \text{ M}$, high concentration probably resulted from efficient acid utilization in dissolving UO_2 .
 $\beta = 1.60 \times 10^{11} \text{ c/m/ml}$
 $\gamma = 2.00 \times 10^{11} \text{ c/m/ml}$
 $\alpha = 3.80 \times 10^9 \text{ c/m/ml}$
 $\text{U} = 0.92 \text{ M}$.

Plant-Scale Dissolver Demonstration

As was pointed out in the discussion of the plant design basis, a "throw-away" maintenance concept in a plant of this size should result in a reduction in both the initial plant investment and the annual operating cost. Because the application of this concept involves the use of equipment of novel design, pilot plant models of a shear, dissolver, equipment rack, and disconnect flange were constructed. This was considered necessary, since it would permit demonstration of the unique features of the equipment and at the same time provide a sound basis for the cost estimation of the FOB plant equipment.

The dissolver was identical in design to that described in the plant process equipment section with the exception that lengths of glass pipe were installed in the vertical legs to permit observation of the hydraulic operation of the equipment. The dissolver was mounted on a disposable rack 8 ft high, 8 ft long, and 3 ft wide constructed of 1-in. angle iron. Other design features were identical to those of the plant model rack. The rack is shown in Figure 19, and details of the locking and alignment device are shown in Figure 20. Tests indicated that up to 1/2-in. lateral and 1/4-in. longitudinal alignment errors can be corrected by the positioning and locking mechanism. The remote disconnect flanges used to connect the dissolver vessel to the external tanks and service line were also similar to those intended for plant use. The details of the connector are shown in Figure 21, and another view of the assembly is shown in Figure 22 (see also Figure 15, p. 38). Three of these units were installed on the prototype rack and a simulated cell face, which was also used as a panel board for the pilot plant instrumentation. The experimental shear previously described was mounted on a platform above the dissolver rack. Two views of the assembled equipment are shown in Figures 23 and 24.

FIGURE 19. EQUIPMENT RACK

KS-8648
Unclassified

FIGURE 20. RACK ALIGNMENT AND LOCKING DEVICE

KS-8646
Unclassified

- NOTES
- ① CLAMP PT-4 & PT-6 TOGETHER ROUGH MACH HOLES UNCLAMP & FINISH MACH AS SHOWN

⑥ EXCEPT AS SHOWN PT-6 SAME AS PT-7

② MACH AS SHOWN FROM 1" HEX SOC HD SCREW

FIGURE 21 REMOTE DISCONNECT FLANGE
KS-20273
UNCLASSIFIED

FIGURE 22. DISCONNECT FLANGE ASSEMBLY

KS-8650
Unclassified

FIGURE 23. DISSOLVING PILOT PLANT - SIDE VIEW

KS-16085
Unclassified

FIGURE 24. DISSOLVING PILOT PLANT - END VIEW

KS-16088

Unclassified

Operation and Testing of the Pilot Plant

Two feed dissolution tests (FDT-1 and -2) were run in the pilot plant. The main purpose was to check the hydraulic operation of the dissolver, but in the process some dissolution data were also collected. The hydraulic operation was not satisfactory in FDT-1 because of the lack of circulation of liquid between the two legs of the dissolver. This resulted in a large difference between the densities of the solutions in the two legs, which could not be corrected easily even upon dilution. This defect was corrected in FDT-2 by the addition of a horizontal member below the liquid level to allow convective circulation of the solution between the two vertical legs. No other hydraulic difficulties were encountered.

Test FDT-1 was made with a feed material consisting of natural uranium dioxide powder and stainless steel rings in the weight ratio specified by the fuel element design; 9.0-M nitric acid was used in the dissolution, during which 2.8 moles of acid were consumed for each mole of oxide dissolved. The average dissolution rate over the first hour was 212 gm/min. No residue was observed in the dissolver after an initial dissolution period of ~1 hr.

Test FDT-2 was made with a feed material consisting of a solid solution of thorium dioxide and uranium dioxide with a uranium-to-thorium ratio of 5 to 1. The material was clad with stainless steel and fabricated by the extrusion technique designated for the plant. The fuel was sectioned without difficulty by the fuel shear unit and charged to the dissolver; 14-M nitric acid was used for the dissolution. For each mole of oxide dissolved, 4.1 moles of acid were consumed. The dissolution rate over the first hour was approximately the same as in FDT-1, but a residue remained after the reaction subsided. This residue was assumed to be thorium dioxide. Heating for an additional 4 hr at boiling point did not bring the residue into solution.

Operation of the disconnect flanges and other auxiliaries was satisfactory in both tests.

Solvent Extraction

Mixer-Settler Laboratory System

The facilities at the Knolls Atomic Power Laboratory for the laboratory study of aqueous solvent extraction reprocessing of spent reactor fuels have been described previously.^{9,10} It was necessary to modify these facilities for the study of systems containing macro-concentrations of plutonium.¹¹ These modifications were of two types: first, the

installation of the mixer-settler banks, product vessels, and sampling equipment in enclosures which satisfied health physics requirements for plutonium containment, and second, the improvement of the mixer-settler drive mechanism so that the contactors could be operated for long periods without maintenance. The use of the modified equipment with existing feed preparation and waste handling facilities resulted in an integrated system for the demonstration of the proposed solvent extraction flowsheet. The modified system is shown in Figures 25, 26, and 27.

Flowsheet Considerations

In order to keep the inventory of fissile material at a minimum, it is necessary to have a fuel reprocessing step capable of handling highly radioactive, short-cooled fuel. Reprocessing must provide for adequate decontamination and essentially complete recovery of uranium and plutonium and must lead to a simple step for adjustment of the metallic ion ratio before preparation of the mixed oxide. Since the high concentration of plutonium present in the fuel will in itself require remote handling, extremely high decontaminations will not be required.

As a first step, a single-cycle process was sought which would reduce the radiation from the fuel by several orders of magnitude. For this purpose it was proposed to use a Purex-type solvent extraction process with 30 vol % TBP in a hydrocarbon diluent as the extracting phase and nitric acid as the aqueous phase. This is a well-understood, simple, and economic process of high capacity. With such a system co-decontamination of uranium and plutonium can be accomplished in two mixer-settler banks with fuel dissolved in nitric acid as the feed.

The system is relatively stable in comparison to other solvent extraction systems with respect to radiation and hydrolytic damage and requires relatively simple processing steps and equipment in comparison to nonsolvent extraction processes. It also supplies a product which is readily sweetened with plutonium and from which the oxide can be prepared without difficulty.

If further decontamination is desired to simplify the requirements for remote handling of product, second and even third solvent extraction cycles similar to the first can be utilized. Since these latter cycles would operate at radiation levels corresponding to those normally encountered in fuel processing, it is expected that data already available will give an accurate picture of results to be expected.

Although the design of a Purex-type flowsheet for the recovery and decontamination of macro-uranium and -plutonium is relatively simple, the high burnup and short cooling necessary for an economic fuel cycle introduce questions concerning the behavior of the flowsheet.

FIGURE 25. MIXER-SETTLER FEED SYSTEM

KS-6471
Unclassified

FIGURE 26. MIXER-SETTLER CASCADE

KS-6472
Unclassified

FIGURE 27. SOLVENT EXTRACTION PRODUCT RECEIVERS

KS-6470
Unclassified

Of the problems that can be foreseen are the following:

1. The effect of radiation damage to the solvent on decontamination and recovery
2. The effect of radiation damage to the solvent on mixing and settling in the banks
3. The effect of macro-concentrations of fission products on decontamination, and
4. The effect of the fast neutron spectrum and short cooling on fission-product distribution in spent fuel and the resulting effect on the process.

It was possible to simulate conditions pertaining to the first two problems by exposing the solvent to varying dose rates before use in laboratory solvent extraction runs. A more extensive study of the effect of radiation on hydraulic behavior is reported elsewhere.¹²

The problems introduced by macro-concentrations of fission products were not studied but are not thought to be insuperable, provided that a suitable feed clarification step is found to remove solid material such as niobium pentoxide and zirconium polymers. Macro-concentrations of zirconium may actually tend to give higher zirconium decontaminations than those obtained in the laboratory runs, since zirconium will probably be present in concentrations greater than the concentration of dibutyl phosphate (DBP), the radiation-formed hydrolysis product of TBP, which adversely affects decontamination because of the strong organic soluble complex it forms with zirconium.

The change in fission-product distribution with fast spectrum is not expected to be a serious problem, since the yields of fission products limiting decontamination will not change markedly. Feed for the Mini mixer-settler runs was spiked with small amounts of mixed oxide fuel which had been subjected to burnups of up to 35% in the MTR pile, which has a neutron spectrum lower in energy than that of the proposed FOB reactor.

Short cooling will introduce a problem, since the fission-product iodine will react with the hydrocarbon diluent, especially with those radicals formed under the influence of radiation. This will limit decontamination of the solvent, because iodine is not removed effectively in the washing step. However, because of the short (8-day) half-life of I^{131} , the buildup of this activity will be limited and, thus, will probably not prevent reuse of the solvent in the first cycle. The behavior of the process with iodine activity of this magnitude was not studied, although it may be assumed from past experience with the Purex process that a significant portion of this activity present in the feed will become associated with the solvent.

Flowsheet Design

Because of the slow dissolution rate of mixed oxide fuel at low nitric acid concentrations, the design of the FOB TBP flowsheet was based on experience with high acid flowsheets. It was assumed that

1. The salting effect of plutonium nitrate in the aqueous phase was not significant
2. The saturating effect of the plutonium TBP complex on uranium distribution could be represented by correcting such distribution for an original TBP concentration lowered by the amount associated with plutonium, and
3. The extraction coefficients of plutonium(IV) nitrate are one-fourth those of uranyl nitrate when both are present in macro-concentrations.¹³

Stages were stepped off on a McCabe-Thiele type X-Y diagram starting from the product end of the 1A extraction system. The theoretical X-Y diagrams for the 1A bank are shown in Figure 28, and profiles of nitric acid, uranium, and plutonium concentrations in an actual run are shown in Figure 29. Despite the difficulty experienced in handling and analyzing these small stage samples, the profiles indicate that the flowsheet performed as expected. Flowsheet conditions are given in Table 3, page 21. No troubles that could be attributed to flowsheet design were experienced during the runs, although not enough allowance was made for the variations experienced in feed concentrations and flowrates as evidenced by occasional high LAW losses in the laboratory runs. More room for analytical inaccuracies and flowrate variation can readily be introduced without major change in the flowsheet by dropping the scrub flowrate by 25%.

Equipment and Procedures

The Mini mixer-settler equipment used in making the runs was described in the previous section. The extraction (1A) bank consisted of seven extraction and nine scrub stages; the stripping (1C) bank contained twelve stages. Runs extended over periods of ~1 week or longer with actual running times of 16 to 50 hr. Most of the runs were subdivided into periods in which solvents having different radiation doses were used. When possible, the use of any one solvent was continued until steady-state losses and decontaminations were obtained. Solvent irradiation consisted of exposing washed, water-saturated, 30 vol % TBP in Ultrasene to radiation from a cobalt source. The solvent was contained in a glass doughnut. A complete description of the solvent irradiation has been presented elsewhere.¹²

FIGURE 28 THEORETICAL X-Y DIAGRAMS FOR IA BANK, FOB FLOWSHEET

KS-20319
UNCLASSIFIED

FIGURE 29 STAGE PROFILES-RUN FOB-102

KS-20342
UNCLASSIFIED

Run Results

The significant effects of irradiated solvent on the process are given in Tables 29, 30, and 31. The dose that the solvent is expected to be subjected to in FOB processing is 1.7 watt-hr/liter. However, tests with a two-phase mixed system under irradiation have shown that the amount of DBP produced under such conditions is approximately three times that obtained by irradiating the solvent alone.¹² For this reason the results

obtained from a solvent given a dose of 5.3 watt-hr/liter are assumed to be comparable to results under actual FOB processing conditions. The behavior of the flowsheet with the solvent having higher doses of radiation confirms the effects noticed at the dose expected under normal FOB conditions. As far as could be observed, the use of irradiated solvent had little if any effect on the 1A bank extraction section. However, it was impossible to ascertain whether or not there might have been some slight formation of precipitate in this bank. Any large amount of precipitate could have been seen. The formation of a light-colored, flocculent solid was observed at the raffinate end of the stripping bank after long periods of running. This precipitate accumulated more rapidly as the radiation dose to solvent increased; however, it was not of a magnitude expected to interfere with mixer-settler operation, since the precipitate accumulated at the interface and was gradually carried over into the organic raffinate. The quantities formed were not large enough to allow a determination of the nature of the precipitate.

TABLE 29. Effect of Solvent Irradiation on Product Loss

	<u>Solvent Irradiation,</u> <u>watt-hr/liter</u>	<u>Loss, %</u>
<u>1AW (Aqueous Waste)</u>		
Uranium	0 to 24	< 0.01
Plutonium	0 to 24	~ 0.01*
<u>1CW (Organic Waste)</u>		
Uranium	0	0.006
	5.3	0.06
	16	0.2
	24	0.6
Plutonium	0	0.007
	5.3	0.05
	16	0.2
	24	0.7

*Slight, if any, increase in loss with irradiation of solvent.

TABLE 30. Effect of Solvent Irradiation on Log
Decontamination of Product (1CP)

	<u>Solvent Irradiation, watt-hr/liter</u>	<u>Log Decontamination Factor</u>
Beta	0	2.9 to 4.2
	5.3	2.7
	16	2.1
	24	<2.2*
Gamma	0	2.5 to 3.5
	5.3	1.5
	16	0.9
	24	<1.5*
Zirconium-niobium	0	2.5 to 3.5
	5.3	1.5
	16	0.9
	24	1.5*,**
Total rare earths	0 to 24	5†
Ruthenium††	-	-
I ₂ ‡	-	-

*Not at steady-state operation. Decontamination was still decreasing at end of run; should be compared with higher of two results with solvent having no irradiation.

**Assumed from gamma decontamination factor.

†Irradiation of solvent has very little effect on decontamination, which is, however, sensitive to hydraulic operation. A log decontamination factor of 5 is expected with good operation.

††Masked by zirconium-niobium.

‡Not known.

TABLE 31. Effect of Solvent Irradiation on Percentage of LAF Activity and Uranium in LCW before and after Washing

	Solvent Irradiation watt-hr/liter	Before Washing, %	After Washing, %*
Beta	0	0.001	0.00004
	5.3	0.01	0.0002
	16	0.1	0.03
	24	0.1	-
Gamma	0**	0.002	0.00005
	5.3	0.3	0.004
	16	4	1
	24	-	-
Plutonium	0	0.007	0.00005
	5.3	0.05	-
	16	0.2	-
	24	0.7	0.1
Uranium	0	0.006	-
	5.3	0.06	-
	16	0.2	-
	24	0.6	-
I ₂ [†]	-	-	-

NOTE: The effect of macro-concentrations of fission products was not known.

*Standard procedure: water, 0.1-M NaOH, water, 0.06-M HNO₃. Each wash was followed by a standing period and discard of the wash solution.

**75% zirconium-niobium, 25% ruthenium. Decontamination factors of 70 for zirconium-niobium and 30 for ruthenium were obtained.

[†]Not known.

The effect of using irradiated solvent is shown very clearly in the losses of the uranium and plutonium to the organic raffinate from the stripping bank and in the carrying of zirconium-niobium activity to the product and spent solvent streams. As far as could be determined, the decontamination of product from ruthenium was not affected by the use of irradiated solvent; however, the analysis was difficult because of the

masking of ruthenium activity by that of zirconium-niobium. Decontamination from rare earths was not affected. Not only was the carrying of zirconium-niobium activity into the spent solvent stream adversely affected by the use of irradiated solvent but also the ability to remove such activity from the solvent by standard washing procedures. It is believed that the behavior of the process will be satisfactory at the radiation doses expected, corresponding to the solvent irradiation of 5.3 watt-hr/liter in these tables.

Material balances and behavior of the flowsheet during a run having good hydraulic behavior are presented in Figures 30, 31, 32, and 33. The alpha material balance was low at the start of the run because the banks originally contained no alpha activity, although they were already at steady-state conditions with respect to uranium and acid. Poor alpha balance subsequent to that time was very likely the result of an error in the feed analysis which had been impossible to trace. This analysis indicated a plutonium molarity of 0.23 M, 30% higher than calculated on the basis of stock dilution. Conditions for four runs are given in Table 32.

TABLE 32. FOB Run Conditions

	Nominal	Run Number			
	<u>Flowsheet</u>	<u>FOB-100</u>	<u>FOB-102</u>	<u>FOB-103</u>	<u>FOB-105</u>
1AF					
Relative flowrate	100	100	100	100	100
Uranium, <u>M</u>	1.05	1.03	1.14	0.98	1.07
Plutonium, <u>M</u>	0.16	0.15	0.16	0.16	0.23*
HNO ₃ , <u>M</u>	2.00	2.16	2.48	2.51	2.34
1AS					
Relative flowrate	66.7	67	67	67	67
HNO ₃ , <u>M</u>	3.00	2.56	2.94	3.08	2.78
1AX					
Relative flowrate	333.3	354	354	333	333
TBP, vol %	30	30	30	30	30
1CX					
Relative flowrate	500	500	500	500	500
HNO ₃ , M	0.02	0.02	0.02	0.02	0.018

*The feed analysis leading to this value is probably in error, since calculations and analyses of the plutonium spike indicated the molarity should be 0.16. This may account for the ~75% material balance on alpha shown in Figure 33.

4 Cycles x 10 division per inch

General Electric Company, Knolls Atomic Power Laboratory

4 Cycles x 10 divisions per inch

General Electric Company, Knolls Atomic Power Laboratory

Material Balance

Run FCB-105

FIGURE 33

Solvent

Bank
Upset

Aqueous Strip

Acid

Uranium

Alpha *

* See note - Table 32

Run Time - Minutes

0 1000 2000 3000

150%

100%

50%

150%

100%

50%

150%

100%

50%

150%

100%

50%

150%

100%

50%

Alternate Flowsheets

The feasibility of using a precipitation process in lieu of a solvent extraction process for the decontamination of spent FOB fuel was investigated on a test tube scale. In this preliminary study the precipitates of phosphoric acid, oxalic acid, and hydrogen peroxide were examined only for completeness of plutonium precipitation and ease of handling.

Simulated FOB feed consisting of 1.1-M uranyl nitrate, 0.16-M plutonium(IV) nitrate, 2-M nitric acid, and 0.3-M sodium nitrate was used as the precipitating medium. An acid feed of this composition was selected because it can be attained in a straight nitric acid dissolution of spent fuel and because fission-product decontamination is better, as a rule, from an acid medium. Direct strikes were made with precipitating reagents in excess of the amount required to precipitate the total plutonium in solution.

Phosphoric acid produced a clear, gelatinous precipitate which was agglomerated by heating at 95°C. Approximately 10% of the plutonium remained in solution. The addition of sufficient sulfate to form the uranium complex did not improve the qualities of the precipitate or the recovery of plutonium from the solution.

~~Oxalic acid~~ produced a precipitate of excellent settling qualities. Approximately 40% of the plutonium remained in solution. The addition of sulfate decreased the amount of plutonium left in solution to ~20%.

Hydrogen peroxide produced a precipitate similar in settling qualities to the oxalate. The supernate contained ~1% of the total plutonium and was water white. This is in sharp contrast to the phosphate and oxalate supernates, which contained a major portion of the uranium.

Of the precipitates investigated in this study, only the peroxide shows any promise of use in a precipitation process for fuel recovery. Further study with this reagent in the presence of high radiation, fission products, and iron will be necessary before a realistic evaluation can be made.

Mixed Oxide Preparation

Equipment

In order to ensure criticality control during the precipitation step, a tall-pipe, batch precipitator surrounded by a Nichrome heating coil to maintain the precipitation temperature was used. Agitation and introduction of the precipitating agent were accomplished by sparging the feed solution with a mixture of nitrogen and ammonia, which then passed through a de-entrainer and a scrubber to the vent system and stack. The course of the reaction was followed with a pH meter. The precipitator was installed

FIGURE 34 FAST OXIDE BREEDER PRECIPITATOR AND OFF-GAS SYSTEM - SCHEMATIC DIAGRAM

on a balcony so that the top of the precipitator was at the same level as the base of the rotary drum filter to which the precipitate was transferred as a slurry from the vessel through a suction dip leg. Details of the precipitator are shown in Figure 34. The top of the precipitator, and the de-entrainer, scrubber, and gas manifold are shown in Figure 35.

The slurry product from the precipitator was filtered on a laboratory model of an Eimco standard rotary drum vacuum filter of stainless steel construction. The drum was 18 in. in diameter and 12 in. long and provided 4 ft² of filter area. Nylon 513F cloth was used as the filter medium, but a stainless steel cloth of similar weave would be specified for plant use. The pumps, vacuum receivers, and similar auxiliary equipment were all part of the laboratory package unit. Criticality control would be achieved in a plant installation by modification of the drum geometry. Constant submergence of the drum was achieved by an overflow

FIGURE 35. PRECIPITATOR AND ASSEMBLY

FIGURE 36 FAST OXIDE BREEDER PRECIPITATION AND FILTRATION EQUIPMENT - SCHEMATIC DIAGRAM

weir in the filter pan. The slurry was pumped to the pan at a rate in excess of the filtration rate, and the excess material overflow was returned to the precipitator. The filter drum, feed pan, and discharge were contained in a special fume hood connected to the laboratory exhaust system and stack. A diagram of the system is shown in Figure 36. Figure 37 is a photograph of the unit.

Experimental Procedure

The selection of the filter medium was based on data for the filtration of ammonium diuranate precipitated at room temperature from a uranyl nitrate solution meeting solvent extraction product specifications. Initial testing of the laboratory filter with a similar slurry indicated that satisfactory filtrate clarity could not be obtained with the specified filter cloth, and discussions with the manufacturer indicated that precipitation from either a more concentrated solution or a normal solution at an elevated temperature should yield the desired filtrate clarity. Since concentration of the solvent extraction product stream would require an additional process step, it was decided that higher temperature precipitation was preferable, but that both process modifications should be investigated.

FIGURE 37. FILTER UNIT

Because of the limited time available for experimentation and the need to establish both precipitator and filter operating conditions, only the first phase of an investigation of the oxide preparation process variables was undertaken. Preliminary testing of the precipitator had established the permissible range of gas flow rates and batch sizes, and an investigation of the literature established an upper limit on the uranyl nitrate concentration. The range of the ammonia addition rate was established from the desired plant operating schedule and data from the literature survey. It can be seen that this process investigation is essentially a study of the precipitation step, since the filtration is a purely physical operation and the fabrication process is the object of another investigation. It was necessary, however, to evaluate the product of the precipitation on the basis of the reproducibility of fabricated and sintered oxide pieces. To achieve this result, a statistical experiment treating the four precipitation variables was designed with a standard oxide conversion, compaction, and sintering operation as an analytical technique. All samples were calcined and all pellets sintered simultaneously in order to eliminate any effect from variations in either temperature or exposure time.

The first phase of any statistical experiment is generally a testing of the specified independent variables to ensure that some other uncontrolled variable does not have an effect on the dependent variable which will mask or modify the effect of the variables of interest. This is accomplished by running a number of tests with some fixed condition of the independent variables, usually with each variable at the midpoint level of its range. The data from these tests are also useful in specifying the experimental test points in the statistical design.

As previously indicated, there was not sufficient time to complete the entire experiment, so it was decided to carry out the first phase, which consisted of five tests, with all variables at their range midpoints. This would permit demonstration of the equipment and would provide a basis from which other investigators could continue the study. The general test conditions are described below.

The uranyl nitrate stock solution was prepared by dissolving UO_3 in concentrated nitric acid. This stock solution was then saturated with TBP to simulate the solvent extraction product. The precipitator was charged with 33 liters of 0.5-M UNH, 0.5-M H^+ feed solution made by diluting the proper volume of the stock solution. The batch was heated to 47°C and sparged with nitrogen. Ammonia was then mixed with the nitrogen and fed to the precipitator at a nominal rate of 0.25 cfm. The course of the reaction was followed with a pH meter, and the settling rate and solids content of a sample of the slurry were determined at the end of

each run. When precipitation was complete (defined for these tests as the point when $\text{pH} = 9.5$), the ammonia was shut off and the slurry was circulated to the agitated feed pan of the filter. The filter was then operated at a drum speed of $1/3$ rpm until the precipitator was empty. A sample of the slurry was also filtered on the test leaf filter.

The filtrate was examined for clarity, and the handling characteristics of the cake were noted. A sample of the cake was dried overnight in an oven at 100°C and crushed and screened until all material passed through a 16-mesh screen. This material was calcined for 1 hr in a hydrogen furnace at 1000°C , then crushed and screened until all material passed through a 50-mesh screen. A sample weighing 0.48 gm was then pressed in a $3/16$ -in.-diam single-acting die to a fixed height of 0.187 in. The pellet was removed from the die, weighed, measured, and then sintered for 2 hr in a hydrogen furnace at 1600°C . The sintered pellets were then reweighed and measured. The green and sintered densities were calculated, and any weight change during sintering was noted. These data were then transmitted to the statistician for analysis.

Laboratory Experiments

Because of the restricted investigation in the pilot plant, the tests were made in laboratory equipment. It was hoped that these experiments would be less time-consuming, and that if there was no significant difference between them and the plant tests, the full statistical process investigation might be carried out in the laboratory.

The experimental procedure was nearly identical to that used in the pilot plant tests; the only variation was the reduced batch size and gas flow rates (1.36 liters of feed solution, 0.018 cfm NH_3). The geometry of the tall-pipe precipitator was duplicated in the laboratory by using precipitators fabricated from 2-in. stainless steel pipe. These were wrapped with Nichrome heating coils and asbestos tape. A single $1/4$ -in. gas-dispersing tube was used in each vessel. This apparatus permitted all four tests to be made simultaneously and, therefore, ensured similar test conditions. The product slurry was removed through a suction dip leg and filtered on a leaf filter with Nylon 518F cloth as the filter medium. This apparatus is shown in Figure 38. Samples from each test were dried, screened, calcined, pressed, and sintered at the same time the samples from the pilot plant were processed.

Results and Conclusions

The experimental conditions of the pilot plant and laboratory runs are shown in Tables 33 and 34. The green and sintered densities of the oxide compacts are given in Tables 35 and 36. Analysis of these data,

FIGURE 38. LABORATORY PRECIPITATION APPARATUS

TABLE 33. Experimental Conditions - Pilot Plant
Precipitation Tests

Run Number	1	2	3	4	5
Feed volume, liters	33	33	33	33	33
Feed concentrations:					
U, $\underline{M}$	0.442	0.487	0.466	0.521	0.469
H ⁺ , $\underline{M}$	0.596	0.606	0.543	0.526	0.575
Average temperature, °C	51	50	47	47	46
Ammonia rate, cfm	0.26	0.26	0.26	0.26	0.26
Nitrogen rate, cfm	1.17	1.20	1.20	1.20	1.20
Time to completion,* min	150	235	180	250	180

*Time to completion is defined as the time required to reach a pH of 9.5.

TABLE 34. Experimental Conditions - Laboratory
Precipitation Tests

Run number	1-I, 1-II, 1-III, 1-IV	2-I, 2-II, 2-III, 2-IV	3-I, 3-II, 3-III, 3-IV
Feed volume, liters	3	1.36	1.36
Feed concentrations			
U, $\underline{M}$	0.479	0.416	0.480
H ⁺ , $\underline{M}$	0.602	0.568	0.600
Average temperature, °C	48	47	47
Ammonia rate, cfm	0.024	0.018	0.011
Nitrogen rate, cfm	Constant, but not measured	0.12	0.14
Average time to completion,* min.	160	110	175

*Time to completion is defined as the time required to reach a pH of 9.5.

TABLE 35. Compact Densities - Pilot Plant Tests

<u>Sample Number</u>	<u>Green Density, gm/cc</u>	<u>Sintered Density, gm/cc</u>
PS1-EF	5.59	9.33
PS2-EK	5.48	9.51
PS2-LF	5.53	9.18
PS3-EF	5.14	9.34
PS3-LF	5.26	9.37
PS4-EF	5.59	9.66
PS4-LF	5.14	9.33
PS5-EF	5.47	10.07
PS5-LF	4.50	9.14
Average = 5.30		Average = 9.44
$s_g = 0.35$		$s_s = 0.28$

TABLE 36. Compact Densities - Laboratory Tests

<u>Sample Number</u>	<u>Green Density, gm/cc</u>	<u>Sintered Density, gm/cc</u>
LS1-I	Broken sample	Broken sample
LS1-II	5.10	10.14
LS1-III	5.62	10.30
LS1-IV	5.42	9.88
LS2-I	5.56	8.69
LS2-II	5.50	8.55
LS2-III	5.10	8.24
LS2-IV	5.59	8.74
LS3-I	5.30	9.48
LS3-II	5.17	9.18
LS3-III	5.51	9.34
LS3-IV	5.18	8.63
Average = 5.37		Average = 9.20
$s_g = 0.20$		$s_s = 0.69$

KAPL-1757

the observations made during the course of the tests, and the detailed reports of the limited statistical analysis of the raw data lead to the following conclusions

1. Satisfactory operation of the precipitator and filter was demonstrated, but exact plant operating conditions were not established since all the tests were made at the mid-points of the variable ranges rather than at the anticipated plant conditions, which lie nearer the range endpoints.
2. Dimensional specifications which will satisfy conditions of geometrical criticality must be established for the filter drum and feed pan, and a means of remotely changing the filter medium must be developed.
3. Variations in the sintered compact densities including those variations introduced by the analytical technique are fairly large. This is indicated by a standard deviation of 0.28 (about a mean of 9.44 gm/cc) in the pilot plant experiments and by a standard deviation of 0.69 (about a mean of 9.20 gm/cc) in the laboratory runs. This variation is larger than can be tolerated in the operating plant.
4. A comparison of variations within each experiment and between the two experiments shows that a considerable part of the over-all variability may be due to differences introduced by the sample preparation technique; that is, the calcination, sizing, compaction, and sintering operations. Since all samples were calcined and sintered simultaneously, it was thought that the variation in green density, which was in a sense an independent variable, might be responsible for the scatter in the final density values. Cross plotting the green and sintered density values, as shown in Figures 39 and 40, indicates that there is no over-all correlation between the green and sintered densities.* This lack of

*Since there was very limited within-run replication in the experiment, little evidence can be obtained for the existence of such a correlation for a single precipitated, calcined, and sized batch of oxide. In the case of the laboratory runs, some evidence of a within-run correlation can be seen in Figure 40. It is obvious that unless such a correlation exists, the use of a "shrinkage factor" (that is, the ratio of the change in density to sintered density) to predict final density has no significance. Further highly replicated tests are required to determine if such a correlation exists.

FIGURE 39 SINTERED DENSITY vs. GREEN DENSITY
PLANT SCALE TESTS FIGURE 40 SINTERED DENSITY vs. GREEN DENSITY
LABORATORY TESTS

strong correlation indicates that variations in the sintered densities are not directly attributable to the variations in green densities. This does not permit the conclusion, however, that the compaction operation does not contribute to the variability of sintered density. Since a rather weak correlation in the laboratory runs is indicated, some undetermined part of the variability may be attributed to real process differences between laboratory and plant runs.

5. It is recommended that the precipitation process investigation be discontinued until the precision of the compacting and sintering technique can be investigated. This can be done on a laboratory scale with a single batch of UO_2 prepared by a typical precipitation procedure. Because variations in the laboratory runs were greater than the variations in the pilot plant runs, it is also recommended that all further process studies be made on the plant scale.

Fuel Element Fabrication

An investigation of the proposed extrusion-fabrication technique was conducted in KAPL facilities to gain an understanding of the mechanical equipment requirements necessary for the design of a remotely operated plant-scale extruder, to obtain an estimate of the dimensional reproducibility of the product, and to determine what process variables were significant. This investigation also provided feed material for the pilot plant dissolver demonstration previously described.

Equipment

The principal piece of apparatus used in this investigation is the hydraulic extrusion chamber and product take-up device shown in Figure 41. Standard batch-type laboratory equipment was used to prepare the oxide and to blend it with water and an organic binder to form a clay for extrusion. The rods were dried in a humidity cabinet in which the trays of extruded rods were positioned above a pan of water. A standard molybdenum-wound hydrogen atmosphere furnace with 4-in.-diam muffle was used for oxide conversion and sintering.

Experimental Procedure

A mixed uranium-thorium dioxide was used in these experiments. This was prepared in the laboratory in 1000-gm batches by bubbling ammonia gas through a nitrate solution with the metallic ions in the desired atomic ratio. The precipitate was filtered, dried, and converted to the mixed oxide by calcining the material for 1 hr at 700°C in the hydrogen furnace. A total of five batches was prepared in this manner with an attempt to keep the preparative conditions constant.

Each batch of oxide was milled in a Hobart mixer until all material passed through a 50-mesh screen. One-half of each batch was then returned to the mixer and milled twice as long as had been required to reduce it to less than 50-mesh size. This was done to see if the increased product surface area would have an effect on the sintering characteristics of the extruded piece. These sub-batches were then blended with 1 or 2 wt % Methocel binder and about 10 wt % water to form a clay for extrusion. The amount of water added varied from sample to sample because water was added until the clay had the proper consistency for extrusion. This clay was then extruded as 0.25-in.-diam rods about 30 in. long and placed in the humidity cabinet to dry. The ends of the rod usually warped badly during drying; therefore, ~3 in. were discarded from each end, and the remaining length was broken into four 6-in. lengths. The diameter, density, and position of each of these pieces within the green rod were determined and recorded. When all batches had been carried through this step,

FIGURE 41. HYDRAULIC EXTRUSION EQUIPMENT

KS-20165
Unclassified

specimens from each batch were selected according to a statistical plan for sintering at either 1500 or 1700°C for either 1 or 3 hr. After the batches were sintered, the densities and diameters were again determined and recorded. These data were then analyzed statistically to determine the significance of the various process steps.

Results and Conclusions

Preparation of 128 six-inch extruded and sintered oxide fuel specimens during the course of this experiment resulted in a satisfactory demonstration of the proposed extrusion-fabrication process. The plant extruder design discussed on page 37 is based on the mechanical analysis of the operation of the extruder during this experiment.

A complete tabulation of the raw data is too bulky for inclusion in this report, but the following results were obtained by statistical analysis of the data.

1. The process variability is rather large, as is indicated by an estimated sintered density standard deviation of 0.37 (about a mean of 9.66 gm/cc).
2. The greatest part of this variability is attributed to variations among the several oxide batches resulting from separate precipitation and calcination operations.
3. The effects of the process variables - grinding time, drying humidity, sintering temperature, and sintering time - and their interactions were not found to have a significant effect on either the sintered density or sintered diameter when compared with the large batch variability effect.
4. It was surprising to find that neither sintering temperature nor sintering time is a significant variable. This can be explained by the high densities that were obtained; that is, the oxide was so "active" that the samples sintered beyond the point where the density changes rapidly with time for the sintering temperatures used in this investigation.

REFERENCES

1. Davidson, J. K., "The Fast Oxide Breeder - A Summary," KAPL-1701, July 1, 1957.
2. Davidson, J. K., "The Reloading Cycle for a Power Reactor," KAPL-M-JKD-11, March 19, 1957.
3. Coplan, B. V., Davidson, J. K., and Zebroski, E. L., "The Pump-Mix Mixer Settler, A New Liquid-Liquid Extractor," Chem. Eng. Progr. 50 (August, 1954), pp. 403-408.
4. Davidson, J. K., Schafer, A. C., Jr., and Haas, W. O., Jr., "Belgian Symposium on Chemical Processing II Session; Aqueous Reprocessing, Application of Mixer-Settlers to the Purex Process," KAPL-1809, May 8, 1957.
5. Lang, H. J., "Cost Estimates," Chem. Eng. September, 1947, pp. 130-133; October 1947, pp. 117-121; June 1948, pp. 112-113.
6. Chilton, C. H., "Cost Data Correlated," Chem. Eng. June 1949, pp. 97-106.
7. Cashin, W. M., "Evaluation of an Oxide Breeder Fuel Element," KAPL-1446, November 18, 1955.
8. "Report of the Chemistry and Chemical Engineering Section for August, September, October, 1955," KAPL-1438.
9. Coplan, B. V. and Smith, D. J., "Remodeling a High Level Chemistry Cell," Nucleonics 12 (November, 1954), pp. 92-93.
10. Alter, H. W., Coddling, J. W., and Jennings, A. S., "Miniature Mixer Settlers for Continuous Counter-Current Solvent Extraction." Anal. Chem. 26 (August, 1954), p. 1357.
11. Miller, R. S. and Smith, D. J., "A Facility for High Level Plutonium Extraction Studies," Proceedings of the 1957 Nuclear Congress, Vol III "Hot Laboratory Operation and Maintenance," Pergamon Press, 1957.
12. Jones, S. S., "The Fast Oxide Breeder - Radiation Effects," KAPL-1758, July 1, 1957.
13. Coddling, J. W., Haas, W. O., Jr., and Heumann, F. K., "Tributyl Phosphate - Hydrocarbon Systems, Organizing Equilibrium Data," Ind. Eng. Chem. 50 (February 1958), p. 145.

NOTE: A bibliography of the KAPL topical reports on the Fast Oxide Breeder program can be found in KAPL-1701, "The Fast Oxide Breeder - A Summary," J. K. Davidson, July 1, 1957, pages 28-29.

APPENDIX

CALCULATION OF NORMAL CORE AND BLANKET PROCESSING RATES

The core and blanket processing rates can be estimated from the core power, the internal and external conversion ratios, α_{Pu} , the burnup, the plutonium concentration in the discharged blanket, and certain of the core volume fractions as follows.

If the flux is assumed to be constant with time and if only one "group" is assumed to describe the neutrons, then we find,

$$N_{49} = N_{28} \left(\frac{\sigma_c}{\sigma_{a49}} \right) \left(1 - e^{-\phi \sigma_{a49} t} \right) + N_{490} e^{-\phi \sigma_{a49} t} \quad (1)$$

where N_{49} = the number of Pu atoms/cc at time t

N_{490} = the number of Pu atoms/cc at time $t = 0$, and

N_{28} = the number of U^{238} atoms/cc (assumed constant).

The total number of plutonium fissions per cubic centimeter, F , occurring in time t is given by

$$F = \int_0^t \phi \sigma_{f49} N_{49} dt; \quad (2)$$

but since ϕ is assumed to be constant,

$$F = \phi \sigma_{f49} \int_0^t N_{49} dt \quad (3)$$

and

$$\begin{aligned} F = N_{28} \frac{\sigma_c}{\sigma_{a49}} \phi \sigma_{f49} t - N_{28} \frac{\sigma_c}{\sigma_{a49}} \left(\frac{\sigma_{f49}}{\sigma_{a49}} \right) \left(1 - e^{-\phi \sigma_{a49} t} \right) \\ + N_{490} \left(\frac{\sigma_{f49}}{\sigma_{a49}} \right) \left(1 - e^{-\phi \sigma_{a49} t} \right), \end{aligned} \quad (4)$$

or the total amount of fuel fissioned stated as a fraction of the original plutonium charged is given by

$$\frac{F}{N_{49O}} = \left(\frac{N_{28} \sigma_{c28}}{N_{49O} \sigma_{a49}} \right) \left(\frac{\sigma_{f49}}{\sigma_{a49}} \right) (\phi \sigma_{a49} t) - \left(\frac{N_{28} \sigma_{c28}}{N_{49O} \sigma_{a49}} \right) \left(\frac{\sigma_{f49}}{\sigma_{a49}} \right) \left(1 - e^{-\phi \sigma_{a49} t} \right) + \left(\frac{\sigma_{f49}}{\sigma_{a49}} \right) \left(1 - e^{-\phi \sigma_{a49} t} \right). \quad (5)$$

If the following definitions are made:

$$\text{burnup} \equiv B \equiv 1 - e^{-\phi \sigma_{a49} t} \quad (6)$$

$$\text{or} \quad \ln \frac{1}{1-B} = \phi \sigma_{a49} t \quad (7)$$

$$\text{initial internal conversion ratio} \equiv \text{ICR} \equiv \frac{N_{28} \sigma_{c28}}{N_{49O} \sigma_{a49}} \quad (8)$$

$$\text{and since} \quad \frac{\sigma_c}{\sigma_f} = \frac{1}{1+\alpha}, \quad (9)$$

Equation (5) can be rewritten

$$\frac{F}{N_{49O}} = (\text{ICR}) \left(\frac{1}{1+\alpha_{49}} \right) \ln \left(\frac{1}{1-B} \right) - (\text{ICR}) \left(\frac{1}{1+\alpha_{49}} \right) B + \left(\frac{1}{1+\alpha_{49}} \right) B \quad (10)$$

$$\frac{F}{N_{49O}} = (\text{ICR}) \left(\frac{1}{1+\alpha_{49}} \right) \ln \left(\frac{1}{1-B} \right) + B \left(\frac{1}{1+\alpha_{49}} \right) (1 - \text{ICR}). \quad (11)$$

Equation (1) can be rearranged to obtain a similar expression for the amount of plutonium present in the fuel at time t ; the basis of the fraction again is the original plutonium charged:

$$\frac{N_{49}}{N_{49O}} = \left(\frac{N_{28} \sigma_{c28}}{N_{49O} \sigma_{a49}} \right) \left(1 - e^{-\phi \sigma_{a49} t} \right) + e^{-\phi \sigma_{a49} t} \quad (12)$$

$$\frac{N_{49}}{N_{49O}} = (\text{ICR}) B + (1-B). \quad (13)$$

Equations (11) and (13) can now be evaluated to give the daily core reprocessing rates. Since ~1 MWD of heat is released in fissioning 1 gm of fuel, a core power of 660 Mw will require the fissioning of 660 gm of plutonium per day.

On the basis of 300-day per year reactor operation and 250-day per year reprocessing plant operation, the fission rate on the plant basis will be

$$660 \text{ gm/day} \times \frac{300}{250} = 792 \text{ gm Pu/day.} \quad (14)$$

For $B = 0.5$
 $ICR = 0.5$, and
 $\alpha_{49} = 0.2$,

the daily feed rate of discharged fuel to the reprocessing plant is found as follows:

$$\frac{F}{N_{49O}} = (0.5) \left(\frac{1}{1+0.2} \right) \ln \left(\frac{1}{1-0.5} \right) + (0.5) \left(\frac{1}{1+0.2} \right) (1-0.5) = 0.497 \quad (15)$$

$$N_{49O} = \frac{F}{0.497} \quad (16)$$

$$\frac{N_{49}}{N_{49O}} = (0.5)(0.5) + (1-0.5) = 0.75 \quad (17)$$

$$N_{49} = 0.75 N_{49O} = (0.75) \left(\frac{F}{0.497} \right) = 1.51F \quad (18)$$

$$(N_{49})_{Pu} = (1.51)(0.792) = 1.20 \text{ kg Pu/day} \quad (19)$$

$$(N_{49})_{PuO_2} = 1.20 \left(\frac{271}{239} \right) = 1.36 \text{ kg PuO}_2/\text{day.} \quad (20)$$

The oxide fission-product feed rate, FP, is simply the fission rate corrected for the difference in reactor and plant operating time:

$$FP = 0.792 \left(\frac{271}{239} \right) = 0.90 \text{ kg FP oxides/day.} \quad (21)$$

If the original uranium-to-plutonium ratio is 4.6 to 1, the uranium discharge rate is obtained from

$$\left(N_{49O} \right)_{PuO_2} = \frac{N_{49}}{0.75} = \frac{1.36}{0.75} = 1.81 \text{ kg } PuO_2/\text{day} \quad (22)$$

$$N_{28O} = 4.6 N_{49O} \quad (23)$$

$$\left(N_{28O} \right)_{UO_2} = (4.6)(1.81) = 8.33 \text{ kg } UO_2/\text{day} \quad (24)$$

$$N_{28O} + N_{49O} = N_{28} + N_{49} + N_{FP} \quad (25)$$

$$(N_{28})_{UO_2} = 8.33 + 1.81 - 1.36 - 0.90 \quad (26)$$

$$(N_{28})_{UO_2} = 7.88 \text{ kg } UO_2/\text{day} . \quad (27)$$

The stainless steel cladding from the core processed per day can be calculated from the core volume fractions and an assumed fraction of core iron present as cladding. The core volume fractions are:

$$VF_{UO_2} + VF_{PuO_2} = 0.26 \quad (28)$$

$$VF_{SS} = 0.16 \quad (29)$$

and

$$\rho_{PuO_2} = 11.0 \text{ gm/cm}^3 \quad (30)$$

$$\rho_{SS} = 7.9 \text{ gm/cm}^3 . \quad (31)$$

If we assume that 50% of the core iron is cladding, we find

$$W_{SS} = 0.5 \rho_{SS} V_{SS} \quad (32)$$

$$W(PuO_2 + UO_2) = \rho(UO_2 + PuO_2) V(UO_2 + PuO_2) \quad (33)$$

$$W_{SS}W(UO_2 + PuO_2) = \frac{0.5\rho_{SS}V_{SS}}{(\rho V)(UO_2 + PuO_2)} \quad (34)$$

$$W_{SS} = \frac{(0.5)(7.9)(0.16)}{(11.0)(0.26)} \times 10.14 \text{ kg/day}$$

$$= 2.24 \text{ kg SS/day} . \quad (35)$$

The blanket fuel processing rate is now calculated on the basis of a discharge concentration of 5000 gm Pu/ton U and an external conversion ratio of 0.9 as follows:

$$\text{destructions in core} = \text{fissions in core} \times (1 + \alpha_{49}) \quad (36)$$

$$P = \text{plutonium formed in blanket} = \text{EBR} \times \text{destructions in core} \quad (37)$$

$$P = (1 + 0.2)(0.792)(0.9) = 0.855 \text{ kg Pu/day} \quad (38)$$

$$U = \frac{\text{gm Pu/day}}{\text{gm Pu/ton U}} = \frac{855}{5000} = 0.171 \text{ tons U/day} . \quad (39)$$

The blanket cladding processing rate (of an element whose diameter is 0.75 in. and cladding thickness is 0.030 in.) is found as follows:

$$V_U = \frac{\pi}{4} \left(\frac{3}{4}\right)^2 L \quad (40)$$

$$V_{SS} \approx \pi \left(\frac{3}{4}\right) (0.030) L \quad (41)$$

$$\frac{V_{SS}}{V_U} = 4 \left(\frac{4}{3}\right) (0.030) = 0.16 \quad (42)$$

$$\frac{W_{SS}}{W_U} = \frac{V_{SS}\rho_{SS}}{V_U\rho_U} = (0.16) \left(\frac{7.9}{18.9}\right) = 0.067 \text{ gm SS/gm U} \quad (43)$$

$$W_{SS}/\text{day} = (1.71)(0.067) = 11.45 \text{ kg SS/day} . \quad (44)$$

A summary of the rates is shown below.

<u>Core</u>	<u>kg/day</u>
PuO ₂	1.36
UO ₂	7.88
Fission-product oxides	0.90
Stainless steel	2.24
<u>Blanket</u>	
Uranium	171.0
Plutonium	0.855
Stainless steel	11.45