MASTER

GEAP-3876

**PLUTONIUM FUEL FABRICATION AND
REPROCESSING FOR FAST CERAMIC
REACTORS**

By
E. L. Zebroski
H. W. Alter
G. D. Collins

February 1, 1962

Atomic Power Equipment Department
General Electric Company
San Jose, California

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.

This report has been reproduced directly from the best available copy.

Printed in USA. Price \$1.50. Available from the Office of Technical Services, Department of Commerce, Washington 25, D. C.

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.

PLUTONIUM FUEL FABRICATION
AND
REPROCESSING FOR FAST CERAMIC REACTORS

Prepared for the
UNITED STATES ATOMIC ENERGY COMMISSION

under

Contract AT(04-3)-189, P.A. #10
General Electric Requisition #474-30750

By

E. L. Zebroski

H. W. Alter

G. D. Collins

February 1, 1962

APPROVED:

K. P. Cohen, Project Engineer
Fast Ceramic Reactor Program

.

.

.

.

.

PLUTONIUM FUEL FABRICATION AND REPROCESSING FOR FAST CERAMIC REACTORS

E. L. Zebroski, H. W. Alter, G. D. Collins

TABLE OF CONTENTS

	<u>Page</u>
I. <u>FAST REACTOR FUEL CYCLE</u>	3
A. Fuel Materials	3
B. Fuel and Blanket Time-Cycle	3
II. <u>RADIOACTIVITY OF RECYCLED PLUTONIUM FROM FAST REACTORS</u>	6
A. Isotopic Dose-Rates from Plutonium	6
B. Dose from Recycled Plutonium	9
C. Alpha-Neutron Reactions and Plutonium-238	10
D. Effect of Plutonium-238 Buildup	10
III. <u>CHEMICAL PROCESSING</u>	15
A. Relation of Residual Plutonium Radiations to Selection of Reprocessing Method	15
B. Gross Fission Product Activity of Irradiated Fuel	17
C. Decontamination Factors Required to Permit Direct Handling	17
D. The Effect of Long Irradiations and Long-Lived Fission Products	20
E. Ion Exchange Processes	22
F. Solvent Extraction Process for Fast Reactor Fuel	24
G. Recommended Reference Process	25
H. Fuel Throughputs and Plant Capacities	26
I. Reprocessing Economics	30
IV. <u>FABRICATION PROCESS FOR OXIDE FUEL</u>	31
A. Choice of the Preferred Process	31
B. Blending	33
C. Precipitation of Mixed Oxides	33
D. Alternate Precipitation Method for Plutonium Alone	33
E. Drying and Calcining	34
F. Pressing and Sintering of $\text{PuO}_2\text{-UO}_2$	34
G. Swaging over Pellets	34
H. Assembly and Quality Control	35
I. Fabrication Costs	35
J. Fuel Fabrication Volumes Due to Growth of Capacity	36
V. <u>TOTAL FUEL CYCLE COSTS</u>	40
A. Basis for Elements of Total Fuel Cycle Costs	40
<u>APPENDIX A</u> - Anion Exchange Flowsheet	43
<u>APPENDIX B</u> - Outline of Development Programs for FCR Fuel Fabrication and Reprocessing	50

-

-

-

-

PLUTONIUM FUEL FABRICATION AND REPROCESSING FOR FAST CERAMIC REACTORS

E. L. Zebroski, H. W. Alter, G. D. Collins

A B S T R A C T

A study has been made of the processes available for fabrication of plutonium-containing fuel from a fast ceramic reactor, and for chemical reprocessing of irradiated fuel.

Radiations from recycled plutonium are evaluated. Adaptation of conventional glove-box handling procedures to the fabrication of recycle plutonium appears practical.

It is concluded that acceptable costs are obtainable using moderate extensions of conventional glove-box fabrication methods and wet processing techniques, provided a significant volume of production is available. The minimum economic scale for the preferred chemical reprocessing method, anion exchange, is about 500 MWe of reactor capacity.

The minimum scale of economic operation for the fuel refabrication facility corresponds to three 500 MWe reactors, if only steady-state refueling provides the fabrication load. The minimum volume required falls to one 500 MWe reactor, if the continued growth of capacity provides fabrication volume equal to that for refueling.

The chemical reprocessing costs obtained range from 0.27 mills/kwhr for 1,500 MWe of reactor capacity, to 0.10 mills/kwhr for 3,000 MWe of capacity.

The estimated fuel fabrication cost is \$211/kg of uranium and plutonium in the core region (excluding axial and radial blankets) or \$1.06/g of plutonium content. When axial blankets, fabricated in the same rods, are included; the combined average is \$134/kg of uranium and plutonium. Radial blanket fabrication cost is \$49/kg of uranium. The overall average of all fuel and blankets is \$84/kg of uranium and plutonium. The fabrication cost is 0.29 mills/kwhr for a production rate corresponding to 3,000 MWe of capacity (or 1,500 MWe of capacity plus growth equivalent to one additional reactor core per year). For one 525 MWe reactor, (plus equivalent growth volume) the fabrication cost becomes 0.42 mills/kwhr. (All fuel throughputs are based on fuel life of 100,000 MWD/T.)

Using the estimates developed here, the total fuel cycle cost for a typical fast reactor design using $\text{PuO}_2\text{-UO}_2$ fuel is estimated to be about 0.9 mills/kwhr.*

Unconventional processes, including "dry" methods and remote refabrication may have interesting economic potential and warrant further evaluation. The estimated costs of the reference processes are sufficiently low, however, that the development of novel methods does not appear essential to the economic viability of the fast reactor with ceramic fuel.

The developmental problems in the reference processes are mainly in the nature of engineering demonstration, and in obtaining data for optimizing large scale process design with respect to unit costs. It appears to be feasible to resolve these questions within the time required for reduction-to-practice of the design of a large scale fast reactor using ceramic fuels (about five years).

* Assuming fabrication and processing corresponding to 3,150 MWe of capacity, 10% inventory charges, and \$6.50/gm fissile plutonium inventory value. (1975 Projection)

I N T R O D U C T I O N

The objectives of this study were to:

1. Explore several alternative fuel fabrication and chemical reprocessing methods to recycle plutonium fuel in a fast ceramic fueled reactor and select reference processes.
2. Estimate fabrication and reprocessing costs for the reference processes.
3. Estimate the development required for reference processes and the possible scope for cost savings through the use of novel methods.

In selecting processes for study, it is noted that with a long-lived fuel (several years) there is no compelling reason for requiring processes which can give very short time cycles for recovery or fabrication of fuel.

Typical fast ceramic fuel reactor designs, using either oxide or carbide fuels, permit fuel lifetimes in the reactor of the order of three years. For example, the present reference design for oxide fuel has a specific power of 1,000 kw/kg of fissile isotopes and average irradiation levels of 100,000 MWD/T in the discharge fuel. These levels of performance have already been demonstrated on a small scale with oxide fuels, and are probably also attainable with carbide fuels. (Such lifetimes might also be attained by metal, alloy or cermet fuels with strong cladding).*

The estimated times required for reprocessing and refabrication of ceramic fuel are: one month cooling; one month for shipping and reprocessing; four months for refabrication and return shipping of the fuel.

A total out-of-pile cycle of about six months appears attainable using conventional wet reprocessing methods, either solvent extraction or ion exchange. These processes provide high decontamination factors so that the fission-product activity of the recovered material is negligible. The commitment of one-sixth of the amount of the in-pile inventory to the external fuel "pipeline" is an economically minor addition to the total fuel inventory costs. If subsequent improvements in fast reactor design permit doubling the specific power of the in-core inventory, there appears to be reasonably good prospects of reducing the out-of-pile time to about three months (especially with relatively large-volume operations) so that the same relationship of total inventory to in-reactor inventory could be preserved.

From the foregoing, it is evident that for long-lived fuels there is no compelling economic or technical need for processes which permit very short time for recycle of fuel. The selection, design, and costing of "reference" reprocessing and fuel fabrication processes accordingly have been based on modest extensions of conventional technology in order to obtain reliable cost estimates.

* L. R. Blake, REACTOR SCIENCE AND TECHNOLOGY, Journal of Nuclear Energy, Parts A/B, 14, 31 (1961).

Within the limits of the uncertainties listed elsewhere in this report, the cost estimates serve to provide reference costs for the fast reactor fuel cycle, and also as a target level from which to measure the incentives or cost-saving potential for process improvements or alternate methods of reprocessing and fuel fabrication. For the processes described, reprocessing and fabrication costs, each of the order of 0.3 mills/kwhr, have been estimated (and lower at large volume of operation). These lead to overall fuel cycle costs of slightly less than 0.9 mill/kwhr for a fast reactor with oxide fuel. Generally similar fuel cycle costs are believed to be possible with carbide fuels.*

In order to attain tolerable fuel inventory costs in reactor systems with short-lived fuels, decontamination and fabrication processes are needed which can function at extremely short cooling times - and which may leave considerable residual fission-product activity. This has focussed attention on remote handling methods for refabrication of fuel. Once remote fabrication is accepted, there is less disadvantage in considering chemical separation processes which yield small decontamination factors. From a neutron loss standpoint, a decontamination factor of 100 (1% of fission products retained with the fuel) is entirely adequate.

If remote fabrication were essential because of inherent radioactivity levels of recycled plutonium, the incentive for high decontamination factor chemical processing methods is also considerably lessened. It is therefore important to analyze the radiations from recycled plutonium in some detail to determine the adaptability of conventional glove-box handling techniques to the specific radiation levels to be encountered. (See Section II.) If glove-box handling is to be practical, the fission-product activity associated with recovered plutonium should also be reduced to levels which are negligible for handling purposes. This requires fission-product decontamination factors near 10^6 , which are at present attainable only by wet processes, solvent extraction or ion exchange.

Finally, the use of "wet" processes with metal fuels, carries the cost burden of the reduction steps from the nitrate to the metal (\$1 to \$2/gram for plutonium). This provides an incentive for "dry" processes which can avoid or cheapen the reduction step. With the mixed oxide fuel the cost of conversion from nitrate to oxide is about 20¢/g of plutonium (see Section IV, Figure IV-2) which tends to eliminate this cost as a significant incentive for dry processing with oxide fuels.

* K. M. Horst, B. A. Hutchins, "Comparative Study of PuC-UC and PuO₂-UO₂ as Fast Reactor Fuel", GEAP-3880-AEC, January 19, 1962.

I. FAST CERAMIC REACTOR FUEL CYCLE

A. Fuel Materials

The fast ceramic fueled reactor is fueled with a mixture of uranium and plutonium in the form of the mixed oxides or carbides of these elements. For this study a reference design has been used which uses a mixture of uranium oxide and plutonium oxide at approximately 4 to 1 atom ratio of uranium-to-plutonium. Generally similar performance, from the standpoint of fuel fabrication and re-processing economics, is believed to be potentially also attainable with carbide fuels. An evaluation of the relative merits of oxide or carbide fuels for fast reactors and their cost relationship, are subjects of a concurrent study.*

For the current reference design of the FCR, "reactor-grade" plutonium is used to provide the initial inventory of the reactor. Such plutonium (typically from a BWR, PWR or other thermal reactors) contains substantial amounts of plutonium-240 and plutonium-241. The blanket of the fast reactor initially contains mainly uranium-238. Depleted uranium (approximately 0.3% uranium-235) is preferred because of its low cost.

For the reference design size, (500 MWe) the total breeding ratio is 1.08. Of this, 0.66 occurs in the core of the reactor. The remaining plutonium generation occurs in the axial blanket (0.24) and in the radial blanket (0.18).

The best economy of the fast reactor would probably be reached in unit sizes near 1,000 MWe. For such reactors, the total breeding ratio would be slightly higher, and the fraction of the total breeding ratio which was internal to the reactor core would increase somewhat relative to the blanket.

B. Fuel and Blanket Time-Cycle

The fuel irradiation lifetime which appears practical from both the material standpoint, and a reactivity-lifetime standpoint for a large fast reactor, is an average of 100,000 MWD/T in the fuel. Preliminary irradiation tests (capsules) have shown good dimensional stability and no failures up to 100,000 MWD/T with mixed uranium-plutonium oxides.**

The axial blanket material is integral with the core fuel and therefore is removed from the reactor on the same schedule as the core fuel. The axial blanket reaches an average exposure of 12,200 MWD/T in the time required by the core fuel to reach an average level of 100,000 MWD/T. The radial blanket, which is subject to a considerably lower flux, could be replaced at a different rate than the core fuel. If 7% of the fuel is removed for each reloading, (70 day intervals at

* K. M. Horst, B. A. Hutchins, "Comparative Study of PuC-UC and PuO₂-UO₂ as Fast Reactor Fuel", GEAP-3880-AEC, January (1962).

** J. M. Gerhart, "Post-Irradiation Examination of PuO₂-UO₂ Fuel for Fast Reactor Application", GEAP-3833-AEC, November (1961).

TABLE I-1
REFERENCE FUEL PARAMETERS

		Blanket	
	Core	Top & Bottom Axial	Radial
I. <u>Dimensions</u>			
O.D., ft.	6.5	6.5	9.0
Overall length, ft.	3.25	5.75	4.5
Pins/Assembly	127	127	-
Pin Dimensions			
Fuel diameter, in.	.220	.220	.420
Clad O.D., in.	.250	.250	.450
Clad Thickness, in.	.015	.015	.015
II. <u>Fuel Material</u>			
Density (oxide) g/cc	10.0	-	-
Wt. % Pu	Initial	0	0
	Final	3.0	0.81
(U + Pu) g/pin	239	-	-
(U + Pu) kg/assembly	30.4	-	-
Pu g/pin	45.9	-	-
Pu kg/assembly	5.84	-	-
III. <u>Time Cycle (80% Capacity Factor)</u>			
MWD/T	100,000	12,200	3,050
Average time in core, days	1,000	1,000	1,000
Refueling interval, days (7% of core & blanket)	70	70	70
Annual fuel throughput* (kg/yr of U + Pu + FP)	3,230	2,485	7,090
Pu throughput, kg/yr	545	75	57

* See Tables III-3, III-4, and III-5 for more detailed breakdowns of fuel throughputs and compositions.

80% capacity factor) and if a similar fraction of the radial blanket is removed at the same time, the average irradiation of the rods discharged from the blanket is 3,050 MWD/T. For the low irradiation levels in the blanket regions, plutonium-239 is generated with only small amounts of higher isotopes. The axial blanket would be expected to contain approximately 1.2% plutonium-240, and .08% plutonium-241; the radial blanket would contain approximately .26% plutonium-240 and .02% plutonium-241. Upon recycling the plutonium, it is assumed that the core and the blankets are combined for reprocessing. For sufficiently large-scale operations, there might be some advantage in segregating the core plutonium with the higher content of non-fissile isotopes, from the plutonium in the radial and axial blankets. There may also be a small economic incentive in optimizing a design and replacement schedule for the radial blanket. For the present analysis, the same replacement rate was taken for the core, axial and radial blankets.

Table I-1 lists the reference fuel parameters* important to chemical processing and fabrication of fuel. Isotopic compositions and radiations from recovered fuel are given in Section II.

* K. M. Horst, B. A. Hutchins, F. J. Leitz, B. Wolfe, "Design Study of a 500 MW Fast Oxide Reactor", GEAP-3721-AEC, December(1961).

II. THE RADIOACTIVITY OF RECYCLED PLUTONIUM FROM FAST REACTORS

A fundamental question in the selection and design of refabrication processes for plutonium-containing fuels is the radiation dose which will be emitted from recycled fuel which contains large amounts of higher isotopes of plutonium. Even the initial fuel, obtained from thermal reactors at 10,000 to 20,000 MWD/T, has large percentages of higher isotopes. A review of the data on the decay schemes and the observed specific activities of plutonium isotopes leads to the conclusion that routine handling of highly irradiated plutonium is feasible using adaptations of the usual glove-box procedures. Fabrication steps such as powder blending, cold pressing and sintering, or vibratory compaction of mixed oxide (or carbide) powders, or cold swaging over powder or pellets, and usual types of end-closure welding procedures all appear feasible. However, operations such as grinding pellets appear seriously less desirable (or more costly) and the assembly procedures must be designed to avoid more than occasional hand contact with the assembly.

Added precautions such as localized shielding and restricted time for direct glove contact are required. The primary consideration in handling, especially prior to encapsulation, is the protection required against the alpha inhalation hazard of the plutonium, but the equipment procedures required are essentially the same as those normally used with low-irradiation plutonium.

One reason for the relatively favorable situation, despite the additional gammas, x-rays and neutrons emitted from highly-irradiated plutonium, is that nearly all of the intense activities are relatively low in penetrating power. They are strongly self-absorbed in plutonium or plutonium-uranium mixtures, and the surface dose can be substantially reduced by modest amounts of shielding. For example, the neoprene gloves with lead or zinc loading, which are used in dry-box handling of plutonium, serve to stop most of the soft gamma and x-ray radiations as well as alphas and betas, from plutonium-238, plutonium-241, plutonium-240, plutonium-239, americium-241 and uranium-237.

The spontaneous neutrons from plutonium-240 and plutonium-238, and the oxygen or carbon alpha-n neutrons from mixtures of plutonium and uranium oxide or carbide, are the predominant penetrating radiations. Plutonium-240 x-rays limit contact handling. (See table on page 11.) The penetrating neutron radiation is significant for handling considerable masses of plutonium, such as a fuel subassembly. For smaller quantities, such as one or a few fuel pins, contact times of the order of one working day per week are possible.

A. Isotopic Dose-Rates from Plutonium

The contribution of activities from various plutonium isotopes from plutonium-238 to plutonium-242 are summarized in Table II-I and II-1B. The sources quoted (see references for Table II-1) represent independent sets of measurements.

TABLE II-1A

PLUTONIUM ISOTOPE RADIATIONS

	<u>Rad'n.</u>	<u>% Decays</u>	<u>Mev</u>	<u>Half-Life</u>	<u>Curies/gm</u>	<u>Spontaneous Fission n/s - gm</u>	<u>Ref.</u>
Pu-238	α	100	5.49	89.6 y	16.1	3,420 (a)	(4)
	γ	.001	.15				
	γ	.008	.10				(1)
	γ	.038	.044				
	L x-ray	13	.017(.014, .024)				
Pu-239	α	100	5.14	21,360 y	.0615	.03	(1)
	γ	.002	.038				
	γ	.007	.052				
	γ	.005	.10				
	γ	.003	.124				
	γ	.002	.384				
	L x-ray	1.4	.014				
	γ	2.2	.017				
Pu-240	γ	0.2	.021	6,580 y	0.227	1,380 (a)	(4)
	α	76	5.16				
	γ	.01	.044				
	L x-ray	10	.017				
	K x-rays	40	.102-.125				(1)
Pu-241	α	.003	4.9	13.3 y	111.5	-	(4)
	β	99.997	.02				
	γ	2×10^{-4}	.145				
	γ	.001	0.10				
Pu-242	α	76	4.89	380,000 y	.004	2,300 (a)	
	α	24	4.86				
	γ	.01	.045				(1)
	L x-ray	10	.017				
Pu-243	β	100	.39	5 hr	-	-	(3,4)
	γ	-	.095				
	γ	-	.12				
U-237 (from Pu-241)	β	100	.245	6.75 d	(b)	-	(4)
	γ	61	.059				(1)
	γ	35	.207				
	γ	4	.334				
Am-241 (from Pu-241)	α	84%	5.48	470 y	(c)	-	(4)
	α	13.6	5.43				
	γ	37	.017				(1)
	γ	2.7	.026				
	γ	.05	.043				
	γ	37	.059				
	γ	.02	.099				

(Footnotes follow Table II-1B on next page.)

TABLE II-1B
PLUTONIUM ISOTOPIC NEUTRON SOURCES

Isotope	Half-Life Years	Alpha Energy Mev	Spontaneous Fission Rate		Alpha-n Reactions; per gram or Kg of Pu Isotope in Compound					
			Neut/sec-gm of pure isotope	rem/hr*	PuO ₂		PuC		PuC ₂	
					n/s-gm	rem/h.*	n/s-gm	rem/hr*	n/s-gm	rem/hr*
Pu-238	89.6	5.49 5.47 5.15	3,420	4.20	30,200	37	30,600	37	58,500	72
Pu-239	24,360	5.14 5.10	.03	--	56.5	.07	58.1	.07	111	.13
Pu-240	6,580	5.16 5.12	1,380	2.24	209	.25	214	.26	409	.49
Pu-242	380,000	4.89 4.86	2,300	2.82	--	--	--	--	--	--

* Calculated surface dose-rate for 1 kilogram sphere of metal neglecting self-absorption. Plutonium isotopes 241 and 243 produce no neutrons. Alpha-n data from reference 2.

Footnotes for Table II-1A and II-1B

- (a) Spontaneous fission also yields an average of 7.5 one-Mev gammas/fission, plus one Mev of fission products per fission at equilibrium (80% of equilibrium value is reached in one day). (1)
- (b) $4.7 \times 10^{-3} (1 - \exp(-.102t))$ curies/gm of Pu-241, where t = days after chemical separation (1).
- (c) $4.25 \times 10^{-4}t$ curies/gm of Pu-241 (t in days after chemical separation up to about 1,000 days). (1)

References:

- (1) W. C. Roesch, Geneva Conference (1958) Volume 23, p. 339, P/755.
- (2) E. D. Arnold, Geneva Conference (1958) Volume 13, p. 237, P/1838.
- (3) J. F. Mech, et.al., Physical Review, 103, July (1956).
- (4) J. M. Hollander, I. Perlman, and G. T. Seaborg, Reviews of Modern Physics, 25, No. 2, 469-651, April (1953); also, D. Strominger, et.al., Reviews of Modern Physics, 30, No. 2, Part II, April (1958).

B. Dose from Recycled Plutonium

Taking the typical composition of the fast reactor discharge plutonium for one cycle (Case A) and five cycles (Case B) of 100,000 MWD/T each, (see Table II-2) and considering neutron emission as the primary penetrating radiation, one obtains the dose-rates shown in Table II-3.

The entries under plutonium-241 in Table II-4 reflect the growth of uranium-237 and americium-241 during the first year after chemical separation of the plutonium. The uranium-237 saturates relatively early, (in about 25 days) but the americium-241 (470 year half-life) continues to grow roughly linearly for two or three years, eventually saturating and then decaying as the plutonium-241 parent is depleted with a 14 year half-life.

Of the radiations which grow with time, the energy of one of them, americium-241, is only 99 Kev, so the dose-rate becomes small with leaded gloves (less than 1 mr/hr net for a 1 gram source at 1 cm). The other activity which grows with time is uranium-237.

The penetrating uranium-237 radiations include 207 Kev and 334 Kev gammas, which are not significantly attenuated (~63 and 84% transmission) by leaded gloves. The unshielded dose-rates (75 and 24 mr/hr respectively at 1 cm for a 1 gram source) nevertheless permit handling time of the order of 2 to 15 hr/week for most operations. For a fabrication plant at the same location as a reprocessing facility, the use of freshly separated plutonium could be used to reduce the dose-rates given above for uranium-237 to the following percentages: 1 day - 9.5%; 5 days - 40%; 10 days - 64%. (See footnotes on Table II-1 for growth parameters.)

However, since the uranium-237 dose-rate is usually small compared to the plutonium-240 gamma, significant investment in chemical decontamination capability is not justified if the fabrication facility is not already near a processing facility.

An important radiation for contact handling in glove-boxes is a group of 102 to 125 Kev x-rays from plutonium-240. The (narrow beam) transmission through leaded gloves ranges from 4.5 to 16%.

Taking an average transmission factor of 0.10, a transmitted dose of 440 mr/hr results from a 1 gram source.

Allowing for glove attenuation, and neglecting self-absorption, the significant contact dose-rates for a 1 gram source are approximately:

Pu-240 (102 to 125 Kev)	440
U-237 (207 and 334 Kev)	67
Neutrons (>1 Mev)	8
	<hr/> 515 mr/hr

The permissible glove-contact handling time is slightly under 3 hrs/wk (1,500 millirem/week permissible dose).

For larger sources, the self-absorption of the gamma rays of 100 to 125 Kev sources limits further significant increase in surface dose-rates. For intermediate-size sources (over 50 grams of plutonium) the uranium-237 activities (207 and 334 Kev) will predominate in the glove-transmitted radiation.

However, for sources in this size range, procedures to minimize or eliminate contact handling are practical. (A combination of distance, handling fixtures, and lightly shielded containers is postulated.)

For large sources, such as a fuel assembly (~5 kg of plutonium), the neutron emission becomes the dominant penetrating radiation, but distances of the order of one meter give working times of the order of 1 d/wk. (See Table II-3.)

C. Alpha-Neutron Reactions and Plutonium-238

The specific alpha activity of irradiated plutonium (Case A, Table II-3) is more than twice that of plutonium-239. The increase is largely due to the plutonium-240 and plutonium-238 content.

The most important source of alpha-neutron emission is from oxygen content of plutonium dioxide, (or plutonium dioxide mixed with uranium dioxide). The neutron emission for mixtures of plutonium with other elements or impurities in processing steps prior to the oxide are relatively unimportant since handling in a glove-box is not required for these steps.

Table II-1 also shows isotopic neutron emission rates for PuO_2 , PuC and PuC_2 . PuO_2 and PuC are nearly the same in α , η yield. Although PuC itself may be unstable under operating conditions, the neutron yield applies also to $(\text{Pu} + \text{U})\text{C}$ solid solutions, which are believed to be more stable.

D. Effect of Plutonium-238 Buildup

For the plutonium compositions used in Table II-2, the main source of neutrons is spontaneous fission of plutonium-240. The amount of plutonium-240 does not change significantly between the first cycle and the fifth cycle as shown by comparison of Case A and Case B in Table II-2. The other significant source of neutrons is alpha-neutron reaction with oxygen in PuO_2 . Using the experimentally determined neutron yield coefficients (Table II-1, Reference 2) the additional neutron source from the alpha-n reaction corresponds to 25% of the total neutron emission.

The plutonium-238 content is difficult to calculate with precision since several of the important cross-sections are not known for the spectrum of the fast reactor.

The most important chain in this respect appears to be as follows:

Estimating that the americium-241 capture cross-section is equal to the plutonium-239 cross-section, the plutonium-238 after a single cycle rises to 0.14% by weight of the plutonium.

The plutonium-238 content would have to rise to 1.7% of the total plutonium on prolonged recycle, if the additional neutrons from this source of alpha-n reaction were to double the total neutron dose-rates given in Table II-4.

The estimated increase in plutonium-238 is about 0.14% for the first cycle, and about 0.11% per cycle for later cycles. Allowing for decay and capture (assumed equal to uranium-238) results in about 0.5% of plutonium-238 after five cycles of 100,000 MWD/T each.

TABLE II-2

CALCULATED GAMMA AND X-RAY DOSE-RATES AT CONTACT FOR A SPHERE CONTAINING 1 KG OF FAST REACTOR RECYCLE PLUTONIUM

Atom % of Isotope In Pu		Days After Chemical Reprocessing	Rad/Hr of Gammas and X-Rays			
Case A*	B**		Hard, 125 Kev		Soft, 125 Kev	
			A	B	A	B
Pu-238	0.1	0.5	-	-	.20	1.0
239	54.7	43.1	.006	.005	.14	.11
240	33.1	40.0	.04	.05	.92	1.1
242	4.5	10.5	-	-	-	-
<u>Total for all Isotopes of Pu</u>		1	.07	.074	1.3	2.2
		5	.15	.14	1.3	2.3
		25	.29	.25	1.4	2.4
		365	.31	.26	2.4	3.1
		1,000	.31	.26	4.4	4.6
					<u>U-237</u>	<u>Am-241</u>
		1	.024	.019	.01	.004
		5	.10	.08	.04	.014
		25	.24	.19	.10	.07
		365	.26	.20	.11	1.0
		1,000	.26	.20	.11	3.0
					<u>U-237</u>	<u>Am-241</u>
					.008	.003
					.03	.011
					.08	.055
					.09	.80
					.09	2.3

* Composition discharged after one cycle of 100,000 MWD/T (Case A).

** Composition discharged after five cycles of 100,000 MWD/T (Case B).

TABLE II-3
PLUTONIUM COMPOSITIONS AND NEUTRON SOURCES IN FCR DISCHARGE AND RECYCLE FUEL

	<u>Atom Percent</u> <u>of Isotope</u>		<u>α, η from PuO_2 or PuC</u> <u>n/sec/g Pu</u>		<u>Spontaneous Fission</u> <u>n/sec/g Pu</u>	
	<u>A</u>	<u>B</u>	<u>A</u>	<u>B</u>	<u>A</u>	<u>B</u>
Pu-238	0.1	0.5	30	150	3	15
239	54.7	43.1	31	24	-	-
240	33.1	40.0	70	84	460	555
241	7.6	5.9	-	-	-	-
242	4.5	10.5	-	-	<u>103</u>	<u>240</u>
			131	258	566	810
Total n/sec/gm			Case <u>A</u>	697	Case <u>B</u>	1,068

Case A uses the plutonium discharge composition after one cycle of 100,000 MWD/T. Input to the cycle is reactor-grade plutonium (from a thermal reactor) which is 60% plutonium-239; 25% plutonium-240; 12% plutonium-241, and 3% plutonium-242.

Ref: P. Greebler, P. Aline, and J. Sueoka, "Fast Oxide Breeder Reactor Physics - Part I", GEAP-3287, November (1959), p. 20, Case 5.

Case B used the plutonium discharge compositions estimated for recycled plutonium after 5 cycles of 100,000 MWD/T each.

Ref: B. A. Hutchins, "Plutonium Isotopic Compositions in FOR Reference Design Fuel", (which is a revision of recycle calculations in GEAP-3087 by P. Greebler, December, 1958), Memorandum, November 10, 1961.

TABLE II-4

FCR-PLUTONIUM: NEUTRON DOSE-RATES

<u>Source</u>		<u>Neutron Dose-Rate Millirem/Hr*</u>		<u>Time Limits Hrs/Week**</u>	
		<u>Distance: 1 cm</u>	<u>100 cm</u>	<u>1 cm</u> (hands)	<u>100 cm</u> (whole body)
One pellet (.22 gm Pu)	<u>CASE</u> A	1.6	1.6×10^{-4}	>40	>40
	B	2.5	2.5×10^{-4}	>40	>40
One pin (46 gm Pu; 3.5 feet long)	A	95.	0.30	16	>40
	B	145.	0.46	10	>40
One assembly (127 pins)	A	2430	33.	0.62	9.1
	B	3720	51	0.40	5.9

* Based on $7.7 \text{ n/cm}^2\text{-sec} = 1 \text{ Mrem}$; distances are measured from the surface along a perpendicular to midpoint of the axis of the cylinder.

Pin calculated as a line source where $\phi = \frac{\Delta}{4\pi Z} [F(\theta_1, \mu t) + F(\theta_2, \mu t)]$ and functions F are given in Nuclear Engineering Handbook, Etherington, Section 7-103, McGraw Hill (1958). The assembly was calculated as a summation of line sources at varying distances.

** Time limit assuming other penetrating radiation sources are small compared with neutron source; tolerance doses for setting time limit are:

1,500 Mrem/Wk to hands; 300 Mrem/Wk to body

Case A and Case B plutonium as defined in Table II-2.

III. CHEMICAL PROCESSING

A. Relation of Residual Plutonium Radiations to the Selection of Reprocessing Method

The purpose of chemical reprocessing of the irradiated fuel from a fast reactor is to recover the plutonium which is produced in a form which permits its convenient reuse in the reactor. Since the net conversion ratio of the fast ceramic reactor with either oxide or carbide fuel is somewhat greater than unity, the reactor is capable of fully self-sustaining operation on its own recycled fuel after the initial inventory is provided.*

Chemical processing methods may be roughly divided into "high-decontamination-factor" and "low-decontamination-factor" processes. Aqueous reprocessing, in which the oxide fuel is dissolved in a nitrate solution, separated from fission products, and reconverted to oxide, is a typical "high-decontamination-factor process" (5×10^7 to 5×10^8).

The selection of a recovery and decontamination process for spent fuel from fast reactors depends critically on the procedures to be used in refabrication of the recovered fuel. This in turn, is limited by the radiation levels which will be emitted by the recycled fuel. This radiation comes from two sources:

1. That which is inherently associated with chemically pure plutonium and the daughter products which grow into initially pure plutonium, (as discussed in Section II).
2. The radiation from residual fission products due to incomplete separation in the decontamination process.

Maximum decontamination factor which is required of the decontamination process is defined by the residual radiation of the recovered plutonium. The minimum decontamination factor which is needed is set by the refabrication process. If the conventional processes for making oxide fuels were to be used, (for example, cold-press and sinter pellets, grind to size or vibratory compaction or cold-swaging) then the cost of such an operation might be seriously increased if every step in the process had to be done by hot-cell procedures with manipulators or automated equipment, rather than by "direct" handling in glove-boxes.**

* In practice, two or three "7%-batch" reloads are required in addition to the core inventory itself in order to fill the pipeline between the reactor, the reprocessing plant, and the fuel fabrication shop. From the standpoint of inventory charges on the fuel, it is the overall kilowatts per kilogram of fuel including the out-of-pile inventory which is important since reduction of overall inventory has the same effect on inventory costs as an increase in the specific power of the in-pile inventory.

** The capital cost objection to fully automated "remote" control fabrication may disappear with a sufficiently large volume of production.

For this reason, it is important to design the fabrication procedure so that exposure from the residual radiation from fission products, plus that from the plutonium itself is kept low, otherwise, fabrication costs are likely to increase beyond acceptable values of about .3 to .4 mills/kwhr.

Two or three cycles of aqueous processing are sufficient to reduce the fission products to such low levels that their residual radiation levels are comparable to that from inherent radioactivity of the fuel material itself.

The "pyro" processes commonly give low decontamination factors. For example: the metal-slugging process under development at Argonne National Laboratory for alloy fuels, or the fused-salt electrolysis process, under development at Hanford Laboratory, typically give small decontamination factors, of the order 10^2 to 10^3 , so that the residual fission products prohibit direct handling procedures for refabrication of fuel or maintenance of the equipment.

The analysis in Section II indicates that the residual radiation from highly-irradiated plutonium is sufficiently low that conventional dry-box handling procedures can be adapted to refabricate fuel. The added radiation is a significant but tolerable addition to the handling restrictions normally applicable to low-irradiation plutonium. This requires additional provisions for limiting the quantity of plutonium handled at one time, provisions for small thicknesses of localized shielding when larger quantities are involved, and the use of handling methods which minimize the need for surface contact handling of plutonium-containing materials. Most radiations associated with highly irradiated plutonium are very soft, being largely self-absorbed, or greatly attenuated by heavy rubber gloves. The limiting penetrating radiation for large masses - of the order of a kilogram - is from spontaneous neutrons and alpha-n neutrons.

Special gloves are used for plutonium handling which are made with lead or zinc-loaded rubber. To the usual 110 mg/cm^2 of 30 gauge rubber gloves, is added the absorption equivalent of 0.5 mm of lead. This eliminates almost completely the abundant radiations below 50 Kev and provides an attenuation factor of about 9 at 100 Kev.

Since the inherent plutonium radiation permits (controlled) direct-handling processes, there is an incentive to reduce the residual radiation from fission products to a level which will be negligible compared with the plutonium radiations themselves.

One of the possible advantages of the "dry processes" for some fuel types is to avoid the cost burden of chemical conversion steps before and after chemical processing. For example: if plutonium alloy is used as a fuel, a wet process would require dissolving the metal and finally reconvert to the metal by way of the nitrate, the oxide, the fluoride, metal-reduction and casting processes. Refabrication of metallic fuels by a melting and slugging procedure by elimination of the conversion steps may therefore offset part or all of the remote handling costs.

In the case of oxide fuel there appears little or no incentive for avoiding "wet" processes. This is because the chemical free energy difference between the oxide and the nitrate is relatively small, so that plutonium can be dissolved, and returned from nitrate to the oxide form by inexpensive steps, with costs of the order of 20¢/gm, compared with \$1 to \$2/gm for metal-reduction.

Carbide fuel fabrication and reprocessing have not been covered in this study, pending the outcome of a current evaluation of reactor design and performance with carbide fuel. However, most of the unit processes used with carbide are similar to those used with oxide. Also, the radiation level analysis in Section II indicates that the alpha-n neutron emission from carbide fuel is almost identical to that from oxide. A reasonable hypothesis for the present is that carbide fuel will have processing and fabrication costs generally similar to those estimated for the oxide. (This assumes a time at which the level of experience in fabrication and in performance testing of carbide approaches that now available with oxide.)

An implied restriction placed upon this study is that the reference reprocessing and refabrication cycle be based on technology for which there is enough experience to give good assurance of feasibility and firm basis for cost estimates. This does not eliminate from serious consideration "dry processes" although only the "wet processes" have been demonstrated and operated routinely on a large scale. Of the "dry" processes directly useable for oxide, only the chloride electrolysis approach is under active study (at Hanford) but it is still in a developmental state technically and economically. It depends on remote refabrication of the fuel which still would contain intense fission-product activity. While this approach merits further study, it is premature to adopt it as a reference design.

The problem of selection of a reference process is narrowed to a choice between different "wet" processes - primarily between an adaptation of the Purex process, which is solvent extraction using tributylphosphate ("TBP") and an ion-exchange process (cation or anion exchange process).

B. Gross Fission Product Activity of Irradiated Fuel

The gamma activity of fast reactor fuel as a function of irradiation and cooling is shown in Figure III-1 based on fast-fission yields of plutonium. The typical values of interest range from 800 to 1,000 days irradiation to give 100,000 MWD/T burnup, and 30 to 100 days cooling. Fuel experiencing 70 days irradiation and 30 to 100 days cooling corresponds to fuel which is removed after one refueling interval in the case of the initial core loading.

C. Decontamination Factors Required to Permit Direct Handling

Table III-1 shows the overall decontamination factors required for various cooling times using the data of Figure III-1 for the initial activity. Because of the long average half-life after 1,000 days irradiation time, there is only a factor of 7 reduction in the decontamination factor required when cooling time is increased from one month to one year.

GAMMA ACTIVITY FROM FAST FISSION OF PLUTONIUM - 239
FIGURE III-1

TABLE III-1

FISSION-PRODUCT DECONTAMINATION FACTORS REQUIRED*

<u>Days Irrad'n.</u>	<u>Days Cooling</u>	<u>Gamma Curies/Gm</u>	<u>DF Required To:</u>	
			<u>a. Equal Pu Rad'n.</u>	<u>b. 10% of Pu Rad'n</u>
1000	30	110	5×10^7	5×10^8
	60	75	3.5×10^7	3.5×10^8
	90	65	3.1×10^7	3.1×10^8
	120	55	2.6×10^7	2.6×10^8
	180	35	1.7×10^7	1.7×10^8
	365	24	1.2×10^7	1.2×10^8

* For plutonium irradiated for one cycle to 100,000 MWD/T at a specific power of 666 watts/g of total plutonium.

The final activity desired should be at most equal to the hard component of plutonium radiations, (neutrons) and preferably only 10% of the plutonium radiation.

The decontamination factors shown in Table III-1 are those required to reduce gamma activity from plutonium irradiated to 100,000 MWD/T and 666 W/gm of total plutonium, to 7,000 gamma photons/sec-gm. This 10% of Rem calculated in Section II from spontaneous fission and alpha-n neutrons from the $\text{UO}_2\text{-PuO}_2$ mixture, assuming 780 gamma photons/cm²-sec = 1 m Rem/hr for 0.7 Mev average fission product energy, and 7.7 neutrons/cm²-sec = 1 m Rem/hr for 2 Mev neutrons.* For a neutron source of 697 neutrons/gm-sec, a residual fission gamma source of 7,000 gamma photons/sec-gm gives one-tenth the dose of the neutron source. See Table II-3 for derivation of the neutron activities.

The values shown in Table III-1 are for "one-cycle" plutonium (Case A of Table II-3). For "five-cycle" plutonium (Case B) the total neutron emission is 1,068 n/sec-gm, or 1.53 times greater than for Case A. The decontamination factors required for Case B are smaller by a factor of 0.65 than those for Case A.

D. The Effect of Long Irradiations and Long-Lived Fission Products

Consideration has been given to the different proportion of long-lived fission products to be expected in a fast reactor fuel irradiated for a total period of roughly 1,000 days, compared with the roughly three-tenths of a year thermal reactor irradiations used to produce the feed material for nearly all process development done to-date. Table III-2 lists the long-lived fission products and their yields calculated for irradiations of 365 and 10,000 days. Of the ten long-lived isotopes, only three produce radiations of sufficiently high energy to contribute appreciably to a handling problem for irradiated plutonium even if poor decontamination factors were found. Of these three, the decontamination factor for only one of them, ruthenium, has been determined for one cycle of the preferred process, anion exchange. For the amount of ruthenium left after 1,000 days of irradiation, a DF of 6.5×10^4 leaves a residue of less than 3 millicuries per gram of plutonium. (Note that core and blankets are mixed together for reprocessing.) An overall decontamination factor of about 10^6 is sufficient to make this radiation source negligible.

The second long-lived isotope emitting hard radiation is Cs-137 with its Ba-137m daughter. Because of the greater chemical dissimilarity of the cesium and barium from plutonium, and their reversible chemical behavior, large decontamination factors are observed for these elements in successive cycles of both solvent extraction and ion exchange processes.

* K. Z. Morgan, Section 7.32, Nuclear Engineering Handbook, H. Etherington, Editor McGraw Hill, 1958. This value of 7.7 n/cm²-sec is slightly more conservative than NBS Handbook 63. 10 Code Fed. Reg., Volume 20, which gives 8.0 n/cm²-sec for 2.5 Mev.

TABLE III-2

YIELDS OF LONG-LIVED FISSION-PRODUCTS*
FROM FAST FISSION OF PLUTONIUM-239 AND 1 YEAR TO 27 YEAR IRRADIATIONS

	<u>Half-Life</u>	<u>Atoms Remaining per 100 Atoms of Pu Fissioned**</u>			<u>Radiation</u>
		<u>365 d</u>	<u>10³ d</u>	<u>10⁴ d</u>	
Ru-106	1 yr	3.78	2.10	.277	Beta (short-lived Rh daughter gives gammas to 2.66 Mev)
Sn-119	250 da	.003	.001	.000	Soft x-ray
Sb-125	2.7 yr	.219	.13	.034	Beta and hard gammas up to .637 Mev
Te-127	115 da	.022	.010	.001	Pure beta
Ce-144	282 da	1.91	1.0	.118	Beta and soft gammas
Pm-147	2.6 yr	2.1	1.2	.327	Pure beta
Sm-151	73 yr	1.29	1.2	1.141	Beta and soft gamma
Eu-155	1.7 yr	.131	0.10	.014	Beta and soft gamma
Cs-137	27 yr	5.36	4.60	3.93	Beta and .66 Mev gamma from Ba-137 ^m daughter
Sr-90	28 yr	2.26	2.02	1.66	Pure beta

* L. Burris and I. Dillon, ANL-5742, June (1957), data for 365 day and 10⁴ day irradiations, 100 d cooling.

** Basis: Half-life between 100 days and 100 years, and excluding the noble gases.

The remaining isotope with an appreciable component of hard radiation is antimony-125. Because of yield and half-life, this contributes less than 5% as much radiation as does ruthenium¹⁰⁶. The yield of antimony for short thermal irradiations is low (.02 atoms/100 atoms of plutonium fissioned, for 30 days irradiation and 100 days cooling). It is probable but not certain that a poor decontamination factor for this element would have been detected in the development work quoted, although zirconium-niobium and ruthenium-rhodium together typically account for over 90% of the residual activity in the decontaminated material.

A poor decontamination factor for antimony is unlikely on chemical grounds, because of substantial dissimilarities in oxidation potentials and ionic species relative to plutonium. (Plus 3 and plus 5 states of antimony are stable, while plutonium is mainly plus 4 in acid solutions.) However, since very large overall decontamination factors are desired, of the order of 10^7 to 10^8 , it would be prudent to check this factor experimentally as well as those for the more abundant activities and as ruthenium and zirconium-niobium.

E. Ion Exchange Processes

Cation exchange processes using Dowex-50 resin have been used routinely on a large scale to concentrate and to complete decontamination of plutonium product from the Purex process.*

This process development has provided extensive data on the adsorption and elution characteristics of plutonium, the valence adjustment steps, the partition from uranium and fission products, and the pressure-drop and flow characteristics of resin beds used in nitrate solutions. Finally, there is extensive data on the extent and rate of damage of ion exchange resin by the alpha particles from plutonium and beta and gamma radiation from fission products.

Better separation of plutonium from uranium and fission products is obtained by using anion exchange rather than cation exchange. Plutonium is efficiently adsorbed from strong nitric acid solutions by anion exchange resin (as a negatively charged nitrate complex) while uranium and fission products are largely rejected. The decontamination factor through one cycle of adsorption and elution on an anion resin such as Permutit SK gives decontamination factors at least as large as those obtained through one cycle of solvent extraction by the Purex process.

The performance observed for the anion exchange process is summarized below, based largely on the work of Ryan and Wheelwright.*

-
- * D. C. Overholt, et.al, "Anion Exchange Process for Plutonium Isolation and Purification", ORNL-1357, October (1952) and ORNL-1520, May (1953), also; P. B. Branin, et.al., "Concentration and Decontamination of Plutonium Process Solutions by Ion Exchange", HW-30063, January (1953).
 - ** J. L. Ryan and E. J. Wheelwright, "The Recovery, Purification and Concentration of Plutonium by Anion Exchange in Nitric Acid", HW-55893 (DEL.), January 2, 1959.

a. Separation of Plutonium from Uranium

Uranium separation factors 1.0×10^6 to 6.8×10^6 were obtained for solutions with a uranium-to-plutonium ratio of 294. Separation factors of 0.4×10^6 to 1.1×10^7 were obtained over a range of uranium-to-plutonium ratios varying from 60 to 1,440.

b. Decontamination Factors for Fission Products

Decontamination factors range from 8.6×10^4 to 4.6×10^5 for zirconium and niobium, using feed containing 7.2 M nitric acid and .01 M HF. Ruthenium-rhodium decontamination factors were 6.5×10^4 (at the limit of measurement). In a longer series of runs, in which HF was omitted from the feed solution, decontamination was somewhat lower, ranging from 1.9×10^4 to 3.4×10^4 for ruthenium-rhodium. For the feed material used in these experiments (less than 5,000 MWD/T), the gross decontamination factors were about 80% of the decontamination factors for zirconium-niobium.

Overall decontamination factors better than 2×10^4 per cycle are reasonably well-demonstrated for the first cycle, and comparable factors are probable but not demonstrated for a second cycle. Overall decontamination factors of 10^7 to 10^8 , as required for material cooled for 60 days appear reasonably well-assured for this process.

c. Separation of Plutonium from Simple Anions

A decontamination factor of 620 was obtained in one cycle from sulphate ion using Dowex-1 resin at 50 C with a 4-column wash with 7.2 M nitric acid. Similar separation from chloride and phosphate is stated to be readily obtainable.

d. Separation Factors from Metallic Impurities

One-cycle separation factors greater than 20,000 were obtained for silver, calcium, chromium, iron, potassium, and lithium ions. Separation factors of 5,000 or greater were found for aluminum, copper, magnesium, manganese, and nickel. A separation of 500 was obtained from sodium ion.

All of the foregoing factors were obtained with a column 90 centimeters in length, 0.28 cm^2 in cross-section, using Dowex-1, X-4 resin, with a wash consisting of 15 bed volumes of 7.2 M nitric acid. The initial concentrations of the impurities were 10% by weight of the plutonium for the elements listed, except iron, which was 1% by weight of the plutonium. The column was operated at 60°C .

F. Solvent Extraction Process for Fast Reactor Fuel

An extensive effort was carried out at Knolls Atomic Power Laboratory to design and demonstrate on small scale the solvent extraction recovery of plutonium from typical fast oxide reactor fuel.*

Fission product decontamination factors of 300 to 3,000 were obtained in one cycle using small scale extraction equipment. Larger factors are normally obtained in large scale equipment because hydraulic instabilities typical of the small scale equipment are eliminated. A typical Purex process flowsheet was tested through one cycle. Both the decontamination factors and plutonium recovery deteriorated significantly, approaching minimum acceptable limits for solvent irradiation levels of 5 watt-hours/liter and above. Because of the lower burnup levels, lower specific power and dilution of the core fuel with lower burnup blanket material typical of the present reference design, the solvent irradiation levels expected are approximately one-fifth of that which was assumed for the KAPL work. For 1,000 days irradiation and 90 days cooling, there is 0.46 watts of beta radiation emitted per gram of plutonium in the process of fuel mixture, compared with approximately 2.2 beta watts/gram of plutonium estimated for the reference conditions of the KAPL work. On the basis of this work, it appears probable that the conventional Purex process is feasible for fast reactor fuel, provided that the selection of the extraction equipment is such as to minimize the residence time and the radiation exposure of the solvent in the extraction section of the first column.

A more certain alternative is the use of "head-end" treatment to obtain a preliminary decontamination factor of 5 to 10 which would substantially reduce the radiation damage problems of the solvent. Scavenging with inorganic adsorbants such as manganese dioxide, on which extensive development has been done, routinely yields decontamination factors (for zirconium-niobium and also iodine) of 10 to 30 with negligible plutonium losses. Overall decontamination factors of about 4×10^7 are routinely attainable in continuous operation of two Purex cycles plus either a "head-end" scavenging process using manganese dioxide, or else a terminal cleanup and concentration step using cation exchange.**

Since the decontamination factor for two cycles of Purex alone would be marginal for the requirements of fast ceramic reactor fuel, either a third cycle of solvent extraction "head-end" treatment, or else a concentration-decontamination cycle following two cycles of solvent extraction, appears necessary. A reference process based on "head-end" treatment with MnO_2 and two cycles of solvent extraction, appears necessary to provide routinely overall decontamination factors of greater of 10^7 to 10^8 .

* J. K. Davidson, W. O. Haas, et.al, "The Fast Oxide Breeder - The Fuel Cycle", KAPL-1757, July 1, 1957.

** E. L. Zebroski and W. O. Haas, "Purex Process Development", TID-2012, March (1954), also;
Reactor Handbook, 2nd Edition, Volume 2, Interscience Publishers, (1961), pp. 147.

G. Recommended Reference Process

On the basis of the data which has been reviewed, two wet processes are feasible:

1. "Head-end" treatment and solvent extraction (possibly followed by an ion exchange concentration and cleanup step) or,
2. Two cycles of anion exchange followed by a precipitation step.

Each offers reasonably good assurance of providing both the required decontamination factors of 10^7 to 10^8 . The solvent extraction process with terminal cleanup offers a considerable margin of safety with respect to the decontamination factors available, over the minimum values required. This implies the adaptability of existing chemical processing units at Idaho, Hanford, Savannah River, or Oak Ridge. The capacity required for the fast reactor is small, however, so that a pilot-plant scale of solvent extraction operation is ample in capacity for many thousands of megawatts of fast reactor refueling at steady-state. For example, a small solvent extraction plant operating at 1 ton per day and 270 days per year would service over 10,000 MWe of fast reactors of the reference design type.

Conversely a plant of this type would operate at a very low fraction of its capacity and with relatively high unit costs, if it depended on fast reactors for its volume.

It appears doubtful that in the next 10 to 15 years solvent extraction will be economically feasible for fast reactors, except perhaps in multi-purpose plants which obtain the bulk of their reprocessing volume from thermal reactors.

The anion exchange process has been chosen as the reference process for the following reasons:

1. The equipment and unit processes involved are somewhat simpler than with solvent extraction, which requires both extraction equipment and a "head-end" treatment or a terminal ion exchange step.
2. The anion exchange process offers the possibility of attaining good reprocessing economics on a smaller scale of operation than in the case of solvent extraction; therefore attractive reprocessing costs are attainable with a small number of fast reactors, totalling 1,500 to 3,000 megawatts electrical, for 10^5 MWD/T fuel life.

The second reason above is important because it makes it possible to establish a reference basis for fast reactor reprocessing costs which is independent of assumptions on the availability and costs of reprocessing fuels from other reactors, or on the efficiency and costs of multi-purpose processing facilities. Details of the reference anion exchange flow-sheet are given in Appendix A.

If the mixed oxides are prepared by the reference method - co-precipitation of ADU and plutonium hydroxide - (see section IV) no significant additional decontamination factors are obtained. However, if necessary, plutonium may be precipitated alone by oxalate or peroxide, which affords substantial added decontamination factors.

Depending on the results of pilot studies, final cleanup by the alternate precipitation method would either be incorporated in the process routinely, or else provided as a method for re-work of occasional off-specification product streams. The cost estimates in Section IV include provision for this alternative.

H. Fuel Throughputs and Plant Capacities

Parameters of interest to reprocessing estimates are taken from the FOR Reference Plant (see Section I-B).

Although other reactor cases may be considered, this study will be confined to the following case so that cost information can be combined with other costs for the reference FOR. (See Table III-3.)

The reprocessing plant should therefore have a capacity of multiples of 46.5 KgU + Pu/day. The reference case (Case I) will process three reactors of 525 MWe each.

Three sizes of reprocessing plant capacities are considered: (See Table III-4.)

It should be noted that an allowance of increased fuel fabrication volume is needed to provide inventory for growth of new capacity (see Section III-J and Table IV-3).

Fabrication capacity corresponding to about twice the current power generation rate is appropriate for typical growth rates.

Chemical reprocessing rates, on the other hand lag behind power generation capacity by the average fuel lifetime and no analogous effect of augmented current processing rate results from the growth of generating capacity.

TABLE III-3

FUEL THROUGHPUTS

<u>Load Factor</u>	0.8
<u>Thermal Power MW (Thermal)</u>	
Core	1220
Axial Blanket	115
Radial	82
Total	1417
<u>Total Electrical Power MW Electric</u>	525
<u>Throughput,* Kg/day of U + Pu +FP</u>	
Core	11.9
Axial Blanket	8.4
Radial Blanket	26.2
Total	46.5
Total Pu Throughput, Kg/day	2.5
<u>Equilibrium Fuel Exposure, MWD/2,000 lb(U + Pu)</u>	
<u>Discharge</u>	
Core	100,000
Axial Blanket	12,200
Radial Blanket	3,050
Average of Core and Blankets	29,500
<u>Specific Power, Kw/Kg (U + Pu), at Equilibrium</u>	
Core	128
Axial Blanket	16.9
Radial Blanket	3.9

* Based on 270 days/year reprocessing plant operation.

TABLE III-4

REPROCESSING PLANT CAPACITIES

	<u>Case I</u>	<u>Case II</u>	<u>Case IA</u>
MWe*	1575	3150	525
Kg/day			
U + Pu + FP	139.5	279.0	46.5
Pu (all isotopes)	7.53	15.6	2.5
Fission Products	4.9	9.8	1.6

Typical distribution of fuel, plutonium, and fission products in core and blanket are shown in Table III-5.

* Basis: Assuming 80% capacity factor on reactor operation, 270 days/year onstream for processing plant. Case IA is the same physical plant as for Case I, operating at reduced capacity.

TABLE III-5

TYPICAL FUEL MASS BALANCE AND THROUGHPUT FOR CORE AND BLANKETS OF A 525 MWe FAST REACTOR

	<u>Core</u>	<u>Blanket</u>		<u>Total</u>
		<u>Axial</u>	<u>Radial</u>	
Kg (U + Pu) initial loading	9,530	7,340	20,900	37,720
Kg Pu {initial final }	1,890	0	0	1,890
	1,610	220	170*	2,000
Kg Fission Products	1,120	102	78	1,300
Kg Pu (fissioned and capture)	1,170	105	75	1,350
Kg Pu Formed	890	325	245	1,460
Final Pu% (avg.)	16.9	3.0	0.80*	
<u>Kg/Yr (80% Load Factor)</u>				
(U + Pu + FP)	3,230	2,485	7,090*	12,705
FP	380	34.6	96.4	440
Pu	545	75	57	677
<u>Kg/Day (U + Pu + FP)</u> <u>(270 days/yr operation)</u>				
Pu	2.02	.28	.21	2.51
FP	1.40	.15	.10	1.63
tMWD/yr	3.56×10^5	3.36×10^4	2.39×10^4	4.14×10^5
MWD/T (average)	10^5	12,200	3,050	29,500

* Weight and average per cent plutonium gained in radial blanket per core life (2.95 yrs at 80% LF). As an alternative, using 5.9 yr radial blanket life would change this item to 1.6% plutonium, 3,545 kg/yr throughput, reduce total throughput to 9,160 kg (U + Pu + FP), and raise the average MWD/T to 41,000. See Section II for isotopic compositions of plutonium.

I. Reprocessing Economics

A plant design and cost analysis has been made for a very small solvent extraction plant handling the fuel for one 180 MWe thermal reactor.* The building and cell designs are ample to accommodate the equipment for the anion exchange process sized for Case I, 1575 MWe. Individual equipment costs were adjusted to allow for differences in criticality limits, waste and off-gas volumes (see Appendix A). The adjusted costs are given under Case I. Cases IA and II were derived by appropriate scaling of each item in Case I.

TABLE III-6

COST ESTIMATES - REPROCESSING BY ANION EXCHANGE

	Case I 1,575 MWe	Case II 3,150 MWe	Case IA (525)**MWe
Labor	511,100	895,000	238,000
Chemicals and Materials	43,900	85,800	14,300
Utilities	11,000	22,000	5,100
Supplies	13,700	24,000	4,600
Waste Storage	97,400	158,500	45,200
Pu Loss (@ \$5.00/gm) x (0.2% loss)***	19,000	38,000	5,800
<u>Buildings and Equipment</u>			
Fixed Charges (16.9% p.a.)	<u>676,000</u>	<u>1,020,000</u>	<u>676,000</u>
	1,371,100	2,243,300	989,000
\$/kg of U + Pu	\$35.90	\$29.40	\$77.80
\$/g of Pu	\$ 0.67	\$ 0.55	\$ 1.46
KwHr/Yr (at 80% LF)	1.10×10^{10}	2.20×10^{10}	3.67×10^9
Mills/KwHr	0.125	0.102	.269

* Idaho Small Processing Plant - "A Study of the Feasibility of a Small Scale Reprocessing Plant for the Dresden Nuclear Power Station", IDO-14521, H. Schneider, et.al., April 28, 1961.

** Case IA is based on operating the plant designed for Case I at one-third capacity.

*** See Section V and Table V-1 for basis of "1975 Plutonium Value".

IV. FABRICATION PROCESS FOR OXIDE FUEL

A. Choice of Preferred Process

A conventional cold-press and sinter pellet process has been selected as the reference process for fuel fabrication.

Alternate procedures which have been considered include:

1. Vibratory compaction of powder.
2. Swaging powder-filled tubes.
3. Isostatic pressing.

Each of the alternate processes appear technically acceptable and compatible with the handling restrictions of recycled plutonium. Each also has had extensive development effort during the last two years, and has been reduced to practice for in-pile testing (although most often for larger diameter fuel rods than those of interest for the core of a fast reactor). The cold-press and sinter process has been selected for this study mainly because it provides the most reliable costing basis, (although vibratory compaction is believed to be more likely to be used in practice).

The reference process was chosen because:

1. It has had the most extensive reduction-to-practice in manufacturing practice and in cost experience with UO_2 fuels.
2. There is extensive irradiation testing experience.
3. Most of the unit processes (and also building, equipment and labor requirements) are closely similar for all three of the alternatives given above. Costing of the alternate processes requires only substitution of one or two of the unit processes, leaving the bulk of the process and the cost estimate unchanged.
4. It is not necessary to assume the less-well-developed processes in order to obtain attractively low reference fuel cycle costs.

The more advanced processes have the potential of decreasing the reference costs but the cost incentives for the alternate processes, at present, appear to be comparable to the probable uncertainties in the overall fuel cost estimate. Accordingly, the alternate processes are regarded as refinements for future study.

A block diagram of the reference process is shown in figure IV-1. A separate process line is used for the blanket rods which contain only uranium. The line which makes mixed plutonium-uranium fuel is in an enclosed operations area. All of the unit processes involving plutonium

* J. M. Cleveland and W. C. Cavanaugh, "Plutonium-Uranium Dioxide Preparation and Pelletized Fuel Fabrication", GEAP-3486, August 15, 1960.

FIG. IX-1 PuO_2 - UO_2 ROD FUEL FABRICATION PROCESSES

pins except final assembly are performed in glove-boxes. Final assembly may be performed in controlled ventilation hoods rather than glove-boxes. Except for special operations connected with maintenance or re-work and except for final assembly, all transfer operations are conducted within the glove-box line.

B. Blending

The starting materials may be either liquids in transfer containers (especially if the fabrication facility is near to the chemical processing facility), or a solid form convenient for shipment or storage. For uranium raw material either uranyl nitrate (UNH) or U_3O_8 or UO_2 feed may be used.

In the case of plutonium the common form for shipment or transfers is expected to be either plutonium nitrate "syrup", or low-fired oxide.

To obtain the greatest assurance of producing homogeneous uranium-plutonium solid solutions, it is preferred that the plutonium and uranium nitrate solutions are mixed and co-precipitated. This insures that there is no inhomogeneity in the uranium-to-plutonium ratio within the fuel. It is also possible to obtain mixed oxide solid solution by mixing the uranium and plutonium powders. However, this operation requires careful quality control to insure that inhomogeneities and off-specification uranium-plutonium ratios are avoided. For conservative costing, the solution-mixing approach has been used, although it is recognized that there may be some cost incentive for using powder mixing.

C. Precipitation of Mixed Oxides*

Plutonium-uranium solution with approximately 4:1 ratio of uranium-to-plutonium is prepared in approximately 2M nitric acid. Ammonium hydroxide is added over a period of about one hour to a pH of 9, and the mixture allowed to settle for one-half hour. The slurry is decanted washed with one-quarter volume each of ammonia and acetone, then filtered.

D. Alternate Precipitation Method for Plutonium Alone

If powder blending is to be used, plutonium alone is preferably precipitated as the oxalate rather than as the hydroxide.

Typical plutonium concentration range for precipitation is 50 to 100 grams/liter, and about 5M nitric acid. The solution is adjusted to about 2% peroxide and .5 M excess oxalic acid. After one hour digestion at 55°C the solution is slurried to a filter, and washed with a nitric acid-oxalic mixture.*

Typically about 1% of the plutonium remains in the filtrate and wash solutions for recycle.

* W. J. Maraman, "Purification and Concentration of Solvent Extracted Plutonium by Oxalate Precipitation", LA-1991, January (1956); also classified reports, HW-30558, HW-31186, HW-26450, HW-45700 and HW-67166.

The mixed oxalate precipitate is filtered, washed, and dried for about one hour at 120°C, and then pre-calcined at 130 to 300°C for three hours.*

E. Drying and Calcining

The mixed uranium-plutonium precipitate is air-dried, then pre-fired in hydrogen-nitrogen mixture at temperatures between 300 and 900°C depending on the stoichiometry and sinterability characteristics desired.

F. Pressing and Sintering of PuO₂-UO₂

Powder is ground in a ball mill to produce a particle size range of -140 to 300 mesh oxide. Pressing of this powder at 22 tsi, followed by sintering in hydrogen-helium mixture (approximate composition: 1 volume hydrogen to 3 volumes helium) results in sintered densities of 93 to 96% of theoretical.**

These results were obtained with mixed oxide from ammonium hydroxide precipitation. The optimum conditions may differ slightly, with oxalate precipitation and powder blending. The probable range of times, temperatures, or pressing pressures required would have no significant effect on the equipment required, and only minor effects on the fabrication cost estimates.

A general description of the details of the unit processes, and of a small-scale glove-box line which has been operated, is available in the work done to make irradiation capsule pin specimens for the Fast Oxide Breeder Project in 1960.*** A larger specialized glove-box facility is now nearing completion at APED.

G. Swaging over Pellets

In conventional cold-press and sinter fabrication of oxide fuel, it is common practice to centerless-grind the sintered pellets to final size.

This is an undesirable operation for plutonium-containing fuel for the following reasons:

1. The production of finely divided plutonium-uranium oxide scrap from the grinding operation makes for heavy contamination and difficulty in maintenance of this equipment.
2. A considerable amount of impure scrap re-work is routinely necessary.

Specifications for diametral tolerances for fast reactor fuel have not yet been established. In the irradiation testing done to-date for fast reactor oxide fuel there has been no significant correlation between gas release and pellet-to-clad diametral gap over a range of about 1 to 8 mills cold-

* M. N. Myers, "Thermal Decomposition of Plutonium Oxalate", HW-45128, August (1956).

** J. M. Cleveland and W. C. Cavanaugh, "Sintering Studies of Plutonium-Uranium Dioxide Pellets", GEAP-3487, August 15, 1960.

*** J. M. Cleveland and W. C. Cavanaugh, "Plutonium-Uranium Dioxide Preparation and Fuel Fabrication", GEAP-3486, August 15, 1960; also GEAP-3486, Part II, M. E. Snyder and W. R. Cowden, "Fabrication of Plutonium-Uranium Dioxide Specimens by Swaging".

gap on the diameter, and for densities varying from 83 to 96% theoretical.* Gas release ranged from 20 to 60% of the gas formed. This would suggest that on the limited data so far, as-sintered pellets might be used without further dimensioning, but possibly with a selection process to limit the range of pellet-to-clad gap within a single element.

A more conservative assumption is that a preferred pellet-to-clad gap will be specified as larger samples of fuel are tested to the limits of the desired performance. A final sizing operation is provided in the reference process, even though it may eventually be eliminated.

The reference sizing process selected is swaging-over-pellets using nylon dies. Starting with initial cold gaps of 20 to 26 mils, nylon die swaging has been used to reduce the diametral gap to less than 3 mils with no pellet breakage.** A series of irradiations in a pressurized water loop showed good dimensional stability for 10-mil-thick cladding for irradiations up to 1,600 MWD/T and heat fluxes up to 550,000 Btu/hr-ft²***

The reference clad material is Type 347 stainless steel, chosen for its strength in respect to the combined thermal and pressure stress in the fuel.

The design conditions specify a linear power generation of 18 kw/ft, 1,800 psi gas pressure, and 1250°F maximum clad temperature for a fuel diameter of 0.220" and clad O.D. of 0.250".****

H. Assembly and Quality Control

Tubing is inspected for cracks and inclusions, and pressure tested prior to pin fabrication. Welds on the fabricated pins are helium leak tested and the dimensional and straightness tolerances checked.

Fixtures are provided to bring the pins into proper relative positions to permit insertion into a channel can (spiral wire design) or into "eggcrate" spacers. Pins are secured by welding to end grids. The assembly jigs and procedure are designed so that no hand contact with the pin bundle is normally required for the assembly operations.

I. Fabrication Costs

Floor area for the fuel fabrication facility, equipment and personnel requirements for the flowsheet of Figure IV-1 are summarized in Table IV-1. Cost breakdown for core and axial and radial blankets for the 76,800 kg/yr rate are given in Table IV-2.

* J. M. Gerhart, J. N. Siltanen, and J. S. Cochran, "The Irradiation and Examination of PuO₂-UO₂ Fast Reactor Fuel", ASTM 64th Meeting, June 25-30, 1961; also J. M. Gerhart, "Post-Irradiation Examination of PuO₂-UO₂ Fast Reactor Fuel", GEAP-3833, November (1961).

** E. A. Lees, GEAP-3687, March 10, 1961.

*** C. N. Spalaris, GEAP-3786, July 1, 1961.

**** K. Horst, B. Hutchins, F. Leitz, and B. Wolfe, GEAP-3721-AEC, December (1961).

J. Fuel Fabrication Volumes Due to Growth of Capacity

In a growing nuclear power economy, a considerable part of fuel fabrication capacity is occupied by first-core fabrication in addition to refueling volume.

Fuel fabrication volume thus leads the level of power production by one to two years (while fuel reprocessing lags by a similar time interval).

After a reactor type begins to demonstrate economics which are competitive with some fraction (f) of the new power market, it will typically exhibit a percentage growth rate which is higher than percentage growth rate of the total power market. Percentage growth rates which are two to three times the rate of total power demand are to be expected until the installed capacity is 50 to 75% of (f) after which the percentage growth rate diminishes and approaches the percentage rate of the total power growth.

Where total power demand is doubling about every seven years, a new economic reactor type may be expected to double roughly every three years for the first 10 to 15 years before its growth rate diminishes due to saturation of its segment of the market.

The fuel residence time in the reference reactor core is closely equal to three years. Refueling the reactor requires fabrication of a total of one full core load every three years.

The fuel required to double installed capacity in three years is also about one full (additional) reactor core load in three years for each reactor operating at the beginning of the three year period.

For the first 10 to 15 years, the total fuel fabrication rate is therefore about twice the steady-state refueling rate of the reactor capacity which is generating power at any instant. This effect is important to recognize because it accelerates the attainment of the fuel fabrication volumes which are needed to get attractive unit fabrication costs.

On this basis, the effective fabrication capacity is for Cases I, II, and IA are accordingly 6,300, 3,150 and 1,050 MWe respectively, as shown in Table IV-3.

TABLE IV-1
FABRICATION PLANT COST BREAKDOWN
(For 76,800 Kg/Yr Rate)

	<u>\$/Yr</u>
Overhead Bldg. and Furnishings	- 60,220
Overhead Personnel	259,000
Miscellaneous O & M	35,400

Total Personnel - 152

TABLE IV-2
FUEL FABRICATION COSTS, CASE I
(For 76,800 Kg/Yr Rate)

	Core and Axial Blanket					Radial Blanket			Grand Total 130,300 rods/yr
	Plutonium to PuO ₂	PuO ₂ -UO ₂ Pellet	UO ₂ Axial Blanket Pellet	Encapsulation 90,700 rods/yr	Sub-Total	UO ₂ Pellet	Encapsulation 39,600 rods/yr	Sub-Total	
Rate kg U + Pu per year	3,840	19,380	14,880		35,100	42,500			76,800 kg/yr
Building	\$ 19,210	\$ 42,800	\$ 3,580	\$ 36,900	\$ 102,490	\$ 10,480	\$ 16,580	\$ 27,060	\$ 129,550
Other	8,930	19,985	1,664	17,150	47,640	4,872	7,707	12,579	60,220
Equipment	47,300	18,810	3,880	173,000	242,990	10,760	78,600	89,360	332,350
Personnel	67,000	195,000	16,200	447,000	725,200	69,900	345,000	407,900	1,133,100
Other	15,315	44,573	3,703	102,175	165,766	14,378	78,860	93,238	259,000
O & M	175,500	47,300	8,290	53,200	284,290	24,400	23,800	48,200	332,490
Other	18,685	5,036	883	5,664	30,268	2,598	2,534	5,132	35,400
G & A - 15%	52,791	56,010	5,730	125,263	239,800	19,558	82,962	102,520	342,314
Tubing			75,900	186,500	262,400		96,000	96,000	358,400
End Caps				111,800	111,800		87,600	87,600	199,400
Hardware				295,000	295,000		175,800	175,800	470,800
UO ₂ Depletion		40,600	70,200		110,800	201,000		201,000	311,800
Pu Loss at \$5.09/gm Pu	97,700	48,850		48,850	195,500				195,500
SHOP COST	502,431	518,874	190,030	1,602,502	2,813,940			1,345,380	4,160,324
Total Price = S.C. x 1.558*	\$782,787	\$808,406	\$296,066	\$2,496,700	\$4,384,118			\$2,096,102	\$6,481,785
\$/kg fuel	\$204/kg Pu Total Conversion & \$211/kg core fuel				\$133.90/kg U+Pu Total Fuel (Core + Axial Blanket)	\$49.30/kgU \$84.40/kg avg. (U+Pu) Grand Total			

$$\frac{6,481,785}{6 \times 525 \times 0.8 \times 8760} = .29 \frac{\text{mills}}{\text{kwhr}} \quad \text{Total Fabrication Costs}$$

* Includes complaints, contingency, working capital, development liquidation and profit.

TABLE IV-3

SUMMARY OF FABRICATION COST ESTIMATES

	CASE	<u>I</u>	<u>II</u>	<u>IA*</u>
Installed Capacity MWe		1,575	3,150	525
<u>Effective</u> Fabrication Rate, (MWe Steady-State)		3,150	6,300	1,050
Volume in Kg/Yr		76,800	153,600	25,600
<u>ANNUAL COSTS (in Thousands)</u>				
Buildings		190	308	190
Labor		1,392	2,420	578
O & M		368	560	191
G & A		342	575	173
Mat'ls		1,340	2,330	555
Pu Loss***		<u>196</u>	<u>392</u>	<u>81</u>
Shop Cost		4,160	7,125	1,966
Adders**		<u>2,322</u>	<u>3,980</u>	<u>1,098</u>
Total Annual Value		6,482	11,105	3,064
Average Fuel Fabrication Cost \$/Kg		\$84.40	\$72.40	\$119.50
Mills/KwHr		0.29	0.25	0.42

* Case IA uses the same building as Case I, but at lower production rate. See discussion of effective rate on page 35.

** Includes: Complaints, contingency, working capital, insurance and taxes, and profit.

*** Plutonium and uranium loss in fabrication taken at 1% at "1975 value" of \$5.00/gm (see Section V).

V. TOTAL FUEL CYCLE COSTS

The total fuel cycle cost is summarized in this section on the basis of the reference design shown in Table I-1. The assumptions and costs are essentially the same as those in the current reference design report* except for the addition of details of production volume dependence of fabrication and reprocessing costs.

A. Basis for Elements of Total Fuel Cycle Costs

1. Fuel Fabrication

Values are taken from Table IV-2. In determining the scale of operation allowance is made for fuel fabrication volume due to growth of installed capacity equal to the volume from refueling of operating reactors.

2. Reprocessing and Shipping

Reprocessing costs are taken from Table III-6. The minimum reprocessing capacity is designed to process 1,575 MWe. Costs for smaller volume are derived by operating the same plant at a smaller capacity. Shipping costs and losses are assumed to be independent of capacity and equal to \$13/kg of fuel for core plus axial blanket and \$10/kg of fuel for the radial blanket (of this \$3 is for losses). Annual cost for one 525 MWe reactor at 80% capacity factor is \$144,400 or 0.039 mills/kwhr.

3. Plutonium Inventory Charge

Inventory cost is applied to the initial value of plutonium loading in the core only. (See Table III-3 for loading and distribution of the production of plutonium; and Table II-2 for average isotopic compositions.) Pending AEC determination of buy-back prices, plutonium values are estimated on their minimum recycle fuel value. The lower value for 1975 assumes a corresponding reduction in the enriched uranium price scale.

TABLE V-1

ASSUMED MINIMUM PLUTONIUM VALUES

	<u>Dollars Per Gram</u>	
	<u>1963</u>	<u>~1975</u>
1. Fissile Pu	9.50	6.50
2. Non-Fissile Pu	1.50	1.10
3. U-235 (at 90%)	13.65	9.80

* K. Horst, B. Hutchins, F. Leitz, and B. Wolfe, "Core Design Study for a 500 MW Fast Oxide Reactor", GEAP-3721-AEC, December (1961).

TABLE V-1 (continued)

<u>AVERAGE VALUES</u>	<u>Dollars per Gram</u>	
	<u>1963</u>	<u>~1970</u>
4. 70% Fissile	7.10	4.90
5. 60% Fissile	6.30	4.30
6. 50% Fissile	5.50	3.80

- (4) Corresponds to first cycle thermal reactor output at about 15,000 MWD/T.
 (5) First cycle fast reactor output (average) (100,000 MWD/T).
 (6) Five-cycle fast reactor output (average).

Inventory charges are based on core inventory at the start of a cycle plus allowance for six months out-of-pile for processing and fabrication, plus 5% in spares for a total out-of-pile inventory which is 22% of the initial core loading of 1,890 kg (see Table III-5). For large volume operation, the spares allowance could be reduced. The 1975 inventory value is used for all cases in the cost summary except the 525 MWe core which uses the 1963 value.

TABLE V-2
INVENTORY COSTS

	<u>1963</u>	<u>1975</u>
Annual Rate %	4.75	10.
Average Pu Value \$/g	7.10	4.90
Inventory Cost Mills/KwHr (80% Load Factor)	.21	.31

4. First Core Capitalization

To start the reactor, fabrication cost of one load of fuel plus 5% spares must be incurred and capitalized. (37,720 kg + 5% from Table III-5)

The average fabrication costs of Table IV-2 are used for the dependence on fabrication volume. This corresponds to the cost of fabricating a new first core at the time at which there is existing capacity as shown.

TABLE V-3

FIRST CORE FABRICATION CAPITALIZATION

	<u>MWe Installed</u>		
	<u>525</u>	<u>1575</u>	<u>3150</u>
Average \$/kg Fabrication	119.50	84.40	72.40
Annual Cost at 14% (including 5% spares) mills/kwhr	0.18	0.13	0.11

5. Plutonium Credit

Excess fissile plutonium isotopes are produced as indicated in Table III-5, at a breeding ratio of 1.08. This corresponds to 110 kg of plutonium in 2.95 years or 37.3 kg/yr of fissile material.

TABLE V-4

PLUTONIUM CREDIT

	<u>1963</u>	<u>1975</u>
Fissile Value, \$/gm	9.50	6.50
Average % Fissile	60	50
Pu Credit, mills/kwhr	.096	.066

TABLE V-5

TOTAL FUEL CYCLE COST SUMMARY FOR REFERENCE REACTOR DESIGN

	<u>Operating Capacity Installed</u>		
	<u>525*</u>	<u>1575**</u>	<u>3150**</u>
1. Fuel Fabrication	.46	.33	.29
2. Reprocessing and Shipping	.27	.13	.10
3. Pu Inventory Charge	.21	.31	.31
4. First Core Capitalization	.18	.13	.11
5. Excess Plutonium Credit	(.10)	(.07)	(.07)
6. Mills/KwHr	1.02	.83	.74
Total Fuel Cycle plus First Core Capitalization			

* First cycle case (Case A in Table II-2). Fissile Pu value \$9.50/g; fuel use charge 4.75%.

** Recycle cases (Case B in Table II-2). Fissile Pu at \$6.50/g; fuel use charge at 10%.

APPENDIX A - Anion Exchange Flowsheet*

The anion exchange flowsheet for plutonium reactor fuels consists of the following steps: (all conducted at 60°C)

- a. Adsorption of plutonium on Permutit SK anion resin from a 7.2 M HNO_3 feed containing uranium, plutonium, and fission products. (Uranium and fission products are not held on the resin and go directly to waste. Depleted uranium has such a low value that recovery is not justified.)
- b. Decontamination of plutonium on the resin by a 7.2 M HNO_3 -0.01 M HF wash solution.
- c. Elution of plutonium by 0.6 M HNO_3 , yielding a solution of decontaminated plutonium at 60 gPu/l.
- d. Second cycle of decontamination by a repetition of steps a. - c. using a second set of ion exchange columns.

A. Fuel Dissolution

a. Chopping

The smaller diameter (0.25 inch) FCR fuel is compared to 0.5 inch diameter fuel would require some modification of the chopper design developed at Idaho Falls. The modification to achieve the required throughput is not considered unfeasible in view of successful experimental chopping work already done at KAPL on similar material. Delivery of chopped material to the dissolver could be identical to the Idaho design. The chopper developed at KAPL for small diameter pins is used to cut the fuel for leaching of oxide. (Similar devices for .5" rods are used in the "Idaho Small Plant" design.) Provisions for radioactive gas and particulate control would be the same as the Idaho plant.

b. Leaching

Nitric acid leaching of plutonium and uranium from irradiated chopped oxide fuel specimens has been confirmed in the FCR work at APED. The dissolver vessel must be geometry-controlled for criticality. If higher dissolution rates are desired than practical in a critically-safe cylindrical geometry, throughput could be increased by using a critically-safe slab.

* Flowsheet adapted from a study by H. W. Alter, March (1961).

FIGURE A-1 - ANION EXCHANGE FLOW DIAGRAM FOR THREE-FCR-PLANT CAPACITY (1575 MWe)

The dissolver is operated to produce a feed with 150 g U/l and 7.2 M HNO_3 . In-line instrumentation and control can eliminate the need for any but minor feed adjustment before ion exchange. Stainless steel ring waste would be disposed of to high-level waste tankage as in the Idaho scheme.

Dissolver product goes to critically-safe cylindrical run-tanks for sampling, minor acid adjustment and NO_2 treatment to stabilize plutonium (IV).

B. Adsorption

The maximum uranium feed concentration is 150 g/l since the solubility limit of uranium nitrate in 7 M HNO_3 is 275 g/l but high uranium concentration also decreases column capacity for plutonium adsorption. Column feed is specified to the composition shown below:

TABLE A-1

ANION RESIN FEED COMPOSITION

Uranium	150 g/l
Plutonium	8.9 g/l
Fission Products	5.8 g/l
HNO_3	7.2 M

For a throughput of Case I, 139.5 Kg (U + Pu + FP)/day, the total feed volume is 845 liters/day.

The number of resin columns required is set by the critically-safe size (6" diameter), the capacity of the column for plutonium, and the cycle time which is a function of feed, wash and elution flow rates. Total cycle time is less than 12 hours. Two cycles per day can be expected of a single column. (See Table A-2.) Two columns must then have a capacity per cycle of $7.53/4 = 1.88$ Kg Pu where the plant throughput is 7.53 Kg Pu/day.

Ryan and Wheelwright's work has shown that a column loading of 70 g Pu/liter of Permutit SK resin bed is feasible. Each of the two primary resin columns therefore requires a volume corresponding to a bed 6" in diameter and 5' long. Each primary column is followed by a 5' cleanup column to absorb the plutonium breakthrough of the fully-loaded primary column. The cleanup columns are washed but not eluted each cycle. They are periodically eluted after several cycles and their product recycled to the feed.

Since plutonium may be fed to a 6" column at 875 gm/hr, the adsorption time per cycle is $1.88/.875 = 2.15$ hours.

C. Wash

A 15 column-volume wash containing 7.2 M HNO_3 and 0.01 M HF is necessary to remove residual amounts of uranium and fission products. The linear flow rate is 20 ml/min-cm². Each primary column has a bed volume of 28 liters requiring a 410 liter wash per cycle. The washing time per cycle is 1.86 hours at the specified linear flow rate and the total throughput of wash for four daily cycles is 1,640 liters.

D. Elution

Plutonium product is eluted by a 0.6 M HNO_3 elutriant. The product cut is at 60 g Pu/l and the elution linear flow rate is held to 1 ml/min-cm². For each cycle, therefore the total volume of product solution is $1.88/.06 = 31.4$ liters. The elution time is 2.88 hours and the daily volume of product and elutriant is 127.6 liters.

E. Second Cycle

The second cycle flowsheet is identical to the first already described, except that uranium is absent and fission product activity levels are typically at least 2×10^4 lower than in the first cycle. The second cycle product can be expected to have an overall decontamination factor in excess of 4×10^8 . The plutonium at a product concentration of 60 g/l may be evaporated to yield a concentrated product or precipitated to make a low-fired oxide for shipping, or else used directly in a fabrication plant to produce mixed $\text{PuO}_2\text{-UO}_2$ powder.

F. Cycle Time

Times for conditioning and recycle together with analysis and makeup are approximate and conservative. Even so, the total cycle time of about ten hours will permit two daily cycles per column.

TABLE A-2
FOR ANION EXCHANGE FLOWSHEET CYCLE TIMES

	<u>Hours/Cycle</u>
Adsorption	2.15
Wash	1.86
Elution	2.88
Condition and recycle	1.00
Analysis and makeup	<u>2.00</u>
Total cycle time	9.89 hrs

G. Waste

The total hot liquid waste consists of NaNO_3 from the dissolver off-gas caustic scrubbers, stainless steel cladding waste, uranium, fission products and HNO_3 from the adsorption step and HNO_3 from the wash step.

It will be assumed that HNO_3 will be recovered by evaporation and recycle. The final concentrated waste will be assumed to have the following compositions:

TABLE A-3

NaNO_3	5 M
Uranium	300 g/l

Concentrated waste will have the following daily volume: (Case I),

	<u>Liters per Day</u>
Uranium	424 l
NaNO_3	57 l
Stainless steel	<u>5 l</u>
Total	486 l

Initial heat generation from fission products is at a level of about 45 watts/liter of waste at 30 day cooling. This high initial heat-generation rate will require more than usual attention to cooling requirements in waste tankage and to the problems introduced by radiolytic gas generation. Heat production in watts per liter will decrease only slowly with time because of long irradiation times used.

H. Radiation Damage of Resin

In the Hanford study Permutit SK resin was favored for use in high activity levels because of high resistance to radiation together with good capacity and mechanical properties. Based on cobalt-60 irradiations of the resin it was found that Permutit SK retains capacity and mechanical properties up to an exposure of 3×10^8 roentgens.

For the calculation of radiation damage to the resin, beta radiation is considered controlling. Only the radiation during the absorption step will be significant. A discharge value of .08 beta watts/gram of FCR fuel will yield a total of 13.3 beta watts/l of feed solution. Since the resin bed can be considered 50% void, exposure in the bed is 6.7 beta watts/liter of bed. The exposure level can therefore be approximated at 2.4×10^6 R/hour during the adsorption step. The alpha dose from .005 alpha watts/gram of fuel is 0.13 watts/liter of resin (average over resin) or 5×10^4 R/hr.

For an exposure of 3×10^8 R the service life of the two primary and two cleanup columns will be 26.6 days at two cycles per day. Ten batches of resin are required for the 270 day operating year. Annual resin consumption is therefore 40 cubic feet, representing only a small chemical and waste-storage cost.

Discarded resin is stored as high-level solid waste. Simplified plant design may favor disconnecting and replacement of entire resin columns including shell rather than remote emptying and loading of fixed columns with fresh resin.

A simple flow diagram for the anion exchange flowsheet is shown in Figure A-1. With the exception of daily throughput volumes to provide the capacity required for the reference case, and minor changes in concentrations, it is identical to one of the flowsheets shown by Ryan and Wheelwright.*

I. Waste Storage

High level liquid waste for Case I is generated at the rate of 486 liters/day of 34,600 gals/yr. The Idaho small plant assumed \$1.67/gal for liquid storage. In view of extra cooling problems \$2.00/gal will be assumed for anion exchange process waste.

Gas storage volume for the fast oxide Case I will cost about 8.8 times as large as for Dresden processing, storage cost is scaled from 180 MWe cost estimate of \$5,910/yr.

TABLE A-4

FCR PLANT ANNUAL WASTE STORAGE COSTS

	Case I <u>1575 MWe</u>	Case II <u>3150 MWe</u>
Gas	\$ 27,000	\$ 44,000
Liquid	69,000	112,000
Solid	<u>1,400</u>	<u>2,500</u>
Total Annual Cost	\$ 97,400	\$158,500

J. Product Losses

Replacement of the discarded uranium costs 0.003 mills/kwhr and is chargeable to fuel fabrication. FCR Case II amounts to 12,200 Kg uranium annually at an assumed cost of \$4.50/Kg (depleted material converted to oxide). The cost of replacing the uranium, about \$55,000/yr, is less than half of the usual cost of recovery and decontamination of recycled uranium.

* J. L. Ryan and E. J. Wheelwright, "The Recovery, Purification and Concentration of Plutonium by Anion Exchange in Nitric Acid", HW-55893 (DEL.), January 2, 1959.

Some product losses must be assumed for plutonium although by the nature of the batch anion process they will be low. A 0.2% loss is assumed for a total annual throughput of 2,050 Kg of plutonium, approximately 4,000 grams are lost. Depending on plutonium values (\$6.50 to \$9.50/gram fissile) this loss corresponds to .002 to .003 mills/kwhr.

APPENDIX B

OUTLINE OF DEVELOPMENT PROGRAMS FOR FCR FUEL FABRICATION AND REPROCESSING

1. Separations Process Study - Exploration of Process Variables
 - a. Gram-scale, trace beta and gamma dF, chemical impurities, resin damage.
(Simulate by $\beta + \gamma$ irradiation.)
 - b. Gram-scale, high level beta-gamma.
2. Dissolution
 - a. Small scale, heat treated but unirradiated samples.
 - b. Small scale, irradiated samples.
 - c. Pilot scale, unirradiated.
3. Pilot Plant Design Study and Optimization, Ion Exchange Process
4. Build and Operate Minimum-Scale Pilot Plant
5. Precipitation and Calcining

dF Available

Chemical Purity

Time and Temperature for Optimum Recovery

Recycle of Loss Streams
6. Plutonium Activity

Establish by calculation and measurement, definitive relationship of dose-rates to isotopic composition, geometry, and attenuation for convenient use in process design and layout.
7. Prepare Process Design for Fabrication Pilot Operation
8. Evaluate Unit Fabrication Processes for Optimum Balance of Cost, Handling, and Maintenance Requirements

9. Build and Operate a Minimum-Scale Pilot Fabrication Line
10. Fuel Assembly Processes; Design of Special Jigs and Fixtures for Minimum Handling and Maintenance Requirements
11. Quality-Control Instrumentation; Evaluate Inspection Procedures and Sensing Elements; Explore Measurements most Likely to Correlate with Fuel Performance and Survival