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COMPARATIVE COST STUDY OF THE PROCESSING OF OXIDE, CARBIDE, AND METAL FAST-BREEDER-REACTOR FUELS BY AQUEOUS, VOLATILITY, AND PYROCHEMICAL METHODS

by

M. Levenson, V. G. Trice, Jr.,
and W. J. Mecham

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Chemical Engineering Division

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I. INTRODUCTION

The economic importance of electric power production by nuclear power plants is of ever-increasing significance. The more rapidly the present economy of thermal reactors expands, the sooner will fast breeder reactors be important in the overall nuclear industry. Although the need for efficient breeding or converting of fissionable material in order to utilize fully our natural resources is well established, the question of the competitiveness of thermal and fast neutron reactors is an extremely complex one. The complexity arises from the many interrelated economic factors involved in the nuclear fuel cycles and the current variety of reactor types, fuel types, and fuel cycles. This study has not attempted to estimate the ratio of fast breeder reactors to thermal reactors, nor to select a "best" fuel or fuel cycle. The final evaluation of economic competitiveness between reactor types requires the development of realistic costs, not only of reactors, but also of fuel processing, fuel fabrication, fuel inventory, and all other associated cost items. For this study it was assumed only that fast breeder reactors would be of importance, and an attempt was then made to determine the potential relative costs of reprocessing oxide, carbide, or metal fuels by aqueous, pyrochemical, or fluoride-volatility technology. While technical feasibility has been demonstrated for some parts of each fuel cycle of the three fuels listed and of the three process technologies listed, complete demonstration has not been done for any process or any fuel cycle of the high-performance, high-burnup class, primarily because such spent fuels do not yet exist.

For the economic evaluation portion of this study, complete technical feasibility of both fuels and processes was assumed. Some of the major questions of process feasibility are listed in Appendix A. Factors such as design philosophy, plant location, or degree of conservatism can affect the costs of a fuel-reprocessing plant as much as or more than the fuel or process selected. Therefore, the numbers developed in any such cost study can only be compared to other numbers developed in the same study or to other numbers developed on exactly the same basis. The cost estimates presented in this report are quite reliable as to their relative values. The absolute values are reliable only if the same philosophies are used in plant design and

operation as were used in the evaluations. The conclusions apply only to a privately financed, privately owned industry wherein the cost of money is a real and significant item and wherein the ratio of risk capital to return is also important. Almost any reprocessing scheme can be made economically attractive on a unit-cost basis if the plant size selected is large enough, but risk capital may not be readily available for such large projects, so that total cost as well as unit costs are of importance in such evaluations.

A. Objectives

Fast breeder reactors are today in a very early stage of development. While several fast reactors are actually in operation today, none is the size of a central power station nor can they even be considered prototype plants. The eventual power economy for the fast breeder reactor will require the development of economically acceptable fuel cycles as well as the development of technically feasible power plants. The objective of this study was to determine the relative economic potential of the various fuel-reprocessing technologies as applied to several possible fast-reactor fuels. At this early stage, when fuel compositions, fuel burnups, fuel-recycle schemes, and all other aspects of the fuel cycle are not fixed, cost estimates of future fuel cycles are of limited value as absolute numbers. As a result, the primary objective of this study was to estimate and/or identify significant differences in the costs of processing fast-reactor plutonium oxide, carbide, or metal fuels by aqueous solvent-extraction, fluoride-volatility, or pyrochemical methods. The goal was evaluation of economic potential of alternative processes for a power economy of the 1980's, rather than an evaluation for a plant in the 1970's. Therefore, each process was represented (in a best-estimate effort) by the lowest-cost conceptual assemblage of processing steps for which technical feasibility has been demonstrated experimentally, or for which reduction to practice can be expected by 1980. None of the proposed plants could be built with confidence on existing information. Every effort has been made to perform the evaluations under a consistent set of requirements and restrictions, both technical and economic.

The capacity of the spent-fuel processing plant is an important parameter in obtaining an answer to the question of what process for what fuel. To explore this parameter, three plant sizes were considered for each of the three processes and three fuels: a close-coupled, on-site plant, serving a single 1000-MWe power station; a close-coupled, on-site plant, serving a three-reactor, 3000-MWe reactor park; and a central plant, serving ten reactors with an installed capacity of 10,000 MWe. The costs considered were reprocessing costs in a much broader definition than usual. Operating and capital costs of the reprocessing plant itself, costs for ultimate waste disposal, out-of-reactor fuel inventories, resynthesis of fuel material, and, in the case of the central plants, fuel shipping costs were included.

In addition to the general objective of determining the economic potential of various processes and plant sizes for various fast-reactor fuels,

an effort was made to identify the sources of significant cost differences and to define fruitful areas of research.

B. Ground Rules

In keeping with the objective of this study, the following common technical and economic ground rules were adopted for the calculation of the costs associated with the cases considered:

1. A common fuel-processing load was adopted as a reference specification for all cases (three fuels and three processes). This reference processing load is stated in Table I in terms of average mass rate of fuel

TABLE I: Specifications of the Reference 1000-MWe Fast Reactor
(All quantities stated as weight of elements)

Total Thermal Power, MWe	2500
Net Electrical Power, MWe	1000
Fraction of Total Power Generated in Core	0.85
Total Breeding Ratio	1.4
<u>Initial Composition of Fuel, Weight Percent on Metal Basis</u>	
<u>Core</u>	
Depleted Uranium	78.0
Plutonium (239, 241)	15.0
Plutonium (238, 240, 242)	7.0
<u>Blanket</u>	
Depleted Uranium	100.0
Core-fuel Burnup, Percent of Actinides	10.0
Blanket-fuel Burnup, Percent of Actinides	0.71
<u>Average Daily Rate of Fuel Discharge, kg per Full Power Day</u>	
<u>Core</u>	
Depleted Uranium	15.66
Plutonium (239, 241)	2.91
Plutonium (238, 240, 242)	1.56
Fission Products	2.24
<u>Blanket</u>	
Depleted Uranium	53.90
Plutonium (239)	1.053
Plutonium (240)	0.058
Fission Products	0.39
<u>Other Constituents</u>	
18.8 SS clad for Oxide and Carbide Fuel	12.0
V-20 w/o Ti Clad for Metal Fuel	12.0
1.78 w/o Ti as Alloy Constituent of Metal Core Fuel	0.405
Bond Sodium in Metal Core and Blanket	0.95

discharged from the reactor per day of full-power operation. The specifications in the table were selected as typical ones, in the light of recent conceptual designs of 1000-MWe ceramic- and metal-fueled fast reactors.^{1,2*} The average burnup of core fuel was specified as 10%. (At present, no clear superiority with respect to burnup for the three types of fuels has been established. Burnups of at least 6% appear to have been achieved for all three types of fuels in irradiation tests, and it appears reasonable to expect 10% burnup to be achievable in practice by 1980.)

2. A plant factor of 80% was adopted as the reference value for both reactor and processing-plant operation. This is in keeping with the usual commercial standards and with AEC guidelines.³

3. The product of the processes was the substance (oxide, carbide, or metal) of the reactor core or blanket in a form suitable as feed for fabrication operations without further chemical conversions.

4. In the reference processes, fission-product decontaminations were higher for the products of aqueous and volatility processing than for pyrochemical processing. In all processes, refabrication of the core fuel requires at least semiremote techniques because of the hard radiation from the fuel actinides themselves. Although an estimate of the costs of refabrication is beyond the scope of this study, a preliminary analysis of cost penalties for low-decontamination blanket processing is included in Appendix G.

5. Cladding was removed by chop-leach, chemical decladding, or mechanical decladding, depending upon which was most favorable to the respective reference processes.

6. The ultimate disposal of high-level radioactive waste as solids at a remote location was taken as the reference method. However, because of present uncertainty as to the cost of solidification of aqueous waste, its disposal costs were calculated on the basis of perpetual on-site storage of concentrated waste solutions, with the expectation that technical developments in aqueous-waste disposal would ultimately permit solid storage at a remote location at an equal cost.

7. To illustrate the relative magnitude of the cost of the processing loss, a processing loss of 1% was assumed for all cases.

8. The two cost elements of processing that were omitted from this study were (1) site acquisition and development costs and (2) liability insurance. The magnitude of these costs was not expected to be large, but more importantly, they were assumed to be matters of economic indifference in the comparison of alternate fuels and processes.

*References are listed at the end of each appropriate section.

9. The aqueous processes adopted in this study are of the generic type referred to as a chop-leach, solvent-extraction method. However, many simplifications and optimistic assumptions were incorporated in this aqueous-plant design compared to what is done in presently operating plants, particularly as regards the plutonium-handling steps. The uranium and plutonium outputs from the solvent-extraction system were converted to oxide in fluid-bed denitration and reduction units. For metal and carbide fuel, tail-end process steps were added to resynthesize the fuel material.

10. The pyrochemical processes for ceramic and metal fuels were adapted from the melt-refining and skull-reclamation processes developed for the Experimental Breeder Reactor II (EBR-II) Fuel Cycle Facility. (A newer process concept, originally developed for oxide fuel, termed the salt-transport process, holds promise for further cost reductions for all fuels. This concept was not developed sufficiently to be selected as the reference process for the cost estimates in the present study, but it is given preliminary consideration in Appendix B.)

11. The fluid-bed volatility processes selected use bromine fluorides (BrF_5 or BrF_3) as a selective fluorinating agent to separate volatile fluorides of uranium (UF_6) and fission products without converting plutonium into a volatile form. Elemental fluorine was used subsequently to form volatile plutonium hexafluoride (PuF_6), thereby effecting the separation of plutonium from both uranium and fission products. The process included the fluid-bed conversion of uranium and plutonium hexafluoride to oxides. The tail-end steps employed to yield carbide and metal products were the same as those used with the aqueous process.

12. For each fuel type, process type, and plant size, the investment in process equipment and building and in auxiliary facilities and spare equipment was estimated, based on requirements established for the flowsheets and processing specifications. Other investment items and the schedule of charges on investment were calculated as follows:

a. Twenty-five percent of the above investments in process plant and auxiliaries was added for construction overhead, including engineering. This overhead factor is based on the assumption that the plants as built are not the first of their type.

b. A one-time charge associated with plant startup was added in an amount equal to 25% of the total annual operating cost.

c. Working capital was added to the total investment as a non-depreciating item. It was charged as 5% of the process building and equipment (including auxiliary facilities), plus 20% of the total annual operating cost.

d. Annual charges on capital were calculated as the sum of two components: the return on capital and the repayment of capital. The return on capital was calculated as 13% of the total capital investment. The fixed charge rate of 13%/yr applied to the nondepreciated portions of the capital investment was taken from the AEC Guide to Nuclear Power Cost Evaluation³ and is based on the practices of investor-owned electric utilities. The components are as follows:

Minimum return	6.75
Property insurance	0.40
Federal income taxes	3.40
State and local taxes	<u>2.45</u>
	13.0%/yr

e. The out-of-reactor fuel inventory was charged as a separate nondepreciating item at the 13%/yr rate. The fuel values employed were \$10/g for the plutonium isotopes 239 and 241; values up to \$8/kg were assigned to other actinides.

f. The other component of capital cost (the repayment of capital) was calculated as the sum of annual charges on sinking funds at the 6.75%/yr interest rate to recover the original amounts of depreciating capital items over their economic lifetimes. The economic lifetimes were assigned as follows:

- (1) Process equipment operating in radioactive areas and for which decontamination was not considered practical: 2 yr.
- (2) Process equipment and mechanical equipment in radioactive areas maintained fully remotely or for which decontamination was practical: 5 yr.
- (3) Chemical and mechanical equipment in nonradioactive areas, including laboratory, office items, and mobile equipment: 15 yr.
- (4) Major building structures, including shielding windows in remote cells: 30 yr.
- (5) One-time charges associated with startup and construction overhead: 30 yr.
- (6) Spare processing equipment: 30 yr.

Capital items not included in the above capital-repayment assignment are plant working capital and out-of-reactor fuel inventory, which are nondepreciating items.

13. The annual operating costs of direct labor, indirect labor for maintenance and analytical laboratory operations, and direct materials (including reagents and utilities) were estimated, based on specific plant requirements. The above directly estimated labor requirements were established in man-hours per operating day. This man-hour requirement was then multiplied by 1.4 to obtain the number of man-hours per day for the actual payroll, which includes allowance for such benefits as vacation and sick leave. An average annual payroll cost per man of \$12,000 was used, which includes fringe benefits. Other items of operating cost were calculated as follows:

a. Supervision and indirect labor (other than that specified above) was taken as 25% of estimated direct labor.

b. The cost of maintenance materials was taken as 20% of the sum of (a) total labor costs and (b) the annual charge for capital replacement for process equipment and building items.

c. An annual overhead charge for administration was taken as 15% of the sum of all the operating costs above.

References for Section I

1. *An Evaluation of Four Design Studies of a 1000 MW(e) Ceramic Fueled Fast Breeder Reactor*, AEC Chicago Operations Office, COO-279 (Dec. 1, 1964).
2. L. E. Link, J. E. Ayer, K. A. Hub, W. J. Mecham, V. G. Trice, Jr., and J. T. Weils, *A Large Metal-Fueled Reactor Study. Part II. Core Design and Fuel Cycle*, presented at the Conference on Safety Fuels and Core Design in Large Fast Power Reactors, October 11-14, 1965, at Argonne National Laboratory.
3. Kaiser Engineers, *AEC Guide to Nuclear Power Cost Evaluation*, TID-7025 (1962).

II. SUMMARY AND CONCLUSIONS

An estimate was made of cost differentials among alternative cases in the processing of commercial fast-reactor fuels in facilities to be operated in about 1980. For each of three alternative fuel types, namely, uranium-plutonium oxide, carbide, and metal, flowsheets and plant designs were specified and capital and operating costs were estimated for three alternative types of processes: aqueous solvent extraction, fluid-bed volatility, and pyrochemical. The conceptual plant components for which the costs were estimated were the processing plant, ultimate waste disposal, analytical laboratories, auxiliary facilities, out-of-reactor fuel inventory, and shipping of the spent fuel. For each of these cost centers, three sizes of processing plant were considered: an on-site plant serving one 1000-MWe fast reactor, an on-site plant serving three 1000-MWe reactors, and a central processing plant serving ten 1000-MWe reactors.

Total annual costs were calculated for each alternative case as the sum of (a) the total annual operating cost, (b) the annual amount for capital repayment, and (c) the annual return on capital investment. The electrical unit costs in mills/kWe-hr were calculated from the total annual costs and the annual electric-energy production at 80% plant factor for the reference reactor power. The unit costs are summarized in Table II for the three fuel types, the three processes, and the three plant sizes, and are used as a basis of comparison.

TABLE II. Unit-Cost Summary
(Costs in mills/kWe-hr)

Fuel Type	Cost Component	Close-coupled 1000-MWe Plants			Close-coupled 3000-MWe Plants			Central 10,000-MWe Plants		
		Aqueous	Volatility	Pyrochemical	Aqueous	Volatility	Pyrochemical	Aqueous	Volatility	Pyrochemical
Oxide	Processing	0.53	0.35	0.44	0.29	0.21	0.25	0.16	0.14	0.18
	Waste disposal	0.15	0.09	0.09	0.12	0.08	0.08	0.10	0.08	0.07
	Inventory	0.17	0.12	0.12	0.11	0.06	0.06	0.109	0.079	0.079
	Shipping	-	-	-	-	-	-	0.050	0.066	0.071
	1% loss	0.016	0.016	0.016	0.016	0.016	0.016	0.016	0.016	0.016
	Total	0.87	0.58	0.67	0.54	0.37	0.41	0.44	0.38	0.42
Carbide	Processing	0.62	0.43	0.46	0.33	0.26	0.26	0.19	0.17	0.19
	Waste disposal	0.15	0.09	0.09	0.12	0.08	0.08	0.10	0.08	0.07
	Inventory	0.17	0.12	0.12	0.11	0.06	0.06	0.109	0.079	0.079
	Shipping	-	-	-	-	-	-	0.050	0.066	0.071
	1% loss	0.016	0.016	0.016	0.016	0.016	0.016	0.016	0.016	0.016
	Total	0.96	0.66	0.69	0.58	0.42	0.42	0.46	0.41	0.43
Metal	Processing	0.59	0.39	0.29	0.32	0.24	0.17	0.18	0.15	0.12
	Waste disposal	0.15	0.09	0.06	0.12	0.08	0.05	0.10	0.08	0.05
	Inventory	0.17	0.12	0.12	0.11	0.06	0.06	0.109	0.079	0.079
	Shipping	-	-	-	-	-	-	0.050	0.066	0.071
	1% loss	0.016	0.016	0.016	0.016	0.016	0.016	0.016	0.016	0.016
	Total	0.93	0.62	0.49	0.57	0.40	0.30	0.46	0.39	0.34

The estimates of unit costs shown in Table II for processing, waste disposal, and inventory, and for shipping of spent fuel, are those made in this study. The cost associated with the assumed 1% plutonium lost is shown to

indicate its relative magnitude. No attempt is made to make a cost evaluation of the three alternative fuel types. Such a comparison would have to include differences in reactor-performance breeding gains and in fuel-fabrication techniques. For example, if credits for bred plutonium were assigned to each fuel specifically, they would not be equal, but a somewhat higher credit would accrue to those fuels having higher breeding potential.

Because this study had the objective of elucidating cost differences and not predicting actual costs, the chief result is the quantitative estimate of the cost differences as provided by Table III, wherein the differences of the sums of the cost of processing, waste disposal, and out-of-reactor fuel inventory are presented in mills/kWe-hr. Also presented in the table are the authors' estimates of the uncertainty of the cost differences.

TABLE III. Summary of Unit-cost Differences between Pairs of Alternative Processes

Note: 1. Cost differences shown may be converted to \$/kg by multiplying by 308.

2. The estimated cost differences, with the confidence limits shown in this table, are the authors' estimate of the interval in which the true difference in cost lies.^a

Fuel Type	Process Pairs Evaluated Process A Minus Process B		Unit-cost Difference ^b for the Sums of Processing and Waste Disposal and Inventory, mills/kWe-hr		
			Close-coupled One Reactor 1000 MWe	Close-coupled, Three Reactors Totaling 3000 MWe	Central Plant, ^c Ten Reactors Totaling 10,000 MWe
Metal	Aqueous	Volatility	0.319 ± 0.133	0.174 ± 0.082	0.067 ± 0.055
	Aqueous	Pyrochemical	0.445 ± 0.124	0.272 ± 0.074	0.118 ± 0.050
	Volatility	Pyrochemical	0.125 ± 0.090	0.097 ± 0.059	0.051 ± 0.043
Oxide	Aqueous	Volatility	0.304 ± 0.121	0.169 ± 0.075	0.066 ± 0.051
	Aqueous	Pyrochemical	0.206 ± 0.130	0.129 ± 0.079	0.016 ± 0.055 ^d
	Pyrochemical	Volatility	0.098 ± 0.103 ^d	0.040 ± 0.066 ^d	0.050 ± 0.050 ^d
Carbide	Aqueous	Volatility	0.310 ± 0.139	0.166 ± 0.086	0.061 ± 0.057
	Aqueous	Pyrochemical	0.279 ± 0.141	0.167 ± 0.085	0.035 ± 0.059 ^d
	Pyrochemical	Volatility	0.031 ± 0.112 ^d	-0.001 ± 0.072 ^d	0.025 ± 0.054 ^d

^aThe estimated limits in this study were based on the statistics presented by W. T. Nichols, Capital Cost Estimating, Ind. Eng. Chem. 43(10): 2295-9 (1951).

^bUnit cost of Process A minus unit cost of Process B.

^cDifference presented includes shipping costs for central plant only.

^dThe confidence interval for the cost difference included zero; therefore, no estimate of the cost difference is presented. Under the arbitrary rules applied in this study, the difference is deemed to be not significant.

From the differences of costs presented in Table III, it is concluded that both pyrochemical and volatility processing are significantly more economical than aqueous processing for all fuel types for the two sizes of close-coupled processing plants. For the large central plant, the cost estimates also showed that the nonaqueous processes were more economical than aqueous processes for all fuels, but the unit-cost differences were smaller.

For oxide and carbide fuel, the cost differentials indicated that both volatility and pyrochemical processing were more economical than aqueous processing for the close-coupled plants. For the large central plant, volatility showed a significant cost advantage over aqueous processing. The cost differences between volatility and pyrochemical processing were not significant for the ceramic fuels.

For metal fuels, the greatest cost advantage was shown by pyrochemical processing for all three sizes of plants. This cost difference of pyrochemical processing, with respect to both aqueous and volatility processing, was significant for all plant sizes. Volatility processing was significantly more economical than aqueous processing for plants of all sizes.

Relative process costs are compared in Table IV where the sums of unit costs for processing, waste disposal, out-of-reactor fuel inventory, and shipping of spent fuel are shown as ratios for the cases of this study.

TABLE IV. Estimated Costs of Pyrochemical and Volatility Processing, Relative to Those for Aqueous Processing^a

Fuel Type	Identification of Process Ratios	Ratios of Unit Costs		
		Close-coupled, One Reactor 1000 MWe	Close-coupled, Three Reactors Totaling 3000 MWe	Central Plant, Ten Reactors Totaling 10,000 MWe
Oxide	Volatility/Aqueous	0.64	0.68	0.84
	Pyrochemical/Aqueous	0.76	0.75	0.95
Carbide	Volatility/Aqueous	0.67	0.71	0.90
	Pyrochemical/Aqueous	0.70	0.71	0.95
Metal	Volatility/Aqueous	0.65	0.69	0.88
	Pyrochemical/Aqueous	0.52	0.51	0.73

^aThe estimated cost considered is the sum of the unit costs for processing, waste disposal, inventory, and shipping.

Figure 1 shows, for each fuel type and process, the combined unit cost for processing, waste disposal, out-of-reactor fuel inventory, and shipping of spent fuel as a function of the number of reactors served. The shipping charge is applied to the central plant but not to the close-coupled plants; therefore, the comparison of unit costs between the 10-reactor and the smaller plants does not show an effect of scale only. The estimated plant investment for processing and waste-handling facilities is also shown in Fig. 1. The investment amounts include auxiliary facilities, but not land or site development. Fuel-shipping charges and ultimate waste-burial charges were not capitalized, but were included in operating costs. (Further details are presented in the statement of ground rules in Section I.B.)

Fig. 1. Summary of Estimated Unit Costs and Plant Investment for Combined Processing and Waste Disposal. (Unit cost also includes charges for out-of-reactor fuel inventory and, for the central plants, charges for shipping of spent fuel.

In the cases shown in Fig. 1, a reduction of about 40% of unit costs was found for the increase of size from one close-coupled reactor to three close-coupled reactors. This result appeared for all types of processing and for all fuels. For the further increase in size accompanying the transition to the central, ten-reactor plant, the unit costs of the aqueous plant incurred a reduction of about 20%, a proportional reduction less than that in the previous threefold increase in size for the close-coupled reactors. For the sum of the cost components shown in the figure, the volatility and pyrochemical plants showed essentially no decrease or, in some cases, an increase in unit costs for the transition from the three- to ten-reactor plants. This result for the nonaqueous plants was due to the higher costs of shipping of shorter-cooled fuel. The shipping charges for all cases included estimates of property and liability insurance. However, no attempt was made to evaluate the effects upon costs of possibly more stringent future shipping regulations.

For each fuel type shown in Fig. 1, the results indicate a comparison between close-coupled and central processing plants, as follows:

Aqueous processing is not competitive with volatility or pyrochemical processing for on-site plants of the sizes considered in this study. If aqueous processing were chosen, the promising application would be only for quite large central plants. However, economic incentives are small for building a large central plant, since nonaqueous processing plants of about one-fourth the size offer the same unit costs for a much lower required investment.

The proper effect of throughput on processing and waste-disposal costs (in the absence of inventory and shipping charges) is indicated in Figs. 2, 3, and 4, in which processing (only) and waste-disposal costs are presented separately for the three fuel types.

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Fig. 2. Estimated Unit Costs for Processing Oxide Fuel as a Function of Plant Throughput from One to Ten 1000-MWe Fast Reactors

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Fig. 3. Estimated Unit Costs for Processing Carbide Fuel as a Function of Plant Throughput from One to Ten 1000-MWe Fast Reactors

108-9344 Rev. 2

Fig. 4. Estimated Unit Costs for Processing Metal Fuel as a Function of Plant Throughput from One to Ten 1000-MWe Fast Reactors

For the unit costs of processing (only) shown in Figs. 2, 3, and 4, the economics of scale are generally similar for the three processes. However, the reference pyrochemical process employs strictly batch operations for all its process steps and therefore shows a lower economy of scale for the large central processing plant than either aqueous or volatility processing, which employ continuous or semicontinuous process steps for major portions of their flowsheet operations.

For the waste-disposal unit costs shown in Figs. 2, 3, and 4, there is in all cases only a small economy of scale because costs correlate closely with the quantity of fission products. The somewhat greater economy of scale shown for the aqueous process is due to the component of cost for waste-solution evaporators that do have an economy of scale.

The cost estimates were made in sufficient detail that the sources of the cost differentials could be identified for the alternative processes. The basic source of cost advantage of the nonaqueous processes was due to the fewer process operations and the more compact processing equipment. This effect had several sources: (1) the much smaller amount or absence of diluents in fuel processing, (2) the simpler accommodation of fuel of high fissionable content and high burnup, owing to the absence of reagents offering neutron moderation and suffering irradiation damage, and (3) the direct process yield of solid actinide products and of concentrated solid radioactive waste. Substantial savings in the cost of shielded process cells and buildings as well as in process equipment accrued to the more compact equipment.

Significant savings were also found for the nonaqueous processes in the charges for out-of-reactor inventory. For the purely batch operations that are characteristic of the pyrochemical process, the low inventory costs were due to the short cooling time and overall rapid processing schedule. For the volatility process, the short cooling time and the relatively small holdup of material in the processing equipment allowed similarly low inventory charges.

None of the conceptual processes adopted in this study has been completely demonstrated for irradiated fast-reactor fuel, although in each case technically feasible steps were selected on the basis of laboratory and pilot-plant results and for which further development could be expected to allow practical use by about 1980. In the process critique and selection of process steps for the three processes, more difficulty was encountered with the less-mature nonaqueous processes in settling upon the particular reference designs for the cost study. This was so, especially for the pyrochemical process, because newer process steps have come into consideration, which have promise of further cost advantage, but which had not been developed to the point of allowing cost estimation in the present study. As a result, the present study does not rigorously adhere to the objective of consistent requirements and restrictions for all cases. If the same degree of conservatism were to be applied to the aqueous processes as was applied to the nonaqueous processes, the differentials would be even larger in all cases studied.

Perhaps the major conclusion derived from the study is that on-site, nonaqueous, reprocessing plants requiring relatively small amounts of risk capital are probably capable of processing spent fast-reactor fuel at unit costs very little, if any, higher than those achievable in larger central plants--no matter what process is used in the central plant.

III. CHEMICAL PROCESS FLOWSHEETS

The essential function of a fuel cycle facility is to restore irradiated fuel to a form suitable for its return to the reactor. For fast-reactor fuel the reprocessing requirements for restoration of the fuel are partial fission-product removal to repair irradiation damage and reconstitution of the heavy atom and inert alloying constituents. This section describes the processes used as the basis for the estimation of processing and waste-disposal costs. Process critiques, in which alternative processing steps and questions of feasibility are discussed appear in Appendix A. In this study it was found that no process has been fully developed that satisfies the requirements for fuel cycles utilizing uranium-20 w/o plutonium irradiated to 100,000 MWD/metric ton. The processes presented should be regarded as representing plausible extensions of the present state of the art of reactor-fuel processing.

A. Aqueous Processing

The aqueous processes adopted in this study as a basis for developing cost estimates of processing oxide, carbide, and metal fast-reactor fuels are of the generic type referred to as the chop-leach, solvent-extraction method. However, the aqueous processes currently employed in the processing of nuclear fuels cannot directly satisfy the processing requirements of the fast-reactor fuels considered in this study, principally because of the high concentrations of fission products and plutonium in these fuels. A concept for an advanced aqueous process for fast-reactor fuels is presented here based on a limited quantity of experimental data. These data have been reported largely for unirradiated fuel and in no case for fuels irradiated to burnups close to the reference amount of 100,000 MWD/metric ton. The flowsheet has not been proven in laboratory or pilot-plant evaluation. Areas of questionable feasibility exist in the conceptual advanced aqueous process for fast-reactor fuels, starting with the dissolution of the fuel. In the following discussion, the main features of aqueous processing are presented in terms of the requirements for oxide fuels, and appropriate tail-end process steps are subsequently described for the metal and carbide fuels. The head-end treatment required for carbide fuels is described separately. The metal fuel does not require a separate head-end step, but employs the same chop-leach concept as the oxide fuel.

The chemical steps in the aqueous process are illustrated schematically in Fig. 5. The functional description of these steps is as follows:

1. Dissolution of the fuel by acid-leaching of chopped fuel elements.
2. Decontamination by solvent extraction to achieve the gross separation of fission products.
3. Partition and stripping by solvent extraction to recover uranium and plutonium.

4. Further decontamination of the uranium product in a third solvent-extraction cycle.
5. Fluid-bed denitration of the separated aqueous products to convert them to $(U,Pu)O_2$ and UO_2 for reuse as core fuel and blanket, respectively.

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Fig. 5. Principal Steps in the Advanced Aqueous Process for Oxide, Carbide, and Metal Fuels

In the aqueous process, core fuel and blanket are processed simultaneously. The fuel and blanket are dissolved in a recirculation-type dissolver. The composition of this solution is adjusted to provide a first-cycle feed containing 140.5 g/liter of U, 8.0 g/liter of Pu (239 + 241), and 3.27 g/liter of Pu (240 + 241). The actinide concentrations in these solutions are lower than in conventional aqueous processing for thermal-reactor fuels where uranium is the principal fissile material. The principal reasons for the limitation on feed composition are to provide for nuclear safety and to avoid the precipitation of fission products in the co-decontamination cycle.

In the co-decontamination cycle, uranium and plutonium are extracted with a solvent consisting of 20% tributyl phosphate in kerosene. Because of its low cost, kerosene was selected for the diluent despite its

relatively high susceptibility to radiation damage (as compared to dodecane, for example). The concentration of degradation products in the solvent is kept at an acceptably low level by scrubbing the solvent with aqueous sodium carbonate solutions and by discarding 10% of the effluent solvent from the co-decontamination cycle.

Partitioning of uranium and plutonium requires the adjustment of the plutonium valency and its maintenance as Pu(III). Ferrous ion is usually employed for this purpose; however, because of the high plutonium content of fast-reactor fuel, its use in the conceptual aqueous process would lead to unacceptably high levels of iron contamination in the products. Therefore, uranous ion is used as a reductant. The resulting plutonium product contains about one part of uranium per 100 parts of plutonium. This level of contamination is well within acceptable limits because the separated plutonium product has to be reconstituted to the core-fuel composition by the addition of uranium.

It is assumed that the radiation hazard from actinide isotopes in recycled plutonium will be such that at least semiremote techniques will be required for fabrication. Consequently, there is no economic justification for further fission-product decontamination of the plutonium product by either solvent extraction or ion exchange. The uranium product, on the other hand, once it is separated from plutonium, fully decontaminated from fission products, and stored for about 60 days to permit the decay of ^{237}U , can probably be handled in refabrication operations by direct-contact methods as long as the daughters of the ^{232}U daughter of ^{236}Pu are kept to a minimum. Therefore, the uranium solution from the partition step is subjected to a third cycle of solvent extraction and passage through a column of silica gel to yield a fully decontaminated product.

The aqueous products of solvent extraction are converted to oxides by fluid-bed denitration (calcination) followed by fluid-bed reduction with hydrogen. In experiments with uranyl nitrate solutions, these operations yielded a dense granular material suitable for fabrication into fuel elements by vibratory compaction. The product for the fabrication of blanket elements is uranium dioxide. The product for the core fuel formed by this procedure is probably a solid-solution uranium-plutonium dioxide; however, this has not been verified experimentally.

B. Head-end and Tail-end Conversion Processes

1. Head-end Process for Carbide Fuels

Carbide fuels are not acceptable feeds for either aqueous or pyrochemical processing. In aqueous processing, carbides can be dissolved but the resultant feed solutions have unsatisfactory solvent-extraction

characteristics; i.e., stable emulsions usually form in the extraction columns. In pyrochemical processing, the presence of carbon results in product contamination and excessive plutonium losses. The head-end step for carbide fuels consists of their conversion to oxides. This is accomplished in the course of the oxidative decladding step. Hence, only nominal increased investment is required in head-end facilities to accommodate carbide fuels.

2. Tail-end Processes

Oxides are the product of the basic aqueous and volatility processes. Tail-end processes are required to convert the oxides to carbide or metal refabrication feed materials. The metal product of pyrochemical processing requires tail-end processes to convert the metallic alloys to oxides or carbides.

a. Conversion of Oxide to Carbide and to Metal (Volatility and Aqueous Processing). The estimate of the cost of converting oxide to carbide was based on the processing sequence shown in Fig. 6. A metal product is an intermediate step in the processing sequence. The processing steps are as follows:

(1) Uranium-plutonium oxide is reduced in a beryllia crucible with magnesium in a zinc solution at 800°C. A salt phase is employed to suspend the magnesium oxide formed. At the completion of the reduction, the salt phase is withdrawn from the crucible, filtered to remove the magnesium dioxide, and then recycled. The product is washed with fresh salt to remove completely all traces of magnesium dioxide. The solvent metal, zinc-50 w/o magnesium, is loaded beyond the solubility limit; therefore the uranium precipitates. Plutonium remains in solution in the solvent metal. The actinide loading of the solvent metal in this step is about 20 w/o.

(2) The crucible is transferred from the reduction furnace to a retort, in which the solvent metal and residual salt are removed by volatilization. A metallic product, which is the refabrication feed material for metal fuel, is obtained in this step.

(3) The metallic product is converted to a powder by hydriding at 300°C.

(4) The metallic powder is converted to carbide by carburization with a methane-hydrogen mixture at 750°C at about 5 atm pressure. This conversion is accomplished in a fluid-bed reactor. The carbon content of the product carbide is controlled principally by adjustment of the hydrogen/carbon ratio in the reagent gas.

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Fig. 6. Pyrochemical Process for the Conversion of Oxide to Metal and Fluid-bed Synthesis of Carbide

b. Conversion of Metal to Carbide and to Oxide (Pyrochemical Processing). The process steps for the conversion of metal to carbide are described in Items (3) and (4) above. To produce oxide, the metallic retort product is oxidized with oxygen and reduced with hydrogen to yield UO_2 for blanket refabrication and $(\text{U,Pu})\text{O}_2$ for core refabrication.

C. Pyrochemical Processing

1. General Description

Pyrochemical processing* refers to a variety of high-temperature, nonaqueous procedures for the reprocessing of both ceramic and metallic fuels. The pyrochemical processes that are evaluated in this study employ liquid metals and molten salts as process media, and the basic separations are effected at about 600 to 1300°C. Because of the radiation stability of liquid metals and salts, these processes can accommodate short-cooled, high-burnup fuels. They utilize simple batch separations that are not designed to provide the high fission-product decontamination factors associated with aqueous-solvent-extraction and fluoride-volatility processes. Their objectives are to achieve sufficient fission-product removal to restore the nuclear and mechanical properties of the fuel and to permit rapid recycle of the fuel in order to minimize the out-of-reactor fuel inventory. The limited degree of fission-product decontamination requires that both the core and blanket assemblies be refabricated remotely. However, the indications

*The frequently used term "pyrometallurgical" processing is more restrictive in that it refers specifically to processes for metallic fuels in which the bulk constituents, e.g., uranium and plutonium, remain in the metallic state throughout the process.

are that extensively recycled fuel from a fast breeder reactor may have to be refabricated at least semiremotelly, regardless of the fission-product decontamination, because of the buildup of radioactive uranium and plutonium isotopes.

Until recently, the development of pyrochemical processes at Argonne National Laboratory has been concentrated on the fuel-cycle requirements of the metal-fueled EBR-II. Although the melt-refining process is being used routinely to process the enriched uranium core fuel of EBR-II, it is not suitable for a complete uranium-plutonium fuel cycle. Therefore, the cost estimates in this study were based upon a considerably different process flowsheet. The individual chemical-separation steps in this flowsheet have been demonstrated, and the major engineering problems have been solved during the course of development work on processes for the recovery of plutonium from the EBR-II blanket and the recovery of uranium from melt-refining residues (oxides and skulls). However, some scale-up of these operations would be required, particularly in the retorting step in which uranium and plutonium are recovered by the volatilization of liquid-metal solvents.

The pyrochemical process used for the cost studies on oxide and carbide fuels is not as well established as the process for metal fuels. The pyrochemical process for oxide fuels was selected because the major engineering problems have been worked out during the development of the EBR-II skull-reclamation process, which utilizes similar separation steps. To provide for the recovery of plutonium and its separation from fission products, a multistage, countercurrent extraction column is included in the process for oxide and carbide fuels. Laboratory studies have shown that 99.9% plutonium recovery can be obtained in six theoretical stages of a liquid metal-molten salt column of this type, and pilot-plant investigations have indicated that the height equivalent to a theoretical stage is about 1.5 ft. However, multistage extraction equipment suitable for use in a pyrochemical processing plant has not been developed. The development of equipment for multistage extraction and for large-scale retorting is the major area in which the engineering technology is lacking in the process described for oxide and carbide fuels.

A new pyrochemical process, which uses a salt-transport technique for the basic separations, shows promise for the reprocessing of the three fuel materials considered in this study. For oxide and carbide fuels, in particular, this more advanced type of pyrochemical process offers the possibility of substantial reductions in fuel-reprocessing costs. This process also provides higher fission-product decontamination factors and utilizes more compact equipment. The application of this type of process to oxide and carbide fuels is described and a preliminary cost estimate is presented in Appendix B. The chemical feasibility of this process has been investigated in laboratory studies; however, engineering studies have begun only recently. Therefore, this process was not selected for formal cost comparison with the more fully developed aqueous and volatility processes.

2. Reference Pyrochemical Process for Metal Fuels

The principal steps of the pyrochemical process for metal fuels that was adopted as the reference process are shown in Fig. 7. The feed to the process consists of separate batches of discharged core and blanket materials, which have been declad mechanically. The following six major processing steps are involved:

a. The core fuel* is melted in a beryllia crucible and contacted with a molten mixture of calcium chloride and magnesium chloride at 1250°C. The rare-earth, alkali-metal, and alkaline-earth fission products are selectively oxidized by the magnesium chloride and extracted into the salt phase, which is discarded as waste. The volatile fission products are released to the off-gas system.

b. The metal phase is next contacted with cadmium chloride-sodium chloride-potassium chloride mixture in a tungsten vessel at 650°C. Uranium and plutonium are converted to their respective chlorides and extracted into the salt phase. The noble-metal fission products remain in the cadmium metal produced by the reaction. This metal phase is discarded as waste.

c. The resulting salt phase, which contains the uranium and plutonium chlorides, is charged to a separate stainless-steel vessel and contacted at 650°C with a liquid cadmium-zinc-magnesium alloy in which the declad radial-blanket fuel has been dissolved directly. The uranium and plutonium chlorides are reduced to the metals by the magnesium in this alloy. The alkali-metal and alkaline-earth fission products from the radial-blanket material are extracted into the salt, which is discarded as waste. The volatile fission products are released to the off-gas system. The rare-earth and noble-metal fission products from the blanket fuel are not removed.

Fig. 7

Principal Steps in the
Advanced Pyrochemical
Process for Metal Fuels

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*For the purpose of this flowsheet, the axial blanket fuel is also included in the core processes.

d. The metal phase from Step c is charged to a beryllia crucible and retorted to remove most of the cadmium and zinc. The uranium precipitates in the remaining magnesium-rich, liquid-metal solution. The plutonium remains in solution in the supernatant liquid metal, which is transferred to a separate beryllia retorting crucible.

e. The uranium product is retorted further to remove the residual solvent metals and melted to form a product ingot. The solvent metals are condensed and recycled to Step c.

f. The magnesium-rich, liquid-metal solution from Step d is retorted to remove the magnesium, and the plutonium product is recovered as a metallic ingot. The magnesium-rich solvent metal is condensed and recycled to Step c.

This process makes no provision for the routine separation of niobium and zirconium from the fuel because it may be desirable to retain refractory metals in the fuel alloy. Refractory metals are currently favored as an alloying constituent in metal fuel as a means of increasing its melting point. Although titanium is regarded as the most promising additive metal, niobium and zirconium are potential alternatives. These three metals show similar chemical behavior in this process.

After the fuel has been cycled through the reactor a number of times, it may be desirable to decrease its refractory metal content. Titanium, zirconium, and niobium can be removed by chlorination with cadmium chloride at 650°C in the presence of a molten-chloride-salt phase. Under these conditions, the chlorides of these refractory metals are volatilized and collected as solid waste in traps in the off-gas line. This procedure would be applied to the salt phase resulting from Step b after separation of the salt from the waste metal.

In the pyrochemical process for metal fuels, the fission-product removals are expected to be approximately as follows:

Volatiles (Xe, Kr, Te, I)	100%
Alkali metals and alkaline earths (Rb, Cs, Sr, Ba)	99%
Rare earths and yttrium	95%
Noble metals (Mo, Te, Ru, Rh, Pd, Ag, Cd, In, Sn, Sb)	95%
Refractory metals (Nb, Zr)	nil

3. Reference Pyrochemical Process for Oxide and Carbide Fuels

The principal steps of the pyrochemical process for oxide and carbide fuels are shown in Fig. 8. For core fuel irradiated to 10% burnup and blanket material in which the bred plutonium content has reached 2%,

the fission-product content of the core fuel is about 15 times that of the blanket material. Therefore, there is little incentive to decontaminate the blanket material routinely in a pyrochemical process. Consequently, the core and blanket material may be processed separately.

108-9339

Fig. 8. Reference Pyrochemical Process for Oxide and Carbide Fuels

The following six major steps are involved in the core process:

a. An oxidative decladding procedure is used for both oxide and carbide fuel. For both fuels, the product of this step is a finely divided oxide mixture that is separated mechanically from the cladding fragments. The volatile fission products are released during this step and are collected in the off-gas waste stream.

b. The mixed oxides are suspended in a molten chloride salt, which is contacted with molten zinc at 750°C in a tungsten crucible. The noble metals are selectively reduced and extracted into the zinc phase, which is discarded as waste.

c. The molten chloride salt containing the mixed oxides is contacted with a liquid magnesium-zinc alloy at 750°C. The uranium and plutonium are reduced to the metals, leaving the alkali-metal and alkaline-earth fission products in the salt phase. This salt, which also contains solid magnesium oxide formed during the reduction, is discarded as waste. At the completion of the reduction step, the uranium-metal product is present as a precipitate, while the plutonium remains in solution in the supernatant

liquid metal. The uranium is dissolved in additional liquid metal and transferred to a surge tank. The liquid-metal solvent in the surge tank is a cadmium-zinc-magnesium alloy, which can be contained in stainless steel.

d. The rare-earth fission products are separated from the uranium-plutonium solution by molten chloride extraction in a 15-ft column having six theoretical stages. The effluent salt phase is subsequently contacted with liquid aluminum-magnesium alloy, which back-extracts the rare earths, thereby permitting recycle of the salt. The aluminum-magnesium alloy containing the rare earths is discarded as waste.

c. The liquid-metal solution of uranium and plutonium from Step d is retorted to vaporize the solvent metals, which are condensed and recycled.

f. In the case of oxide fuel, the uranium-plutonium product is reoxidized with oxygen, followed by reduction with hydrogen, to yield a dense, granular product. In the carbide process, the metallic product from the retorting step is first hydrided to form a powder, then treated with a hydrogen-methane mixture to regenerate the carbide. These product-conversion steps are discussed in greater detail in Appendix A.

The first two steps of the blanket process for oxide and carbide fuels are identical to Steps a and c of the core process. However, at the completion of Step c, the supernatant liquid-metal solution containing the plutonium is combined with the liquid-metal product stream from the core process as it enters the retorting step. The precipitated uranium is redissolved and transferred to a uranium-retorting step, which is followed by the product-conversion procedures indicated in Step f of the core process.

The net fission-product removals expected for the combined core and blanket processes for oxide and carbide fuels are expected to be approximately as follows:

Volatiles	100%
Alkali metals and alkaline earths	95%
Rare earths and yttrium	90%
Noble metals	85%
Refractory metals	10%

The routine removal of zirconium and niobium is not provided. Their buildup in the product can be controlled by periodically chlorinating a portion of the metallic product from the retorting step and separating them as volatile chlorides, as described in the process for metal fuels.

D. Fluid-bed Fluoride-volatility Processing of Fast-breeder-reactor Fuels

To provide a basis for the cost estimates, reference flowsheets were adopted for the fluid-bed fluoride-volatility processing of the three types of fast-breeder-reactor fuels, namely, oxide, carbide, and metal fuels. The reference flowsheets constitute an assemblage of individual process steps having a high degree of feasibility on the basis of laboratory and pilot-plant experience, although demonstration is far from complete. Because the important steps of the volatility process are less well known than in the case of aqueous processing, somewhat more detail is required in the description to make clear the process applications. Further details, references, and a process critique are presented in Appendix A.

The generic features of fluoride-volatility processes consist in the formation of the volatile hexafluorides of uranium and plutonium and in the use of volatility-separations methods to effect fission-product decontamination of the uranium and plutonium hexafluorides. Fluid-bed techniques are employed in the fluorination and in other steps involving solid-gas reactions because of the desirably high rates of heat transfer attainable in fluid beds, and because of the process convenience that results from the ability to transport the fluidizable solids through pipes.

Uranium and plutonium can be separated in the volatility process by several methods. For this study, the method selected is based on the ability of certain fluorinating agents to convert uranium to volatile uranium hexafluoride while plutonium is converted to nonvolatile plutonium tetrafluoride. The latter compound is then converted, in a separate step, to volatile plutonium hexafluoride.

Fluid-bed volatility processing, as applied in this study to the three fast-reactor fuel types, is illustrated in Figs. 9, 10, and 11 for uranium-plutonium oxide, carbide, and metal fuels, respectively. For all three fuels,

108-9437 Rev. 1

Fig. 9. Simplified Flow Diagram for Fluid-bed Fluoride-volatility Processing of Oxide Core and Blanket from a Fast Reactor

108-9436 Rev. 2

Fig. 10. Simplified Flow Diagram for Fluid-bed Fluoride-volatility Processing of Carbide Core and Blanket from a Fast Reactor

108-9438 Rev. 1

Fig. 11. Simplified Flow Diagram for Fluid-bed Fluoride-volatility Processing of Metal Core and Blanket from a Fast Reactor

uranium hexafluoride and plutonium hexafluoride is separated from the non-volatile fission-product fluorides in the fluorination step. Uranium and plutonium hexafluorides are separated from volatile fission-product fluorides by distillation.

In all the reference fluid-bed volatility plants of this study, the process feed is a combined batch of core and blanket fuel. The plant input consists of core and blanket fuel assemblies cooled a minimum of 15 days after removal from the reactor. Prior to chemical processing, the assemblies of core fuel and blanket rods are trimmed of excess structural metal without broaching the cladding. Batches of trimmed blanket and fuel are charged to the fluorinator vessel for the first chemical processing step.

In the first step, the cladding is chemically removed, and consequently, cladding material appears in process waste streams. As distinguished from the chop-leach methods employed in aqueous processing, chemical decladding avoids the necessity of chopping the fuel into short segments, and also avoids the production of a separate waste stream consisting of the hulls of the cladding segments, which must be monitored for fuel accountability. The above mechanical head-end steps are common to the volatility processing of all three fuel types.

1. Chemical Processing of Oxide Fuel

Chemical processing starts with charging to the reaction vessel of standard batches of combined core and blanket rods, which have been given the mechanical head-end treatment described above. The reference oxide fuel is assumed to be clad in Type 304 stainless steel.

a. Oxidation. As indicated in Fig. 9, the first step is an oxidation of cladding and the fuel charge. The oxidation of the fuel is conducted in a bed of fluidized alumina grain, a commercially available refractory, which is thermally stable and nearly inert to process reagents. The reagent used for the oxidation of the cladding is oxygen, together with up to 50% of hydrogen fluoride. The hydrogen fluoride acts as a catalyst, in that its presence is necessary to obtain a practical reaction rate. The cladding reacts completely at an operating temperature of 550°C, and the reaction products of the disintegrated cladding, chiefly oxides, mix with the alumina fluid bed.

In the oxidation step the fuel material is reacted to give products shown by the following principal reactions:

The oxide fuel disintegrates with the formation of the above solid products, which also mix with the fluid bed. Solids are retained in the fluid-bed vessel by sintered metal filters, through which the effluent gases and vapors pass in leaving the vessel. The behavior of fission products during the oxidation step and the methods employed for waste-gas treatment are described later in this section. The physical plant is described further in Section IV-D below, and equipment designs are discussed in Appendix A.

b. Fluorination. The fluid-bed fluorination step is conducted in the process vessel used for the above oxidation. The fluorination is conducted in two stages: a primary fluorination in which only UF_6 is formed, and a secondary fluorination in which PuF_6 is formed. Residual amounts of UF_6 accompany the PuF_6 fluorination product.

The primary fluorination employs bromine pentafluoride vapor as the fluorinating agent at about 300°C. In the primary fluorination the uranium reactions are

The iron, chromium, and nickel from the stainless steel are fluorinated in this step. A small portion of the chromium is expected to form a volatile fluoride, but otherwise the cladding constituents remain as solids in the fluorinator.

Before the secondary fluorination, plutonium dioxide is converted to nonvolatile PuF_4 . The secondary fluorination step employs elemental fluorine at about 500°C to convert PuF_4 to PuF_6 vapor. The maximum mole ratio of PuF_6 to fluorine in the vapor phase, as determined by the chemical reaction equilibrium, is about 1/100 at 500°C.

c. Hexafluoride Condensation and Distillation. The primary fluorination-gas effluent contains principally UF_6 , BrF_5 , and Br_2 , together with product oxygen and diluent nitrogen. For recycle purposes, the Br_2 product is immediately converted back to BrF_5 reagent by a gas-phase fluorination. This refluorination of Br_2 is carried out partially in the fluid-bed fluorinator by the addition of fluorine to that vessel. Subsequently the reaction is carried virtually to completion in a separate "regenerator" vessel located between the main fluorinator and the condenser. Operational control of the fluorination reaction with BrF_5 is achieved by regulating the rate of cooling of the vessel wall. It is assumed that the BrF_5 concentration in the nitrogen diluent is maintained at a constant value and the bed (reaction) temperature is controlled by cooling the vessel wall. The UF_6 - BrF_5 effluent from the regenerator is condensed as a liquid, or possibly as a slurry of UF_6 in BrF_5 . By setting the condenser temperature sufficiently low, the resulting partial pressure of the UF_6 - BrF_5 liquid condensate is sufficiently small that losses of these compounds are negligible in the volume of waste-gas bleed associated with the formation of free oxygen during the fluorination step.

The liquid condensate from the primary fluorination step is subsequently fed into a fractionation tower where the charge is separated batchwise into five fractions in the following order of decreasing volatility:

- (1) Highly volatile waste fraction;
- (2) Recycle BrF_5 fraction;
- (3) UF_6 product fraction;

- (4) BrF_3 fraction (recycled with BrF_5);
- (5) Low-volatility waste fraction.

Sharp separations are not required in this fractionation because of the following considerations: (a) Any UF_6 contained in fraction (2) is not lost but is recycled with the BrF_5 ; (b) any BrF_5 or BrF_3 in the UF_6 product in Step (3) is expected to be eliminated from the ultimate UO_2 or PuO_2 product by the hydrolysis reaction in the subsequent conversion step.

The effluent gas from the secondary fluorination contains PuF_6 at a concentration of the order of $1/2$ m/o in diluent gas, which is principally fluorine. The PuF_6 product is condensed as a solid in a large cold trap at about -70°C . The total quantity of PuF_6 collected in the trap is not large. Therefore, by this technique a PuF_6 product can be collected in a concentrated form suitable for feed to the subsequent step for conversion to dioxide. Further purification of PuF_6 may be required, though this point is not yet proved.

d. Conversion to Dioxide Product. A fluid-bed reactor is used for the conversion of uranium and plutonium hexafluorides to product dioxides by the overall reaction,

where M stands for uranium or plutonium. A semicontinuous scheme is used employing excess H_2 - H_2O reagent gas at about 650°C in alternate $1/2$ -hr periods of hexafluoride feed and oxide-product cleanup to remove fluoride. The process takes place by the deposition of solids on the surface of the fluid-bed particles to form high-density dioxide particles. In this operation, hexafluoride vapor is fed into a fluid bed of the product solids, and portions of the product are removed at the end of each complete cycle. Since the process takes place by the accretion of solid material on the fluid-bed particles, particle size is controlled by periodic in situ partial oxidation of the fluid bed, which results in fracturing of the particles.

Before UF_6 is converted to UO_2 for blanket fuel, the UF_6 product from the distillation step is stored for 60 days to allow the ^{237}U content to decay to a level that would allow direct-contact techniques in subsequent processing steps for blanket fuel. The ^{237}U content before storage will be high, because of the short cooling period before reprocessing. As in the case of the completely decontaminated uranium product from the aqueous process, the daughter of ^{232}U (itself a daughter of ^{241}Pu) is assumed to be sufficiently low to make contact handling feasible.

e. Fission-product Separation and Waste-gas Treatment. In the oxidation step, fission products xenon, krypton, iodine, and probably tritium are released from the fuel matrix into the gas phase where they accompany the O_2 , HF, and H_2O effluents. The water is removed to low levels by a partial condenser, which yields a concentrated hydrofluoric acid condensate. The other gases, including the bulk of the HF, are recycled and retained in the reactor system until the completion of the batch operation. The accumulated gases are then removed from the reactor system through a bed of granular soda lime (NaOH-CaO), which reacts with hydrogen fluoride and iodine and removes them from the waste gas. The soda lime is contained in a disposable reactor, which is replaced when spent. Oxygen is used to purge the reactor system. Oxygen is removed from the xenon-krypton gas stream by conversion to water by reaction with hydrogen on a palladium catalyst at ambient temperatures in a recycle system. The resulting moisture is removed on Molecular Sieves. A blower is used to recycle the gas until the reaction is complete. The Molecular Sieves are periodically regenerated with heated air. The concentrated rare gases xenon and krypton are sent to the waste-disposal facility.

The fission product tritium, while produced only in small amounts, is of some concern because it is present in a combined water waste stream, from which it is not separable by chemical means. The tritium is oxidized during decladding of the fuel by HF- O_2 mixtures and is collected with the product water in the effluent from the decladding step. The hydrofluoric acid condensate is revaporized and put through a soda lime bed, which fixes all halides as solids and allows noncondensable gases and some water to pass through for atmospheric dispersal. The residual tritium is assumed to be safely disposed of by dilution and dispersion to the atmosphere through a 200-ft stack.

In the primary fluorination, several volatile fission-product fluoride species are formed, such as SbF_5 , MoF_6 , RuF_5 , NbF_5 , TcF_6 , TeF_6 , and ruthenium and technetium oxyfluorides, and leave the reactor with the UF_6 product. The species that are condensed with the UF_6 are subsequently separated in the fractional-distillation step. An important amount of activity is associated with fission product TeF_6 , some of which leaves with the oxygen waste bleed from the condenser during the primary fluorination. The balance of the TeF_6 is separated in the fractional distillation as the highly volatile waste cut. These volatile fission-product fluorides are all fixed in disposable soda-lime beds. Fission-product molybdenum, while not radioactive, is present in high yield, and forms a volatile hexafluoride, which is treated the same as the TeF_6 .

Technetium is a high-yield, low-specific-activity fission product, which forms a hexafluoride with a volatility very close to that of UF_6 . Technetium may be removed from the product UF_6 by selective

sorption of technetium on a bed of magnesium fluoride at about 100°C. This technique may also remove neptunium, which is present as a hexafluoride with a volatility close to that of UF_6 .

In the secondary fluorination, the product PuF_6 is expected to be accompanied by minor quantities of residual volatile fission-product fluorides remaining from the primary fluorination. No fractional distillation is provided for the further decontamination of PuF_6 , but substantial decontamination from any SbF_5 , NbF_5 , or RuF_5 present can be achieved in the course of the vaporization and condensation operations involved in the transfer of PuF_6 from the original cold-trap collector to the feed boiler for the fluid-bed conversion to dioxide.

A large amount of fission-product radioactivity is associated with the rare earths, which form nonvolatile fluorides and remain as solids in the alumina fluid bed. The solid material is drained from the fluorinator after the secondary fluorination and conveyed to a waste packaging operation, which is further described in Section IV-E on waste disposal.

The waste-gas effluent from the fluid-bed conversion of mixed hexafluorides to oxides is expected to contain very little fission-product activity, but does consist of substantial amounts of hydrogen fluoride. This HF is condensed in water, and the condensate is piped to waste facilities for neutralization and further treatment.

2. Chemical Processing of Carbide Fuel

The fluid-bed fluoride-volatility processing of carbide fast-reactor fuel requires a flowsheet consisting of an assemblage of steps only slightly different from those described for the oxide and metal processes. The reference carbide fuel is assumed to be clad in stainless steel with a helium gas bond. The mechanical head-end steps are similar to those used for the oxide fuel.

a. Oxidation. The charge of carbide fuel rods is given an oxidation treatment similar to that used for the oxide fuel to effect the chemical removal of the stainless-steel cladding and the oxidation of the fuel matrix. In the carbide fuel, however, the oxidation is extended over a longer time because substantially all the carbon must be converted to carbon dioxide. To minimize formation of carbon monoxide due to oxygen-deficient conditions in the fluid-bed reactor, the reagent oxygen is added at several points of elevation and in excess quantity. To ensure the removal of carbon before the fluorination step, the final period of oxidation is conducted at a higher temperature (about 550°C). The composition of the fuel residue following the oxidation step is essentially the same as for the oxide fuel.

b. Fluorination. The primary fluorination, in which UF_6 is produced, is the same as for the oxide-fuel process. The secondary fluorination in which PuF_6 is produced is the same as for the metal-fuel process described in Section 3 below.

c. Fluid-bed Carbide Synthesis. The fluid-bed system for metal hydriding and carbide synthesis with hydrocarbon gases is essentially the same as that described for carbide synthesis in the aqueous process (Fig. 6). The preparation of metal feed for this step is identical with that described in Section 3 below for the reduction step for metal fuel.

d. Fission-product Separation and Waste-gas Treatment. In the oxidation step of carbide-fuel processing, the waste-gas effluent contains carbon dioxide in addition to the other process-gas constituents, O_2 , HF, H_2O , and the fission-product gases Xe, Kr, and I_2 . A condenser and caustic scrubber are used to remove HF, CO_2 , and I_2 . The residual gas is recycled; oxygen and hydrogen fluoride are added to the recycle gas as required. The fission-product gases that accumulate in the recycle gas during the oxidation step are purged from the system and disposed of in the same way as for oxide-fuel processing.

3. Chemical Processing of Metal Fuel

The processing of the reference metal fast-reactor fuel employs the same mechanical head-end treatment and shares the basic fluorination and chemical-separations features of the fluid-bed fluoride-volatility process as described in paragraph 1-b above for oxide fuel. However, the chemical steps in the present case differ in several respects from the volatility flow-sheet for oxide fuel. A simplified process-flow diagram for the metal fuel was shown in Fig. 11.

For both core and blanket metal fuel, cladding is assumed to be a vanadium-20% titanium alloy and sodium metal present in the rods as a thermal bond. In addition, titanium is assumed to be present in 9 a/o as an alloying constituent in the core fuel. Since both titanium and vanadium form fluorides that are volatile at $300^\circ C$, no separate chemical step is required for decladding, and, as shown in Fig. 11, chemical processing begins with fluorination. A series of chemical separation steps yields intermediate products UO_2 and PuF_4 as feeds for the reduction to metal products.

a. Fluorination. The primary UF_6 fluorination for metal fuel is similar to that for oxide fuel except that a longer time is allowed for processing a standard batch because of the larger total heat of reaction. In general, the restrictions as to allowable rate of heat transfer will determine the practical processing rate in the fluorination step of volatility processing. The operational control of the fluorination rate is achieved by regulating the temperature and the BrF_5 concentration of the reagent gas entering the

fluorinator. In the reagent gas, nitrogen is used as an inert diluent. It may also be desirable to include oxygen in the fluorinating gas. In this way, oxide fines would be produced and dispersed in the fluid bed, providing a solid of higher surface area to enhance the overall fluorination rates. The primary fluorination is continued with appropriate adjustments of temperature and reagent composition through the successive reactions of the vanadium-titanium cladding, the sodium bond, and the uranium-plutonium metal fuel.

In the secondary fluorination step, the residual PuF_4 is reacted with fluorine in a F_2 - BrF_5 mixture. The PuF_6 product is condensed as a liquid solution in BrF_5 . During the fluorination step, BrF_5 is recycled and fed to the fluorinator with the fluorine gas; this step in other ways is basically identical to that employed for oxide fuels. The BrF_5 is recycled following separation from the BrF_5 - PuF_6 condensate in a distillation tower.

Collection of PuF_6 in a liquid solution offers the following advantages to collection of the product by condensation of the solid:

- (a) Liquid condensation allows the use of a smaller condenser; (b) transfer of the product from one process vessel to another is facilitated; and,
- (c) a PuF_6 - BrF_5 mixture may be used directly as the feed material for the reduction of PuF_6 to PuF_4 by the use of BrF_3 .

b. Hexafluoride Condensation and Fractional Distillation.

The effluent gas from the primary fluorination contains the volatile fluorides of cladding elements (as VF_5 and TiF_4) as well as those of the fuel and of the fission products. Because of the substantial quantity of relatively high-boiling TiF_4 , a modified condenser is employed which incorporates a short packed section for fractionation of the incoming vapor. The high-boiling fluorides of Ti, Ru, and Nb are substantially separated from the effluent gas as the bottom product of the fractionating section. Uranium hexafluoride and BrF_5 are collected from the liquid reflux stream in the upper, low-temperature portion of the condenser. Uncondensed gas passing through the condenser is recycled to the fluorinator. The UF_6 - BrF_5 condenser product is subjected to a batch fractionation similar to that described in paragraph 1-c above for the oxide fuel. The disposition of the fission-product waste constituents are further described in paragraph f below.

The BrF_5 composition of the gas effluent from the secondary fluorination is controlled so that the average mole concentrations are about 200/20/1, respectively, for the fluorine, BrF_5 , and PuF_6 components. This composition allows the PuF_6 to be condensed with the BrF_5 as a liquid at low temperature in a manner similar to that employed for the UF_6 condensation in the primary fluorination. Fluorine is not condensed and is recycled to the fluorinator. The PuF_6 - BrF_5 condensate is fed continuously into a separate fractionating tower. The overhead product of the tower is substantially pure BrF_5 vapor, which is recycled to the fluorinator. The bottom product, accumulated during the batch run, has a nominal composition of 20% PuF_6 and 80% BrF_5 .

c. Reduction of PuF_6 to PuF_4 . The bottom product of the above fractional distillation is transferred as liquid to a vessel for the reduction of PuF_6 to PuF_4 . The reduction is carried out in the condensed (liquid) phase with bromine trifluoride as the reductant. The reaction proceeds as follows:

The bromine fluorides are distilled from the reduction vessel and recycled. The PuF_4 product is removed from the reduction vessel to serve as feed for the next step, which is reduction to plutonium metal.

d. Cooling of UF_6 and Conversion to UO_2 . The UF_6 product from the fractional distillation step is cooled for 60 days to reduce the ^{237}U content and is then converted to UO_2 to serve as feed in the final step of reduction to metal. A fluid-bed conversion step is carried out in a manner similar to that employed for the oxide fuel. However, substantial fluoride content is allowable in the present metal-processing case, since fluoride is only slightly less desirable than oxide as a feed material for the pyrochemical reduction step. It was assumed that small amounts of vanadium are allowable in the uranium oxide conversion product.

e. Reduction to Metal. Pyrochemical processes for reduction to metal are used for both uranium and plutonium. Separate facilities are used for the two products. The uranium facility operates on less stringent equipment requirements because of the absence of radioactivity or criticality restrictions for uranium. These pyrochemical reduction, retorting, and casting operations are essentially those described in paragraph III-B-2-a above in conjunction with the aqueous processing of metal fuel. Separate metal ingots of uranium and plutonium are the final products of the processing plant and serve as feed to subsequent fabrication operations.

f. Fission-product Separation and Waste-gas Treatment.

The general scheme of separation and fixation of volatile fluoride waste constituents for the processing of metal fuel is similar to that for oxide fuel. Because no oxide is present in the incoming fuel, no production of oxygen gas is entailed in the primary fluorination. Therefore recycle of noncondensable gas can be made virtually complete. The rare-gas fission products xenon and krypton accumulate in the noncondensable gas recycled to the fluorinator, and at the end of the fluorination are purged with oxygen from the system through a soda lime bed. The rare gases are concentrated by removal of oxygen in the manner described in paragraph III-D-1-e above for oxide fuel. Fission-product iodine and tritium are expected to be fluorinated to IF_7 and ^3HF , respectively, which accompany the UF_6 stream and are separated like TeF_6 in the highly volatile waste stream. The TiF_4 is separated with RuF_5 and NbF_5 in the low-volatility fraction. The vanadium (as VF_5) exhibits a volatility similar to BrF_5 (the boiling point of VF_5 is 48°C) and likewise is partly recycled and partly sent with the UF_6 product fraction as feed to the fluid-bed conversion step. A tendency for the BrF_5 content to build up in the

BrF_5 recycle stream is overcome by an occasional special fractional distillation of the BrF_5 - VF_5 mixture. Waste VF_5 is fixed in a soda-lime bed.

The effluent gas from the conversion step contains waste HF and bromine. These waste constituents are removed from the converter off-gas in a scrub tower by the same method as described in paragraph III-D-1-e above for oxide fuel.

IV. PROCESSING PLANT DESIGN

A. Design Philosophy for Commercial Operation

Two contrasting design philosophies for radiochemical processing plants are based on different modes of maintenance, viz., maintenance by remote operations and maintenance by direct personal contact. In this study, the definition of these maintenance concepts by Farrow¹ are accepted; namely, "Remote maintenance implies a plant in which the equipment is designed for installation and replacement by a remotely controlled crane without decontamination prior to replacement or the need for personnel access to the highly radioactive areas, whereas, contact maintenance requires decontamination of the equipment prior to its replacement so that personnel may enter the process areas and perform the necessary maintenance with essentially conventional techniques and tools."

The clearest presentation of the extremes of maintenance philosophy was presented in connection with the Hanford remote-maintained plant concept^{1,2} and the Idaho "small plant concept"^{3,4} using (nominally at least) contact maintenance. The Hanford evaluation of the two concepts was summarized by Harty² as follows: "An alternative to remote operation and maintenance of a separations facility is remote operation but contact maintenance following decontamination. It is conceivable that a contact-maintained separations facility can be built for two-thirds the cost of a remotely maintained separations facility, but it is estimated that such a contact-maintained facility would be 'on the line' less than one-third of the time that the remotely maintained facility would be. Hence, for equal production capacity, a contact-maintained separations facility would cost more than twice that of a remotely maintained facility. In addition, it may be possible to increase the capacity of a remotely maintained separations facility as much as threefold by locating 'bottleneck' equipment and designing suitable replacements; such a capacity increase is not believed to be obtainable in a contact-maintained plant."

For an aqueous plant serving plutonium-production reactors, in which a processing capacity of a number of tons per day is required and a plant availability factor in excess of 80% is desired, a plant design based on remote maintenance appears most economical. However, for a small-scale aqueous plant, processing one ton or less per day with no requirement for a high factor of plant availability, a plant design based on contact maintenance is indicated. In the present study, a processing-plant factor of 80% is one of the ground rules and small-scale is explicit in the throughputs which range from 0.08 tons/day for a plant serving one reactor to 0.8 tons/day for a plant serving ten reactors. Such plants fall in neither of the above classes.

A plant design based in part on a modified version of the Idaho small-plant concept^{3,4} was employed in the development of the processing-cost estimates presented herein for aqueous and volatility processing. The small-plant concept has as its principal objective the minimization of the cost of aqueous processing. This concept promises unit processing costs substantially lower than those for plants based on other design philosophies, and is an attempt to realize, by means of a novel design philosophy, the reliability of a remotely maintained plant and the economy in capital investment of a contact-maintained plant.

To realize comparatively low costs for building and equipment in the small-plant concept, the bulk of the chemical-processing equipment is virtually packed into a pit 30 ft deep. The equipment, in functional-unit groupings, is mounted on racks suspended from plugs in the top of the pit. Process, instrument, and service lines go through the top plug. Lines connecting functional units are laid through troughs in the top plugs. Quick-disconnects are strategically located to facilitate the isolation of any top plug and thus to permit the removal of the rack-mounted functional unit of equipment supported therefrom. The economy of cell space in the small-plant concept far exceeds that of either remote or contact plants because space allowances are not required for personnel or for remote-handling tools.

The small-plant maintenance philosophy, as described by Slansky,³ involves in situ decontamination followed by removal of the unit from the cell and contact maintenance. This feature of the small-plant concept has been the source of adverse criticism because complex equipment probably cannot be adequately decontaminated quickly to levels allowing contact maintenance. Operation of the processing plant at a plant factor as high as 80% under this mode of maintenance appears unlikely. To meet the plant-factor requirement of the present study, a mode of maintenance by functional-unit replacement was specified. If a component in a functional unit fails, the unit is drained and removed from the process cell, and a replacement unit is installed. Defective functional units from the very high-activity-level portion of the processing pit are taken to a dismantling cell and junked. Defective functional units, employed after the primary separation of fission products, are drained and removed to a hot maintenance facility where they are repaired.

A sectional view of the conceptual processing cell for the aqueous plant is shown in Fig. 12. The figure illustrates the maintenance mode (used both in aqueous and volatility plants) in which unit assemblies of equipment are replaced.

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Fig. 12. Sectional View of Conceptual Aqueous-processing Cell, Illustrating Equipment Replacement

It is believed that the Idaho small-plant design concept yields lower estimates of processing costs than the more conventional design philosophies of either contact-maintained or remotely maintained plants. Consequently, it was the concept used for the aqueous and volatility plants of this study.

The plant design for the pyrochemical processing plant was based on the fuel-cycle facility for EBR-II. The EBR-II Fuel Cycle Facility, which is close-coupled to the EBR-II reactor, uses pyrochemical procedures to process fuel. Processing operations are conducted remotely in a heavily shielded annular cell. Operations in the cell are viewed directly through strategically placed shielding windows.^{5,6}

References for Section IV-A

1. W. H. Farrow, Jr., *Radiochemical Separations Plant Study. Part II. Design and Cost Estimates*, DP-566 (March 1961).
2. W. M. Harty, *The Design Philosophy of Remote Operation and Maintenance of Separations Facilities*, AIChE Symposium Series, No. 13, Vol. 50 (1954).
3. C. M. Slansky and J. A. McBride, *Design Philosophy for Small-size Radiochemical Processing Plants*, 1962 Nuclear Congress of the Engineers Joint Council, Preprint No. 81, New York (June 1962).
4. H. Schneider, R. D. Fletcher, J. W. Coddington, R. G. Bearden, E. E. Erickson, S. J. Horn, M. E. Jacobson, R. F. Murray, and E. L. Rowe, *A Study of the Feasibility of a Small Scale Reprocessing Plant for the Dresden Nuclear Power Station*, IDO-14521 (April 28, 1961).
5. Reactor Fuel Processing 7(4):250 (Fall 1964).
6. J. C. Hesson, M. J. Feldman, and L. Burris, *Description and Proposed Operation of the Fuel Cycle Facility for the Second Experimental Breeder Reactor (EBR-II)*, ANL-6605 (April 1963).

B. Aqueous Processing

1. Oxide Fuel

The operations flow diagram for aqueous processing of oxide fuel appears in Fig. 13. Core and blanket fuel assemblies enter the remote-handling inert-atmosphere cell through a transfer lock. In the remote-handling cell, the assemblies are dismantled and the fuel chopped into short segments. Batches are prepared and loaded into baskets. The baskets are transferred directly through a dissolver chute into the dissolver. Other operations conducted in the remote-handling cell involve the hydrogen reduction of calcine produced in the fluid-bed denitration of uranium-plutonium aqueous product and the packaging of UO_2 - PuO_2 refabrication feed material.

Most of the rare gases are expected to be released to the remote-handling-cell atmosphere when the fuel elements are chopped. A bleed of the cell atmosphere is diverted to holdup tanks and released to the atmosphere when weather conditions permit.

The following equipment is housed in the processing pit:

- a. Dissolvers and most of the off-gas system.
- b. Solvent extraction columns, feed tanks, surge tanks, and solvent-purification mixer-settlers.
- c. Aqueous waste and intercycle evaporators.

Facilities are provided for scrapping or repairing defective equipment according to the maintenance mode adopted in this study. For defective equipment from the very high-activity-level portion of the plant, an equipment-disposal facility is used. For the maintenance of equipment in other parts of the process, a decontamination pit, a decontamination cell, and an equipment-disposal facility are provided. The decontamination pit accommodates the defective units from the processing pit which are mounted on racks. A spare assembly is installed to replace the defective unit. After decontamination, the defective unit is repaired in hot maintenance facilities and then becomes a part of the spares inventory. The decontamination cell serves the same function for the remote-handling cell. The equipment-disposal facility accommodates rack-mounted units from the processing pit and provides a remote-handling facility for the scrapping of equipment. It is also used to remove unsalvageable components from rack-mounted processing assemblies before decontamination.

Part of the used solvent is diverted to a waste-solvent incinerator to help maintain a low level of degradation products in the solvent. (The incinerator is housed in a separate building.) Shielded facilities are also provided for the 60-day storage of fully decontaminated aqueous uranium product to permit the decay of ^{237}U . Unshielded processing facilities are provided for acid recovery, conversion of aqueous uranium product to uranium dioxide, and preparation of reagents and materials introduced into the process. Also provided in the conceptual plant are offices, shops, warehouse space, stockrooms, analytical laboratory, and other necessary auxiliaries.

2. Carbide Fuel

The facilities provided for the processing of carbide fuel include, in addition to those provided for oxide fuel, the following:

- a. Inert-atmosphere remote-handling cell facilities which contain:
 - 1) Vibratory furnace for the conversion of carbide to oxide;
 - 2) Pyrochemical processing equipment for the conversion of $(\text{U}, \text{Pu})\text{O}_2$ intermediate product to metal;
 - 3) Hydriding and fluid-bed carburizing equipment for the conversion of metal to carbide.
- b. Unshielded pyrochemical facilities for the reduction of fully decontaminated uranium dioxide.
- c. Facilities for the fluid-bed conversion of uranium to uranium carbide and for the packaging of product.

3. Metal Fuel

Metal fuel is an intermediate product in the production of carbide, and the tail-end requirements for processing are implicit in the discussion of carbide processing. No head-end facilities are required.

C. Pyrochemical Processing

The cost estimates for the pyrochemical reprocessing of ceramic and metal fuels are based upon a plant design similar to that used in the EBR-II Fuel Cycle Facility in Idaho.¹ All the reprocessing and refabrication operations on the highly irradiated fuels are conducted remotely in a heavily shielded circular cell. An on-site fuel-cycle facility for this process is described below. The operations associated with the processing of both ceramic and metal fuels are also discussed.

1. Processing of Metal Fuels

A conceptual design of a facility for the reprocessing of metal fuels is shown in Figs. 14, 15, and 16. The discharged-fuel assemblies are transferred in a shielded coffin from the reactor to the Fuel Cycle Facility via a tunnel that terminates in a large air lock on the argon-atmosphere process cell. The fuel assemblies are stored in gas-cooled pits while awaiting processing. The processing and refabrication steps proceed in a counterclockwise direction around the process cell. The refabricated-fuel assemblies are retained in storage pits until they are required for refueling of the reactor.

The mechanical operations in the cell are performed with 5-ton bridge cranes, heavy-duty bridge manipulators, and master-slave manipulators. The manipulators have gastight seals to prevent gas leakage in or out of the cell. The master-slave manipulators are installed along the outer wall of the process cell to serve the work stations. The bridge cranes and manipulators are operated remotely by means of plug-ins provided both in the central control room and in the outer operating annulus. The transfer operations are viewed directly through shielding windows located in the inner and outer cell walls. The processing and refabrication operations are conducted remotely from the central control room. The electrical power supply units are located in the subcell and in the space above the control room.

The supporting facilities are located along the perimeter of the fuel-processing cell and in two wings extending outward from the cell. A small wing on one side of the cell contains a lobby, offices, a conference room, a lunch room, and a locker room. A large wing on the opposite side of the cell houses a warehouse, a stockroom, and an area for preparing

1. TUNNEL CONNECTING REACTOR AND FUEL CYCLE FACILITY
2. HOT MAINTENANCE SHOP
3. DECONTAMINATION CELL
4. ANALYTICAL LABORATORY
5. OUTER OPERATING ANNULUS
6. REFAB. INFLUENTS GLOVEBOX
7. ACCESS TUNNEL TO SUBCELL
8. WAREHOUSE
9. ADMIN. OFFICES, LUNCHROOM, AND LOCKER ROOM

10. PROCESSING ANNULUS
11. MAINTENANCE ANNULUS
12. INNER OPERATING ANNULUS
13. OFFICE SPACE
14. PROCESS CONTROL ROOM
15. SUBCELL
16. COLD RECEIVING DOCK
17. ELECTROMECHANICAL MANIPULATOR
18. BRIDGE CRANE

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Fig. 14. Sectional View of a Conceptual Fuel Cycle Facility for a Metal-fueled 1000-MWe Fast Breeder Reactor

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Fig. 15. Plan View of a Conceptual Fuel Cycle Facility for a Metal-fueled 1000-MWe Fast Breeder Reactor

108-9308

Fig. 16. Elevation View of a Conceptual Fuel Cycle Facility
for a Metal-fueled 1000-MWe Fast Breeder Reactor

refabrication hardware that will be transferred into the process cell through a large glovebox. An annular space along the perimeter of the process cell contains an analytical laboratory, including a shielded counting room, various utilities, a health-physics office, a shop for the repair and maintenance of low-activity-level equipment, and a cell in which equipment can be decontaminated. An instrument shop, a machine shop, and an area for preparing items such as crucibles and process reagents for introduction into the process cell, are located beneath the central control room. The process materials are transferred into the cell through the air lock in the waste-processing area of the cell.

The flow diagram of the operations involved in the pyrochemical processing of metal fuels is presented in Fig. 17. The metal fuels require fewer, simpler, and more economical processing operations than the oxide fuels (see Fig. 18). For all types of fuel (ceramic or metal), the core and blanket materials are processed separately. The main differences in the processing of oxide and metal fuels are as follows:

- a. The metal fuel is declad mechanically in an operation that is only slightly more expensive than the procedure used to perforate the cladding material for ceramic fuels.
- b. The oxidative decladding furnace used to separate the ceramic fuel from the perforated cladding is not required.
- c. Since the metallic core and blanket materials enter the process directly as metals, the oxide-reduction step is not required.
- d. A simple halide slagging procedure is used for the removal of electropositive fission products from metal fuels, whereas a multistage extraction technique is employed with ceramic fuels.
- e. In the case of metal fuels the metallic products of the process are suitable for direct fuel refabrication, while oxide and carbide fuels require a product conversion step before refabrication.

2. Processing of Oxide and Carbide Fuels

A flow diagram of the operations involved in the pyrochemical processing of oxide fuels is shown in Fig. 18. The operations required for the processing of carbide and oxide fuels are similar, except that the metallic products are converted to carbides instead of oxides. All operations indicated in the flow diagram are conducted in a process cell similar to that shown in Fig. 14 for metal fuels. These operations are described below.

Fig. 17. Operations Flow Diagram for Pyrochemical Processing of Metal Fuel

108-9338

The core and blanket materials are processed separately. The core assemblies are dismantled, and the fuel elements are cropped to remove excess structural metal. The decladding operation entails perforation of the cladding to allow access of oxygen to the fuel, thereby facilitating oxidation of the fuel and splitting of the cladding. The fuel is pulverized by the oxidation procedure and is recovered as a granular powder. The oxidation step is carried out at 700°C in a furnace containing a vibratory basket for separating the oxidized fuel from the cladding. Carbide fuel is declad and converted to oxide by a similar procedure. However, the carbon dioxide produced in the reaction is removed from the off-gas stream from the decladding furnace by means of a scrub tower containing a sodium hydroxide solution.

When the fuel elements are cropped and perforated, a substantial portion of fission-product krypton and xenon is released to the cell atmosphere. The remaining portion is released during the oxidative decladding of the fuel. To limit the equilibrium concentrations of these fission-product gases in the cell atmosphere, a bleed stream of the atmosphere is removed continuously and treated according to the procedure described in Section IV-E on waste disposal.

After the decladding step, the oxidized fuel is treated with hydrogen to reduce the uranium from U_3O_8 to uranium dioxide. The hydrogen treatment lowers the magnesium requirement in the subsequent reduction step and decreases the quantity of magnesium oxide formed and, therefore, the amount of salt waste. The hydrogen reduction also has the function of providing further comminution of the fuel for the reduction step.

After the hydrogen-reduction step, the uranium and plutonium dioxides are separated from fission products and reduced to the metals in a sequence of batch operations. These batch operations are conducted in resistance-heated furnaces, which are covered with bell jars to contain their own argon atmosphere. They are also fitted with the necessary agitators, transfer lines, instrumentation, etc. The furnace heaters are integral with the bell jars, and solder seals are used for the transfer lines and the base-plate closure of the bell jars.

The reduction step yields a solution of uranium and plutonium in liquid cadmium-zinc-magnesium alloy. This solution is extracted by a molten chloride salt in a multistage, countercurrent column to remove rare-earth fission products. The rare earths in the molten-salt effluent are removed by a single-stage extraction with liquid aluminum-10% magnesium alloy, and the purified salt is recycled to the rare-earth extraction step.

Uranium-plutonium alloy and metallic uranium are recovered from the liquid-metal solvents (cadmium-zinc-magnesium alloy) by evaporation of the solvent metals in intermittently fed retorts. The retorting equipment incorporates pumps, surge tanks, beryllia crucibles, silicon carbide-lined, sodium-cooled condensers, fractionation columns, and graphite-lined, solvent-metal receivers. The recovered solvent metals are recycled.

The uranium and uranium-plutonium alloy from the retorting operations are converted to uranium dioxide and a uranium dioxide-plutonium dioxide mixture, respectively, by a two-step oxidation-reduction procedure in a small, resistance-heated furnace. In the first step, the uranium is converted to U_3O_8 and the plutonium is converted to PuO_2 by oxidation of the metals with oxygen. The second step consists of reduction of the U_3O_8 with hydrogen to form uranium dioxide. In the case of carbide fuel, the metals are first hydrided in a small vessel, then carburized by treatment with a mixture of hydrogen and methane or propane in a fluid-bed reactor. The hydriding furnace and the fluid-bed reactor are an integral unit. During hydriding, the metals are converted to powdered hydrides, which are pneumatically conveyed into the carburizer by the hydrogen stream. The accumulated hydride is then carburized, and the carbide product is conveyed pneumatically into the product receiver.

References for Section IV-C

1. J. C. Hesson, M. J. Feldman, and L. Burris, *Description and Proposed Operation of the Fuel Cycle Facility for the Second Experimental Breeder Reactor (EBR-II)*, ANL-6605 (April 1963).

D. Fluid-bed Fluoride-volatility Processes

Equipment and building requirements were based on the reference flowsheets described in Section III-D for fluid-bed fluoride-volatility processing of core and blanket of the three types of fast-reactor fuel. Combined core and blanket fuel are fed to the plant in batches limited in size to twice the daily load specified in Table I (p. 11). This batch-size limitation was selected for nuclear safety and is based on the plutonium content of the batch. The same batch-mass limits were arbitrarily imposed on all the plutonium-containing functional units of the process indicated schematically as boxes in the operations-flow diagrams of Figs. 19, 20, and 21. On the other hand, the separated uranium blanket fuel is not subject to criticality limits, and the size of the process equipment used for blanket fuel is therefore determined by the plant throughput requirements.

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Fig. 19. Operations Flow Diagram for Fluid-bed Volatility Processing of Oxide Fuel.

108-9310

Fig. 20. Operations Flow Diagram for Fluid-bed Volatility Processing of Carbide Fuel

108-9311

Fig. 21. Operations Flow Diagram for Fluid-bed Volatility Processing of Metal Fuel

The conceptual volatility plant contains the following six major operating areas:

1. A remote operations cell, which contains (a) equipment for dismantling and trimming the irradiated fuel assemblies and for preparing the batch charges to the fluorinator, (b) the disposable fluoride waste-gas treatment reactors, and (c) facilities for packaging the alumina waste from the fluid-bed fluorinator. Also located in this cell are the plutonium fuel processing equipment: (a) the fluid-bed converter for preparation of uranium and plutonium oxide from the hexafluorides; (b) in the case of metal and carbide fuels, the components for reduction of PuF_6 to PuF_4 and metal; (c) in the case of carbide fuel, the equipment for synthesis of carbides; and (d) facilities for solids sampling of the fluid-bed reactors. The cell is equipped with shielding windows, a crane, manipulators, an inert-gas atmosphere system, and other facilities for completely remote manipulations. Transfer locks are provided for introducing and removing equipment and materials. This cell has provisions for the remote maintenance of contained equipment.

2. A decontamination cell, adjacent to the remote operations cell, for decontaminating containers or other items removed from the remote-operations cell.

3. A process hot cell, for the volatility-separations equipment. The major items in this cell are the fluid-bed fluorinator, the bromine fluoride regenerator, the hexafluoride condensation system, and the fractional-distillation system. Also located in the process hot cell is equipment for the treatment of waste gas from the oxidation step and the rare-gas concentration system. In addition, intermediate product receivers and the hexafluoride converter feed boilers are located here. The process equipment is preassembled in functional units on support racks. Each equipment rack is designed to be removed and replaced as a unit. The racks are suspended from removable shielding plugs at the top of the cell. Connections to process and instrument lines are located above the shielding plugs. Coupling operations are performed semiremotely with local shielding.

4. The equipment-disposal cell, a remote-operations cell specifically designed for the removal of equipment from the process hot cell and for the disposal of this equipment by cutting it up and packaging the sections into waste containers. The cell provides access to the process hot cell, but no process operations are carried out in this cell.

5. The process cold area, which contains all nonradioactive chemical facilities. It includes reagent feed storage and metering tanks, equipment for the treatment of the converter off-gas, and a large scrubber for the ventilation air of the process hot cell and of the process cold area.

Uranium blanket conversion steps, including metal reduction and carbide synthesis are conducted here. Equipment for monitoring the ventilation exhaust air is also included. Although remote-operating techniques are not required, some of the equipment requires ventilated enclosures or hoods.

6. A shielded area, for the UF_6 storage tanks for interim cooling of the uranium (containing ^{237}U) before the conversion steps to blanket oxide, metal, or carbide.

The conceptual plant also includes offices, utility rooms, a warehouse, an analytical laboratory, and active and inactive maintenance shops.

The fluorination equipment is constructed of nickel and high-nickel alloys. Design concepts, equipment specifications, and other plant details are discussed in Part 2 of Appendix A.

E. Waste Disposal

1. General Design of Waste-disposal Facilities

The basic criteria for the disposal of radioactive fission-product wastes arise from the radiological safety factors governing the indefinitely long periods of storage. On the basis of existing knowledge, complete criteria cannot be established now for ultimate storage methods expected to be in use in 1980. The reference concept adopted in this study was that substantially all the fission-product activity be contained in high-activity-level solid waste, sealed in cylindrical metal containers. These waste packages are allowed to cool in a storage canal under water until the internal heat generated by radioactive decay is about 300 Btu/(hr)(cu ft), or 3 W/liter, when they are permanently stored under conditions providing a high degree of safety against possible dispersion and with minimum requirements for surveillance. It was assumed that the location of permanent storage would be separate from the processing plant and, therefore, shipping would be required after interim cooling.

Because of the high rate of heat generation of the short-cooled waste, inert-gas cooling is required for the waste containers in the remote-operations cell of the processing plant, where the waste is packaged. The waste from the remote-operations cell is transported to the interim storage canal by means of shielded casks equipped with water-cooling.

The interim storage facility for the packaged waste consists of a central canal for receiving and removing the cylinders, additional canals for the storage of the cylinders, and a service area containing the water-cooling and -purification equipment. The central and storage canals are constructed of reinforced concrete with walls and floor 1 ft thick. The inside surface of the concrete is covered with a radiation-resistant, protective

coating as an aid in maintaining high-purity water and for easier decontamination. The depth of the water above the stored containers provides

Fig. 22. Plan View of a Conceptual Canal for Interim Cooling of Containers of High-activity Waste from a Pyrochemical Processing Plant Serving Ten 1000-MWe Reactors

by an overhead 5-ton bridge crane. The storage canals are housed in a light-construction steel building.

The canal water is recycled to the service area for cooling and demineralization. The cooling system consists of pumps, intermediate heat exchangers, and a cooling tower. In the primary loop, water is circulated by pumping from the ends of the canals, through the heat exchangers, and back to the central facility. In the secondary loop, cooling water is pumped through the heat exchangers and returned to the cooling tower. Cooling systems are designed to handle the maximum heat-generation rate. A rise of about 15°C in the water during passage through the canals is allowed.

The water-purification equipment consists of a filter and deionizers through which the water in the canals is circulated at a rate equivalent to a turnover time of 1 week. The water is deionized by passing first through a bed of strongly acid resin, and then through a bed of weakly basic resin. Tanks are provided for the storage of acid and sodium carbonate regeneration solutions.

biological shielding. The central canal serves as an area for underwater loading of the cylinders into casks prior to the shipping off site. A plan view of the canal is shown in Fig. 22.

The cylinders of solidified waste are stored upright in the canal. They are arranged in parallel rows with a space between rows for moving them in and out. Each cylinder is allowed a space of twice its diameter in both directions. The area of canal required for storage of the cylinders in this array is a function only of interim storage time and independent of the diameter of the cylinder.

To keep the waste cylinders in position during storage, the cylinders are placed in individual stands constructed of a structural aluminum alloy and anchored in the floor. The cylinders are placed in and removed from these stands

The design concepts and cost sources employed in the above high-activity, solid-waste-disposal scheme were adapted from a waste-disposal evaluation reported by Oak Ridge National Laboratory.¹⁻³ Waste-disposal costs may be reduced further through the development of satisfactory means of interim storage in larger containers (bins) and by provision for on-site permanent storage. However, such improvements are likely to benefit most processes and therefore are of economic indifference in the present comparative study.

The above scheme for ultimate disposal of high-activity solid waste is immediately applicable to volatility and pyrochemical processes because the high-activity-level wastes are separated directly as solids. In the case of aqueous processing, an additional step must be provided for solidification of the radioactive material in the aqueous-waste solutions. Rather than choose among the competing methods currently under development for aqueous-waste solidification, it was decided in this study to base the aqueous-waste-disposal cost on perpetual on-site storage of concentrated liquid waste⁴ resulting from the evaporation of plant effluents. This method avoids charges for solidification, for shipping, and for separate final burial, and has the status of a highly developed, economical approach, even though it does not embody the safety features associated with active waste in solid form. By taking this avenue for estimating aqueous-waste-disposal costs, the principle of economic equivalence is assumed. By the time of the establishment of a fast-reactor economy, say 1980, the technology of aqueous-waste disposal will have advanced to the point where the costs of waste solidification, interim storage, shipping, and perpetual storage in a remote area are as low as current costs for on-site perpetual storage of concentrated waste solutions.

Certain fission products that are not contained in the solid or liquid waste streams of any of the processes are handled separately. The fission-product rare gases xenon and krypton are collected in concentrated gas form in the processing plant and are transferred to the waste facility where they are stored in tanks under pressure for 100 to 150 days to allow the radioactive xenon to decay. The remaining krypton activity is diluted with air and is discharged under administrative control from the main 200-ft-high stack of the waste facility when meteorological conditions are favorable. All vent streams are passed through a charcoal bed and an absolute filter for removal of residual activity before discharge to the stack.

Most of the activity of fission-product iodine decays with a short half-life (8 days), and substantial reduction of this activity takes place during the period of initial cooling of the discharged fuel. Although this decay is advantageous for the longer-cooled fuel used in aqueous processing, no great advantage is achieved for the short-cooled fuel used in the volatility and pyrochemical processing. The iodine remaining in the fuel is assumed

to be released as vapor in the initial step of chemical processing. Sorbent beds are provided to retain the iodine during a period of decay. Spent sorbent is buried as low-activity solid waste. Thousands of curies of iodine resulting from the processing of 15-day-cooled EBR-II fuel are presently handled in this manner.

Fission-product tritium, although present in small amounts, is of concern because it has a moderately long half-life (12 yr) and can react to form T_2O , which, when mixed with natural water, is practically inseparable. In the reference plants, tritium is assumed to form water, and this water is evaporated and discharged under suitable meteorological conditions to the environment through the main 200-ft stack of the plant. The schemes for atmospheric dispersal of krypton and tritium were based on the studies by Farrow⁵ and ORNL⁶.

2. Aqueous-processing Waste Disposal

For the aqueous-processing plant, the waste facility includes the main exhaust stack of the plant, evaporators to concentrate aqueous waste, waste-solution neutralization facilities, and a system of tanks for perpetual liquid-waste storage. Provision is made for trapping of iodine and for interim storage of fission-product rare gases. Various inactive and low-activity-level wastes are stored on site.

The high-level fission-product activity is contained in the aqueous-waste effluents from the aqueous-processing plant. These aqueous wastes are concentrated in a system of three waste evaporators, and provision is made for recovery of acid values. Radioactive aqueous wastes from other sources, such as the analytical laboratory and decontamination operations, are added to the major waste stream. The degree of volume reduction of the aqueous-waste solution achieved during evaporation is limited by fission-product heating and by the solubility of the dissolved waste solids. The principal dissolved solids, other than fission products, are iron from the dissolution and solvent extraction steps, and sodium from the carbonate wash of solvent.

The concentrated waste solution is neutralized and stored in 200,000-gal, carbon-steel tanks. Some design features of the small-plant concept⁷ were used in the design of this conceptual waste facility.

The kerosene solvent is washed with sodium carbonate solution to remove the products of solvent degradation resulting from the high levels of irradiation. This spent washing solution is added to the high-level aqueous waste of the plant. Solvent is replaced on a basis of 10% discard of the stream from the co-decontamination cycle. This solvent is burned in an incinerator; the combustion gas is filtered and washed. The ash and wash are added to the main active waste-storage tanks. Disposal of solvent by incineration is reasonably well developed.^{8,9}

The perpetual storage of liquid wastes requires high-integrity vessels and provision for everlasting surveillance of the wastes and future replacement of the vessels. In the plan adopted, one waste tank is being filled at all times, and an empty tank is always on hand as a spare to receive waste transferred from any of the storage tanks that may become defective. When the waste tanks are filled, the tank formerly serving as a spare is then put into service and a new spare tank provided. (In the aqueous-waste facility for one 1000-MWe reactor, a 200,000-gal waste tank is filled in 6 yr.) Each tank put into service is assumed to have a lifetime of 50 yr, when a new tank is provided to receive its contents and the old tank is emptied. Surveillance and operating charges for the storage period (after filling) must be provided for each tank put into service. Because the entire plant is given an economic lifetime of 30 yr, charges for all tank replacements and operating expenses of perpetual storage are funded over the 30-yr economic lifetime.

Present worth concepts, with no allowance for inflation or deflation, were employed in the computation of the annual charge over the 30-yr lifetime of the facility for the on-site perpetual storage of concentrated, neutralized, high-activity-level, aqueous wastes. Two 200,000-gal tanks per reactor were provided in the initial investment in plant and equipment. Future costs were converted to a uniform annual charge against current operations over the 30-yr lifetime of the plant by summing the following components:

- a. A uniform annual charge to provide 200,000-gal tanks, as needed, 6, 12, 18, and 24 yr hence.
- b. A uniform annual charge to provide for the perpetual replacement of each in-service tank at 50-yr intervals.
- c. A uniform annual charge to establish a fund at the end of 30 yr, which would thereafter provide \$1,500/yr per reactor for perpetual surveillance.
- d. A uniform annual charge, amounting to 20% of the annual charge, for perpetual replacement to provide for perpetual maintenance.

In addition to the provisions made for disposal of liquid wastes, provisions are also made for the disposal of waste gases.

A substantial portion of the rare gases xenon and krypton are expected to be released in the mechanical chopping of fuel. The rare gases released to the inert atmosphere of the processing cell are continuously removed by passing a bleed stream of gas from the cell through a charcoal bed at low temperature. The gaseous fission products remaining in the

feed after chopping are released in the initial chemical steps. For carbide fuel, the fission-product gases are expected to be evolved in the dry oxidation of the chopped core and blanket rods. For oxide and metal fuels, the gases are evolved in the acid dissolution of the chopped fuel.

The fission-product waste-gas effluent from the dry oxidation of carbide fuel is passed through a charcoal bed and an absolute filter to remove iodine, and a caustic scrubber to remove carbon dioxide. The rare gases are then dried and absorbed on charcoal at low temperature to separate them from other noncondensable gases, which are vented. The fission-product waste-gas effluent from the acid dissolution of oxide and metal fuel is first passed through a caustic scrubber to remove nitric oxide present in the dissolver off-gas. The waste gas is then dried and passed through a charcoal bed and filtered to remove iodine. After more complete removal of water, the xenon and krypton are separated from other gases by sorption on charcoal at low temperature.

The rare gases xenon and krypton, which have been sorbed on charcoal beds, are periodically desorbed and stored under pressure in steel tanks for a maximum of 150 days. The gas is then dispersed to the atmosphere under suitable conditions by venting through the main 200-ft stack of the plant.

3. Fluid-bed Volatility-process Waste Disposal

For the fluid-bed volatility-processing plant, the waste facility consists of (a) the main exhaust stack of the plant, (b) a storage canal for interim cooling underwater of packaged high-activity-level solids wastes, (c) a system for interim storage of fission-product gases, and (d) facilities for indefinite on-site storage of low-level and chemical wastes of various forms. Aqueous radioactive wastes from laboratory and decontamination sources are evaporated and stored on site in stainless-steel tanks.

The high-activity-level process-waste streams and the associated waste facilities constitute the following:

a. The waste alumina bed from the fluid-bed fluorinator, containing nonvolatile fission-product residues. This very high-activity material is packaged and sealed in disposable containers in the processing plant. The containers are stored under water in the interim storage canal. The containers are of welded steel, 13 in. in diameter and 4.5 ft long. They are aluminized for external corrosion resistance and are stored two-high in the canal.

b. The spent soda lime in the disposable waste-gas reactor, containing fluorides from the off-gas waste from the fluorination and distillation steps. This very high-level activity is principally niobium and

ruthenium. These containers are stored for 1 yr under water in the interim-waste storage canal. The containers are 10 in. in diameter and otherwise similar to those used for the alumina waste.

The rare-gas fission products, xenon and krypton, from the fluid-bed fluorinator purge gas are collected and concentrated in the processing portion of the plant. The waste facility provides for the low-temperature condensation of this gas and revaporization under pressure to fill interim storage tanks where the gas is held for 150 days. The cooled gas is dispersed to the atmosphere under favorable atmospheric conditions by means of the exhaust-air stack.

Some fission products, such as iodine and technetium, are collected on solids in disposable waste containers. These containers, which are expected to be at low-activity level, are buried on site.

Aqueous hydrofluoric acid condensate waste from converter off-gas treatment is a process-waste stream with very low activity. A facility is provided for neutralization of this waste with lime. Tanks are provided on site to collect the calcium fluoride sludge from the neutralized lime slurry. The bromide-containing overflow solution is concentrated by evaporation and stored in tanks. The water from the evaporator overhead is released to the environment.

For carbide fuel, there is a waste stream of spent caustic solution from the off-gas scrubbing of the effluent of the oxidation step. This stream contains fission-product iodine and fluoride and carbonate chemical waste. This stream is combined with the above hydrofluoric acid waste stream.

Soda lime (NaOH-CaO) is effective for general removal of halogens and volatile halogen compounds. However, to insure the high removals required for fission-product iodine and tellurium hexafluoride, charcoal traps are employed as a backup in the waste-gas-treatment system. The spent traps are buried in all cases.

4. Pyrochemical-process Waste Disposal

The high-activity-level fission-product wastes resulting from pyrochemical processing appear in two major waste streams: (a) a metal waste stream, which is a combination of two metal effluent streams, and (b) a salt waste stream, which is a combination of two salt effluent streams. These metal and salt streams are handled separately in the waste-disposal operations. The metal waste consists of aluminum, magnesium, cadmium, and zinc and contains the rare-earth, refractory-metal, and noble-metal fission products. The salt waste is composed mainly of the chlorides of

magnesium, sodium, and potassium, along with some oxide and fluoride. The alkali-metal and alkaline-earth-metal fission products appear in the salt waste.

The containers used for final disposal of the metal and salt wastes in the procedures for their handling are similar to those employed for the high-activity-level fluoride-volatility-process wastes. The containers, made of steel with a protective aluminum coating, are 15 in. OD by 4 ft long for the metal-waste containers, and 24 in. OD by 4 ft long for the salt-waste containers. The containers filled with the metal waste are cooled under water in the interim storage canal for 8 yr, and those filled with the salt waste are similarly cooled for 1 yr. The melting points of the salt and metal wastes and the heat-transfer rates under these conditions are such that a solid phase is always maintained at the inside wall of the container, even under the initially high heat fluxes resulting from fission-product decay. After the interim cooling periods, the fission-product decay heat has diminished sufficiently to permit burial of the waste containers.

In the pyrochemical processes, the metal fuels are declad mechanically, whereas the clad oxide and carbide fuels are chopped into small pieces. Experience in the EBR-II Fuel Cycle Facility has shown that only a slight release of the fission-product gases, krypton and xenon, occurs during the mechanical decladding and chopping of metal fuels. However, larger amounts of these gases will probably be released during the chopping of ceramic fuels. In the conceptual fuel-processing facility, the fission-product gases are allowed to build up to an equilibrium level in the argon atmosphere of the processing cell. This equilibrium level is maintained by continuous passage of a bleed stream of the argon atmosphere through a low-temperature charcoal bed, which removes the krypton and xenon. Final disposal of these fission-product gases is by the same method as that described below for the fission-product gases released during the halide slagging of metal fuels.

In the halide-slugging procedure, fission-product krypton, xenon, iodine, and tellurium are volatilized. The major portions of the iodine and tellurium are collected on a silicate fiber-fume trap, which rests on top of the halide-slugging crucible. The remaining portions are removed by passing the argon atmosphere from the process furnace through a charcoal trap. The effluent gas from the charcoal trap consists of argon, containing fission-product krypton and xenon. This gas mixture is stored under pressure in steel tanks for 120 to 150 days to allow most of the xenon to decay. When the meteorological conditions are favorable, the suitably cooled gas is discharged from the tanks to the atmosphere through the 200-ft stack of the plant.

The spent fume traps described above are packaged in disposal containers similar to those used for the salt wastes. After interim cooling of about 4 months, the waste containers are buried on site. The spent charcoal traps and the empty cladding hulls are disposed of similarly, but interim cooling is not required.

References for Section IV-E

1. R. L. Bradshaw, W. J. Boegly, Jr., F. M. Empson, H. Kubota, F. L. Parker, J. J. Perona, and E. G. Struxness, *Ultimate Storage of High-level Waste Solids and Liquids in Salt Formations*, in Proceedings of the Symposium Treatment and Storage of High-level Radioactive Wastes, held by the International Atomic Energy Agency in Vienna, October 8-12, 1962, pp 153-173, Vienna (1963).
2. J. O. Blomeke, J. J. Perona, H. O. Weeren, and R. L. Bradshaw, *Evaluation of Ultimate Disposal Methods for Liquid and Solid Radioactive Wastes. Part III. Interim Storage of Solidified Wastes*, ORNL-3355 (Oct. 21, 1963).
3. J. J. Perona, R. L. Bradshaw, J. O. Blomeke, and S. T. Roberts, *Evaluation of Ultimate Disposal Methods for Liquid and Solid Radioactive Wastes. Shipment of Calcined Solids*, ORNL-3356 (Oct. 18, 1962).
4. S. M. Stoller Associates, *The Perpetual Care of High-level Radioactive Wastes in New York State*, NP-12486 (Oct. 12, 1962).
5. W. H. Farrow, Jr., *Radiochemical Separations Plant Study. Part II. Design and Cost Estimates*, DP-566 (March 1961).
6. Oak Ridge National Laboratory, *Chemical Technology Division Annual Progress Report for Period Ending May 31, 1964*, ORNL-3627, pp 113-114 (Oct 1964).
7. H. Schneider, R. D. Fletcher, J. W. Coddington, R. G. Bearden, E. E. Erickson, S. J. Horn, M. E. Jacobson, R. F. Murray, and E. L. Rowe, *A Study of the Feasibility of a Small Scale Reprocessing Plant for the Dresden Nuclear Power Station*, IDO-14521 (April 28, 1961).
8. R. R. Stiens, *Disposal of OMP High Boilers by Combustion*, NAA-SR-5410 (Nov. 15, 1961).
9. J. A. Harper and L. Lolly, Jr., *A Method of Disposal of Large Quantities of Degraded and Low-level Contaminated Solvent*, DPSPU 64-30-8B (May 1964).

V. SUMMARY OF UNIT COSTS AND IDENTIFICATION OF SIGNIFICANT DIFFERENCES IN COST

A. Data Sources and Plant Models

The plant-design concept and cost sources for aqueous processing were mainly derived from the Idaho small-plant concept presented by Schneider *et al.*¹ This process concept offers the lowest costs of aqueous plants for comparison with advanced nonaqueous processes. The concept was modified to accommodate maintenance of the hot portion of the plant while retaining the requirement of an 80% plant factor.

The plant design concept and cost sources for the pyrochemical process were mainly derived from the EBR-II Fuel Cycle Facility operating at Idaho.²

The volatility-equipment design concepts and cost sources were derived from ANL pilot-plant experience. Plant concepts were selected to conform to those for the aqueous or the pyrochemical plants as far as was appropriate.

Models relating costs to complexity and size were employed to estimate costs for the processing plant and its auxiliaries for the several cases in a consistent manner. The chief models and factors of estimation were based on methods developed by Stockdale,³ Zevnik and Buchanan,⁴ and Bauman.⁵

The basic design concepts and cost sources for interim storage of high-activity-level solid waste was taken from Blomeke *et al.*⁶ The shipping charge for the cooled waste was based on the study by Perona *et al.*,⁷ and the charges for final burial on the study by Bradshaw *et al.*⁸

The provisional cost estimate of aqueous-waste disposal by perpetual liquid care was patterned after the study by Stoller.⁹

For the central processing plants, out-of-reactor inventory and shipping charges for spent fuel were based on a study by Nelson.¹⁰ Because longer cooling of spent fuel increases inventory costs but lowers shipping costs, optimum cooling times were found for which the sum of inventory and shipping charges were minimum.

Auxiliary facilities for which cost estimates were made in this study included "hot" and "cold" maintenance shops, warehouse, offices, personnel convenience areas, and analytical laboratory.

B. Unit Costs

For the reference cases of this study, unit costs in mills/kWe-hr were calculated to serve as a unit of measure of the relative cost of the alternative fuels and processes. These unit costs are summarized for the three reference plant sizes in Tables V, VI, and VII. The relation between the electrical unit costs shown and the other measures of possible interest, namely, total annual cost and unit costs in \$/kg of fuel processed, can be seen from the reference items of annual production shown in the tables.

TABLE V. Unit Cost Summary for Close-coupled Plant Serving One Reactor (1000-MWe)

Plant Factor, 80%

Annual Production of Electrical Energy, 7.01×10^9 kWe-hr

Fuel	Cost Component	Unit Cost, mills/kWe-hr		
		Aqueous	Volatility	Pyrochemical
Oxide	Processing	0.53	0.35	0.44
	Waste Disposal	0.15	0.09	0.09
	Subtotal	0.68	0.44	0.53
	Inventory	0.17	0.12	0.12
	Shipping	-	-	-
	Total	0.85	0.56	0.65
	1% Loss	0.016	0.016	0.016
Carbide	Processing	0.62	0.43	0.46
	Waste Disposal	0.15	0.09	0.09
	Subtotal	0.77	0.52	0.55
	Inventory	0.17	0.12	0.12
	Shipping	-	-	-
	Total	0.94	0.64	0.67
	1% Loss	0.016	0.016	0.016
Metal	Processing	0.59	0.39	0.29
	Waste Disposal	0.15	0.09	0.06
	Subtotal	0.74	0.48	0.35
	Inventory	0.17	0.12	0.12
	Shipping	-	-	-
	Total	0.91	0.60	0.47
	1% Loss	0.016	0.016	0.016

TABLE VI. Unit Cost Summary for Close-coupled Plant Serving
Three Reactors (3000 MWe)

Plant Factor, 80%

Annual Production of Electrical Energy, 21.0×10^9 kWe-hr

Fuel	Cost Component	Unit Cost, mills/kWe-hr		
		Aqueous	Volatility	Pyrochemical
Oxide	Processing	0.29	0.21	0.25
	Waste Disposal	0.12	0.08	0.08
	Subtotal	0.41	0.29	0.33
	Inventory	0.11	0.06	0.06
	Shipping	-	-	-
	Total	0.52	0.35	0.39
	1% Loss	0.016	0.016	0.016
Carbide	Processing	0.33	0.26	0.26
	Waste Disposal	0.12	0.08	0.08
	Subtotal	0.45	0.34	0.34
	Inventory	0.11	0.06	0.06
	Shipping	-	-	-
	Total	0.56	0.40	0.40
	1% Loss	0.016	0.016	0.016
Metal	Processing	0.32	0.24	0.17
	Waste Disposal	0.12	0.08	0.05
	Subtotal	0.44	0.32	0.22
	Inventory	0.11	0.06	0.06
	Shipping	-	-	-
	Total	0.55	0.38	0.28
	1% Loss	0.016	0.016	0.016

TABLE VII. Unit Cost Summary for Central Plant Serving
Ten Reactors (10,000 MWe)

Plant Factor, 80%

Annual Production of Electrical Energy, 70.1×10^9 kWe-hr

Fuel	Cost Component	Unit Cost, mills/kWe-hr		
		Aqueous	Volatility	Pyrochemical
Oxide	Processing	0.16	0.14	0.18
	Waste Disposal	0.10	0.08	0.07
	Subtotal	0.26	0.22	0.25
	Inventory	0.11	0.08	0.08
	Shipping	0.05	0.07	0.07
	Total	0.42	0.37	0.40
	1% Loss	0.016	0.016	0.016
Carbide	Processing	0.19	0.17	0.19
	Waste Disposal	0.10	0.08	0.07
	Subtotal	0.29	0.25	0.26
	Inventory	0.11	0.08	0.08
	Shipping	0.05	0.07	0.07
	Total	0.45	0.40	0.41
	1% Loss	0.016	0.016	0.016
Metal	Processing	0.18	0.15	0.12
	Waste Disposal	0.10	0.08	0.05
	Subtotal	0.28	0.23	0.17
	Inventory	0.11	0.08	0.08
	Shipping	0.05	0.07	0.07
	Total	0.44	0.38	0.32
	1% Loss	0.016	0.016	0.016

The unit costs in Tables V, VI, and VII are presented, respectively, for the 1000-MWe on-site, the 3000-MWe on-site, and the 10,000-MWe central-processing plants. For the single size in each table, the plants are classified as to type of fuel and type of process. The cost components of each case are shown for comparison.

The processing-cost component refers to the processing plant proper, including head-end and tail-end processes and all stocks of spare equipment and auxiliary facilities. The auxiliary facilities include maintenance shops, analytical laboratory, warehouse, offices and convenience areas for personnel, mobile equipment, and the electrical substation. The plants are described in Section IV.

The waste facilities are shown separately because their design and operation are not determined by the processing requirements in the same way as are the processing facilities. The waste-disposal facilities are also described in Section IV.

The unit costs for the sum of processing cost and waste-disposal cost are shown in the tables as a subtotal. This sum is the prime measure of relative cost among the several fuel and flowsheet cases and incorporates the basic estimates made in this study for equipment, buildings, and operating labor and materials.

Inventory costs were calculated for the out-of-reactor fuel inventory. (Details are given in Appendix H.) Shipping of the spent fuel from the reactor to an off-site processing plant is applied only to the central plant, and this expense is not incurred in the close-coupled facilities. The shipping costs of central plants were based on a previous study,¹⁰ and details are given for a range of fast-reactor processing cases described in Appendix E. Because of the dependence of both shipping and inventory cost on the time allowed for the spent fuel to cool after it is discharged from the reactor, the costs of inventory and of shipping are not independent. The longer the cooling time, the lower the shipping cost and the higher the inventory cost. Accordingly, the approximate minimum cost was determined for the sum of shipping and inventory costs for the central plants. The reference distance from reactor to central-processing plant was 1000 miles.

The total is shown in Tables V, VI, and VII to indicate for comparison the unit costs of the components listed above.

The 1% fuel-loss figure is based on the annual processing rate and the value of fuel. The processing loss was arbitrarily considered a matter of indifference for the comparisons of this study and is shown only to indicate its order of magnitude relative to the cost components that are determined by the alternative fuels and processes.

C. Waste-disposal Cost Estimate

The estimate of the cost of waste disposal includes all charges for further treatment of waste streams emerging from the separations of the processing portion of the plant. The waste operations include perpetual storage of all fission-product activity with the exception of tritium and

krypton, which are dispersed to the atmosphere. This will probably not be an acceptable ultimate solution for the gases, but the costs associated with storage of the gases will not change the relative process costs since such costs are expected to be greatest for aqueous and least for pyrochemical processes.

1. High-activity-level Wastes

For pyrochemical and volatility processing, the bulk of the fission products are contained in a concentrated, high-activity-level solid-waste stream separated in the processing portion of the plant and packaged in disposable containers. The container costs were estimated from shop costs of similar units. The costs of storage canals for the interim cooling of the packaged wastes were derived from an analysis of similar facilities for equal container capacity and heat duty reported by Blomeke⁶ et al. Charges for shipping of cooled solid waste were made by the model presented in a study by Perona⁷ et al. Estimates of the cost of permanent underground storage of cooled solid waste were based on an analysis of Bradshaw et al.⁸

The cost of disposal of high-activity-level wastes from aqueous processing was based on the perpetual on-site storage of liquid-waste effluents from solvent extraction after concentration in evaporators. The assumption was made that, by 1980, methods for the conversion of aqueous wastes to solids would be fully developed; thus, at that time, aqueous-waste disposal would involve conversion to solids and would incorporate a procedure for interim storage and final burial of solids similar to that specified for volatility and pyrochemical processing at a total cost assumed to be no greater than that for perpetual, on-site, liquid storage. Evaporator costs were based on those presented in the Idaho small-plant concept.¹ The costs of tanks in a perpetual care scheme were based on the analysis of Stoller.⁹

2. Gaseous Wastes

The costs of iodine, xenon, and krypton waste operations were based on an analysis of the Idaho small-plant scheme for the aqueous process.¹ Applicable portions of this scheme were used for similar operations for the pyrochemical and volatility processes. The cost of equipment for concentration of waste gas containing xenon and krypton in the volatility process was based on unpublished costs of EBR-II units for removal of oxygen from an inert gas atmosphere.

3. Other Waste Facilities

The costs of other waste facilities were based on cost analyses derived from the Idaho Chemical Processing Plant as reported by Hull and Zeitlin¹¹ and by Robertson and Stockdale.¹² These facilities include the

ventilation exhaust air filtration and blower system, the 200-ft stack, and the lagoon for retention of inactive liquid wastes. Costs of the incinerator for disposal of degraded solvent were derived from a cost analysis by Stiens.¹³

References for Sections V-A through V-C

1. H. Schneider, R. D. Fletcher, J. W. Coddling, R. G. Bearden, E. E. Erickson, S. J. Horn, M. E. Jacobson, R. F. Murray, and E. L. Rowe, *A Study of the Feasibility of a Small Scale Reprocessing Plant for the Dresden Nuclear Power Station*, IDO-14521 (April 28, 1961).
2. J. C. Hesson, M. J. Feldman, and L. Burris, *Description and Proposed Operation of the Fuel Cycle Facility for the Second Experimental Breeder Reactor (EBR-II)*, ANL-6605 (April 1963).
3. W. B. Stockdale, "Estimating Costs for Radiochemical Plants," in *Cost Engineering in the Process Industries*, edited by C. H. Chilton, McGraw-Hill Book Company, Inc., New York, p. 304 (1960).
4. F. C. Zevnik and R. L. Buchanan, *Generalized Correlation of Process Investment*, Chem. Eng. Progress **59**, No. 2, 70 (1963).
5. H. C. Bauman, *Fundamentals of Cost Engineering in the Chemical Industry*, Reinhold Publishing Corp., New York (1964).
6. J. O. Blomeke, J. J. Perona, H. O. Weeren, and R. L. Bradshaw, *Evaluation of Ultimate Disposal Methods for Liquid and Solid Radioactive Wastes. Part III. Interim Storage of Solidified Wastes*, ORNL-3355 (Oct. 21, 1963).
7. J. J. Perona, R. L. Bradshaw, J. O. Blomeke, and J. T. Roberts, *Evaluation of Ultimate Waste Disposal Methods for Liquid and Solid Radioactive Wastes. Shipment of Calcined Solids*, ORNL-3356 (Oct. 18, 1962).
8. R. L. Bradshaw, W. J. Boegly, Jr., F. M. Empson, H. Kubota, F. L. Parker, J. J. Perona, and E. G. Struxness, "Ultimate Storage of High-Level Waste Solids and Liquids in Salt Formations," pp. 153-175, in *Proceedings of the Symposium on Treatment and Storage of High-Level Radioactive Wastes* held by the IAEA in Vienna, October 1962. International Atomic Energy Agency, Vienna (1963).
9. S. M. Stoller Associates, *The Perpetual Care of High-level Radioactive Wastes in New York State*, NP-12486 (Oct. 12, 1962).
10. R. C. Vogel, M. Levenson, and F. R. Masten, *Chemical Engineering Division Semiannual Report, July-December 1964*, ANL-6925, pp. 261-263 (May 1965).
11. H. L. Hull and H. R. Zeitlin, *A Design and Cost Estimate Study of a Purex Plant*, ORNL-1558 (March 31, 1954).
12. P. L. Robertson and W. G. Stockdale, *A Cost Analysis of the Idaho Chemical Processing Plant*, ORNL-1792 (Jan. 4, 1955).
13. R. R. Stiens, *Disposal of OMP High Boilers by Combustion*, NAA-SR-5410 (Nov. 15, 1961).

D. Sources of Differences in Unit Costs for the Alternative Processes

The estimates developed in this study primarily indicate a significant difference in the costs associated with aqueous and nonaqueous processing. In most cases the quality of the estimates is not adequate for drawing conclusions concerning the relative economic merits of pyrochemical and volatility processing. Important technical questions remain to be answered for all the processes, and in this study it is assumed that the processing requirements can be satisfied by the conceptual processes described here. In addition, the unit charges for plutonium loss and plutonium credit, which were treated as matters of indifference, certainly will not be identical for the different processes or for the different fuels. Since the estimation of fabrication costs were beyond the scope of this study, we did not determine the penalty to be levied against pyrochemical processing to account for the additional cost incurred because the blanket must be refabricated remotely. This topic is discussed in Appendix G. On the basis of a preliminary evaluation of refabrication requirements, the choice of remote versus semiremote refabrication, although not a matter of indifference, imposes only a nominal penalty on pyrochemical processing, which does not affect the conclusions developed in this study. The frame of reference for the discussion of the source of the differences is primarily the on-site Fuel Cycle Facility serving a single reactor. The effect of throughput on the differences is discussed in Section V-C below.

1. Oxide Fuel

The unit-cost estimates for the processing of oxide fuel were presented graphically in Fig. 2. The volatility process provides the lowest cost for oxide fuel. For close-coupled processing of the output of a single reactor, in comparison with aqueous processing, the unit costs are lower by 35% for processing, 44% for waste disposal, and 30% for inventory.

For the major separations portions of the processing plants, similar conceptual plant designs were adopted for aqueous and volatility processing. The lower unit cost for processing in the volatility process is attributable primarily to the much smaller number of units of equipment required for volatility processing. The difference between aqueous and volatility processing in the mode of maintenance accounts for a minor portion of the difference in cost between the two processes. In aqueous processing, a 5-yr expected lifetime was assumed for all processing equipment employed after the gross separation of fission products; however, for volatility, equipment employed in high-activity-level service, a lifetime of only 2 yr was conservatively assumed. Partially offsetting the lower cost in aqueous processing that derives from the longer lifetime of the equipment is the expense associated with the provision of facilities for decontamination. There is no requirement for a comparable facility in the volatility plant, since it was costed on a replacement rather than repair

basis. For aqueous processing there appears to be a cost advantage for the decontamination mode of maintenance compared to that for simple replacement. This advantage exists for the plant serving a single reactor and becomes increasingly greater as the throughput is increased, because the plant throughput has little effect on the cost of the decontamination facility. The diminishing effect of the cost of the decontamination facility is typical of many costs that account in part for the convergent trend in aqueous and volatility processing of the unit costs for processing as the throughput is increased.

The unit cost for processing for the pyrochemical flowsheet is intermediate between aqueous and volatility processing for the close-coupled plants. The pyrochemical flowsheet employed in this study for oxide fuel is a modified version of the EBR-II skull-reclamation process. The modifications were necessary to provide for the recovery of plutonium, which currently is not a requirement in the processing of the enriched uranium EBR-II fuel. The difference in the unit cost for processing between the pyrochemical flowsheets for oxide and metal fuel is attributable to the cost associated with multistage extraction to separate rare earths and the oxidation of the metallic retort product to provide an oxide refabrication feed material. The newer pyrochemical processes being developed primarily for ceramic fuels are expected to reduce this difference.

The lower unit cost for waste disposal in volatility processing versus aqueous processing is attributable to the smaller volume of high-activity-level wastes produced and the fact that the wastes are solids in a form convenient for packaging and storage. The waste-disposal costs for pyrochemical and volatility processing of oxide fuel are practically identical. In volatility processing the unit costs for waste disposal are the same for all fuels. However, in pyrochemical processing the unit cost for waste disposal is higher for ceramic fuel than it is for metal fuel, primarily because of the added cost of the disposal of the magnesium oxide that is produced in the reduction of the ceramic fuels to metal.

2. Carbide Fuel

The unit-cost estimates for the processing of carbide fuel are presented graphically in Fig. 3. The nonaqueous processes provide the lowest costs for carbide fuel. The quality of the estimates does not afford a distinction between pyrochemical and volatility processing in the estimated unit costs for either processing or waste disposal in close-coupled processing. In the central plant serving 10 reactors, the volatility process yields the lowest unit cost for processing. In comparison to aqueous processing, for a close-coupled volatility plant serving a single reactor, the unit costs are lower by 31% for processing, 44% for waste disposal, and 31% for inventory.

In each alternative process, the flowsheet for oxide fuel modified by the addition of appropriate head-end and tail-end steps is employed in the processing of carbide fuel. The principal source of the differential in the unit costs for processing between oxide and carbide fuels is the expense associated with the conversion of the primary product to carbide. This differential is greater for volatility and aqueous processing than it is for pyrochemical processing. In volatility processing, the steps are the conversion of oxide to metal and the conversion of metal to carbide. In pyrochemical processing, the only step required is the conversion of metal to carbide. The unit costs for processing are about the same for the volatility and pyrochemical processes because the lower cost of the conversion of the primary product to carbide in pyrochemical processing offsets the small advantage in the cost of processing enjoyed by volatility processing for oxide fuel.

The waste-disposal costs are virtually the same for oxide and carbide processing. In the processing of carbide fuel, a small added cost is associated with the provision of a caustic scrub tower to remove carbon dioxide from the process off-gases, but this cost is not detectable in the computed unit cost for processing.

3. Metal Fuel

The unit cost estimates for the processing of metal fuel are presented graphically in Fig. 4. The pyrochemical process provides the lowest cost for metal fuel. For close-coupled processing of the output of a single reactor, in comparison to aqueous processing, the pyrochemical costs are lower by 50% for processing, 60% for waste disposal, and 30% for inventory. As the plant capacity increases, the relative advantage of pyrochemical processing decreases. However, even for the central plant serving 10 reactors, the pyrochemical-processing unit costs are significantly lower than aqueous-processing costs in all categories except shipping. Higher shipping costs are to be expected because the overall economics demand that fuel to be processed pyrochemically be shipped after very little cooling, and the shipping costs are primarily a function of the heat-dissipation requirements.

The lower cost for pyrochemical processing derives from the compactness and simplicity of the equipment and from the relatively simple process. A breakdown of the expense of aqueous and pyrochemical processing shows that (a) the head-end operations employed in both aqueous and pyrochemical processing to prepare the fuel for processing are, on the basis of cost, virtually identical; (b) the cost of aqueous-processing equipment for dissolution, solvent extraction, denitration, uranium product storage, and acid recovery alone exceeds all costs of equipment for pyrochemical processing; and (c) the cost of equipment employed in the aqueous-processing operations to convert the oxides to uranium-plutonium alloy for use in core

refabrication, and facilities for the conversion of uranyl nitrate solution to uranium for blanket refabrication, is about equal to the difference in equipment costs for the two processes.

In aqueous processing, the requirement of an 80% plant factor necessitates the maintenance of completely assembled functional units for the high-activity-level portion of the aqueous plant and the availability of the facilities needed to assure the expeditious replacement of defective functional units of processing equipment. These facilities are the decontamination pit and the equipment-disposal facility. In pyrochemical processing, the fuel is processed batchwise and the processing units are essentially isolated from one another. Therefore, the failure of one unit does not have an immediate effect on the other units. The demonstration of the remote replacement of defective components of pyrochemical processing units in the operation of the EBR-II Fuel Cycle Facility provides the basis for the expectation, which is implicit in the facilities provided in the conceptual pyrochemical processing plant, that an 80% plant factor can be obtained without an inventory of preassembled spare units, but rather, only spare parts are required. Since pyrochemical processing is batchwise and not continuous, the processing units are not assembled into integrated functional groupings of equipment as in aqueous processing. Consequently, there are no requirements for the special-purpose decontamination pit and equipment-disposal facility specified for the aqueous-process plant. The absence of these facilities is another important factor in the difference in the unit cost of processing between the aqueous and nonaqueous processes.

The lower waste-disposal costs for pyrochemical processing obtain from the much lower quantity of wastes produced and their convenient form. The difference in the waste-disposal costs of aqueous processing and pyrochemical processing are largely accounted for by the expense incurred in aqueous processing for the neutralization and reduction in volume of the aqueous waste produced and the incineration of organic wastes.

E. Sources of Differences in Unit Costs for Three Levels of Plant Throughput

The unit-cost differences between pairs of alternative processes were summarized in Table III. The differences are between summations of unit costs for processing, waste disposal, and inventory charge. The differences are greatest for an on-site plant serving a single reactor. As the throughput is increased, the unit differences tend to diminish, but since the number of units is increasing, the absolute dollar difference does not diminish, so that the economic incentive for improvement remains large. Additional information pertinent to the effect of increased throughput is provided by the presentation of ratios of costs for the alternative processes

in Table IV. Comparisons of the ratios of summations of unit costs for nonaqueous and aqueous processing show that the cost of nonaqueous processing relative to that of aqueous processing increases in going from a close-coupled plant serving three reactors to a central plant serving ten reactors. Note that the central plant summations include the estimated unit costs for shipping.

The sums of the estimated unit costs for the cost categories of processing, waste disposal, out-of-reactor fuel inventory, and shipping of spent fuel are shown graphically in Fig. 2 for the reference facilities servicing one, three, and ten 1000-MWe fast reactors. The one- and three-reactor cases are close-coupled plants for which shipping of spent fuel is not required. The charges for shipping estimated in this study accrue only to the 10,000-MWe central plant.

1. Close-coupled Processing; Comparison of Unit Costs for Plants Serving One and Three Reactors

Unit costs were estimated for close-coupled plants serving one and three reactors. The data in Tables V and VI show that the summation of the unit costs for processing, waste disposal, and inventory are lower for the plant serving three reactors than they are for the plant serving one reactor by about 40% for aqueous processing, 37% for volatility processing, and 42% for pyrochemical processing. Thus, the effect on the summation of unit costs of increasing the plant throughput to serve three reactors is approximately the same for all processes. However, the sources of the reductions differ for the three processes.

a. Unit Cost for Processing. The components of the summation of unit costs are the unit costs for processing, waste disposal, and inventory. The reduction in the unit cost for processing is approximately 40% for all processes. In aqueous processing, which is a continuous process, the primary sources of the reduction in the unit cost for processing are the lower cost per unit of throughput of larger-capacity equipment and the small effect of increased capacity on direct labor costs in a highly automated continuous-processing facility.

In the pyrochemical process, the principal source of the reduction in the unit cost for processing is increased utilization of the processing equipment. Nominally, the plant factor for the entire pyrochemical plant is 80%. However, because of the batchwise character of pyrochemical processing, the most economical mode of operation results in much lower utilization factors for most of the individual equipment items. For example, the utilization factor of the halide slagging furnace in the plant serving one metal-fueled reactor is about 20%. The only components that are operated at a utilization factor of 80% are the multistage extraction facility required in the processing of ceramic fuel and the retorts that are

specified in all the pyrochemical flowsheets. Thus, only a relatively small increase in the cost of processing equipment was required to effect a three-fold increase in throughput. However, a penalty associated with batch processing partially mitigates the beneficial effect of the increased utilization factor; i.e., the increase in the cost of direct labor. As a consequence of these compensating factors, the reduction in the unit cost for processing is about the same for the aqueous and the pyrochemical processes when the plant throughput is increased threefold.

In the volatility process, the plutonium-containing items of equipment are operated semicontinuously with a standard mass amount of plutonium as a nuclear safety restriction. The items of equipment that do not contain plutonium are in general scaled up in size in the same manner as continuous equipment. The volatility process thus is subject to the cost influences described above for batch and continuous equipment for the pyrochemical and aqueous process, respectively. The compensating cost factors result in nearly the same reduction in unit costs for all three processes as the plant throughputs are increased corresponding to the reference 1000- and 3000-MWe close-coupled processing plants.

b. Unit Cost for Waste Disposal. The threefold increase in throughput affords reductions in the unit cost of waste disposal of about 20% for aqueous processing and about 10% for nonaqueous processing. The greater reduction in aqueous processing is attributable to the much lower cost per unit of throughput for the aqueous-processing facilities required for the concentration of aqueous waste and the incineration of organic wastes. An added factor is the cost of direct labor associated with these facilities because of the small effect of throughput on direct labor in highly automated operations.

The nonaqueous processes produce low-volume, concentrated wastes, which are encapsulated and placed in an interim storage facility to cool sufficiently to allow ultimate disposal off site. The size and number of the waste containers are determined by the heat-generation rate of the wastes. Therefore, the number of containers to be stored and the prime cost for encapsulation are directly proportional to the throughput. Other waste-disposal facilities in nonaqueous processing (for example, the stack and the heat exchangers in the storage canal) are relatively insensitive to throughput, so that the net effect is only a 10% reduction in the unit cost of waste disposal.

c. Inventory Charge. Based on the ground rules of this study, for a plant serving three reactors with uniformly staggered refueling schedules, compared to a plant serving a single reactor, the reduction in the unit charge for inventory is 35% for aqueous processing and 50% for nonaqueous processing. This difference derives directly from the

ground-rule minimum requirements of 120-day cooling of the fuel before aqueous processing and 15-day cooling of the fuel before nonaqueous processing.

The ratios of the unit charges for inventory for nonaqueous to aqueous processing in the one- and three-reactor cases are about 0.70 and 0.54, respectively. These data show that substantial savings in inventory charges can be realized by nonaqueous processing and that the savings are greater for the plant serving three reactors than they are for the plant serving one reactor.

2. Central-plant Processing

In this section the unit costs for a central plant serving ten reactors are compared with those for a close-coupled plant serving three reactors. The central-plant costs include a charge for shipping, which is not incurred in close-coupled processing. To make this comparison possible it is assumed that in a future fast-breeder-reactor economy the regulations affecting the shipment of large quantities of highly irradiated fuel will not impose a prohibitive economic burden on central-plant processing. The shipping costs in this study are in accord with the optimistic point of view of fuel shipment. Future regulations and increased shipping costs could affect the on-site versus central-plant costs, but should be of economic indifference as between different processes.

a. Unit Cost for Processing. The reductions in the unit cost for processing, as the throughput is increased from the output of three to that of ten reactors, is 44% for aqueous processing, 36% for volatility processing, and 28% for pyrochemical processing. From these data, and from the presentations in Figs. 2, 3, and 4, it is apparent that for the nonaqueous processes the effect of increased throughput on decreased cost is not as great in going from three to ten reactors as it was in going from one to three reactors. This difference in scaling factors arises from the ground rules used and the basis of estimating costs. The pyrochemical flowsheet selected and the plant-design philosophy used dictate batch-size limits such that even if all the plutonium in solution precipitated from the solvent metal, no criticality incident would occur. As a result, the "ten-reactor plant" contains multiple lines of equipment throughout. The same ultraconservative philosophy was applied to the volatility plant for all plutonium-handling equipment. Because of this, the reduction in cost with increased throughput is not as great as it might otherwise be. For the aqueous plant, only a single solvent-extraction line was funded, and therefore, the cost decreases more rapidly with increased throughput. However, the resulting aqueous plant does not have the same degree of reliability as do the multiple-line volatility or pyrochemical plants. If multiple lines were to be included for the aqueous solvent-extraction plant, the costs of processing in such a plant would increase significantly.

b. Unit Cost for Waste Disposal. The discussion presented for close-coupled processing is applicable to the throughput increase from the three- to the ten-reactor case. No additional considerations are involved.

c. Unit Charge for Inventory and Shipping. Because of their interdependence in the central-plant case, the unit costs for shipping and inventory are properly considered together. In the central-plant case, the ratio of the joint costs for nonaqueous processing to those for aqueous processing is 0.92. The comparable ratios for the one- and three-reactor cases are 0.69 and 0.52, respectively. These data show that the cost advantage enjoyed by nonaqueous processing in close-coupled plants (because they can accommodate short-cooled fuel) is largely lost in the central plant, because the shipping costs are greater for short-cooled fuel. The estimates of shipping costs were based on optimistic assumptions, which made no allowances for the possibility that liability-insurance rates may be substantially higher for fuel cooled only 15 days than for fuel cooled 120 days.

The nonaqueous processes can accommodate very short-cooled fuel. The reason for the minimum cooling requirement of 15 days for nonaqueous processing is to allow for the decay of fission products to a level at which the heat-generation rate can be tolerated in the remote-handling and -processing operations. The reagents used in aqueous processing, however, are subject to radiation damage, and some of the degradation products adversely affect the decontamination and recovery of fuel values. A minimum cooling requirement of 120 days was specified for aqueous processing to allow for the decay of fission products to a level at which the rate of energy release can be compensated for by a practical procedure for the maintenance of satisfactory solvent quality. The effect of short cooling on costs shown in this study is in the reduction of the inventory charge. Of the cases evaluated in this study, the most pronounced effect of short cooling was observed in the analysis of a close-coupled plant serving three reactors. In central-plant processing, the combined effects of shipping costs as used in this study yield a smaller advantage for short cooling because of the increased cost of shipping short-cooled fuels.

F. A Preliminary Estimate of the Effect of Burnup on Processing Costs

Core burnup affects both processing costs and out-of-reactor inventory, but does not significantly affect other cost categories considered. Waste-disposal costs and shipping costs are not primarily dependent on the rate of discharge of fuel, but only on the rate of discharge of fission products (i.e., the power of the reactor).

The influence of core-fuel burnup on processing costs is governed by the plant throughput. A special analysis was not made of the effect on processing costs for increasing core-processing rates at constant levels of

blanket throughput. However, in the light of throughput effects discussed above, an exponential scaling factor of about $1/2$ would probably be an appropriate estimate of the maximum effect of burnup on processing costs. That is, a factor-of-two reduction in burnup would result in an increase in processing throughput such that the new total annual processing cost would be no more than $2^{1/2}$ or 1.41 times the processing cost of the reference-burnup case. On this basis, the decrease of core burnup from 10 to 5% would result in an increase of 41% in the processing cost as the cost difference for the two burnups at a given electrical power level.

The effect of core-processing rate on out-of-reactor inventory does not allow an economy of scale, and the inventory costs for a burnup of 5% is twice that for 10%. The inventory costs are smaller than the processing costs in the cases studied, ranging from 23 to 63% of the processing component of fuel-cycle costs in this study.

Allowable burnups between different types of fuels, as established in 1980, probably would not be different by as much as a factor of two, and the preliminary conclusions are: (1) Expected differences in core burnups of fast reactor fuels have an effect on reprocessing cost considerably less than that indicated by a direct proportionality of throughputs, and (2) the effect of reduced burnup does not change the comparative cost of the alternative processes as estimated for 10% burnup, but rather increases the magnitude of the estimated cost differences.

APPENDIX A

Critique and References for Chemical Flowsheets and Engineering Designs Used in This Study

1. Critique of Aqueous Processing

For this study it was assumed that spent-fuel reprocessing procedures would be required to accommodate plutonium fuels irradiated to 100,000 MWD/metric ton. The aqueous processes currently employed for use in the reprocessing of lower-burnup uranium fuels may not be satisfactory for the processing of fast-reactor fuel. The areas of principal concern regarding process feasibility are fuel dissolution, solubility of fission products in process streams, and radiation damage to process streams.

a. Dissolution

The conceptual process selected accomplishes the dissolution of uranium and plutonium from oxide and metal fuels by a chop-leach procedure.^{1,2} The clad fuel pins are chopped into sections about 1 in. long to expose the fuel, which is then dissolved in 8M nitric acid containing about 0.05M HF. In the presence of a small amount of hydrofluoric acid, the nitric acid dissolution of high-fired solid-solution uranium-20 w/o plutonium dioxide proceeds to completion at a reasonable rate.³ Although the presence of a substantial quantity of noble metals in highly irradiated fast-reactor metal fuel will almost certainly prevent its complete dissolution, this study has assumed that 99.9% of the uranium and plutonium can be recovered by leaching. This assumption has yet to be demonstrated on high-burnup plutonium fuel. To prepare it for aqueous dissolution, the carbide fuel is declad and converted to oxide by controlled oxidation in a vibratory furnace, using a technique developed for the decladding of oxide fuel.⁴

(1) Oxide Fuel. Although in certain cases, sintered, solid-solution uranium-plutonium dioxide can be dissolved in nitric acid,³ an insoluble residue of plutonium dioxide frequently remains. The dissolution behavior of uranium-plutonium dioxide may be greatly affected by the behavior of the fuel during irradiation. The effect of high burnup at high temperature on the physical properties of solid-solution (U-Pu)O₂ is not definitely known, but segregation to some degree during irradiation appears likely. If plutonium segregation results in a material not soluble in nitric acid, a serious feasibility problem arises. Experimental programs to study the behavior of oxide fuels in fast reactors are just getting under way. It will be several years before the dissolution characteristics of spent (U-Pu)O₂ fast-reactor fuel can be realistically assessed.

The solubility characteristics of the fission products at the concentrations specified in the conceptual flowsheet are not well known. It is assumed here that complete solubility will occur; however, if this is not true, then some form of feed clarification will be required and a cost increase will result.

(2) Carbide Fuel. Uranium and plutonium can probably be completely leached from mixed carbide fuel. However, the resultant aqueous solution is not a satisfactory solvent-extraction feed, because of the presence of soluble organic compounds and insoluble graphitic residues, which cause emulsification problems in solvent-extraction columns.⁵ An efficient method for removing carbon is to burn the mixed carbide fuel in oxygen. This procedure was adopted in the conceptual flowsheet and was used as the basis for the cost estimates developed in this study. No information is available on the feasibility of dissolving uranium-plutonium oxides produced by oxidation of the carbides.

(3) Metal Fuel. The fast-reactor metal fuels currently considered most promising are alloys of uranium and plutonium containing about 1.5 w/o of a refractory metal. Zirconium and titanium are the candidate refractory metals. After irradiation, the composition of the fuel is uranium-plutonium-10 w/o fission products-1.5 w/o refractory metal.

The nitric acid dissolution of alloys that contain as little as 1% zirconium is hazardous because of the formation of compounds that deteriorate spontaneously.⁶ The formation of explosive compounds can be avoided by the addition of four to eight moles of hydrofluoric acid per mole of zirconium. However, the presence of hydrofluoric acid eliminates stainless steel as a material of construction, unless a very dilute solution is employed. A stainless-steel dissolver can be used to contain a boiling solution composed of 6.5M nitric acid and not more than 0.04M hydrofluoric acid.⁷

b. Feed Preparation

Once the fuel is dissolved, the subsequent solvent-extraction problems are similar for all fuels. A major problem concerns the high concentration of fission products from fuel irradiated to 10% burnup. Specifically, because of the relatively low solubility of the noble-metal fission products, the conventional Purex flowsheets cannot be employed in the processing of high-burnup fast-reactor fuel.

The conceptual aqueous process for fast-reactor fuel is based to a large extent on processing studies of uranium-plutonium-molybdenum fuels because of the common problem of avoiding the precipitation of molybdenum and other noble metals during feed preparation and during the co-decontamination cycle. In the processing of uranium-plutonium-molybdenum

fuels, precipitation was avoided by reducing the concentration of alloying components by dilution of the feed with nitric acid until a stable solution was obtained.⁷ In the preparation of feeds from uranium-plutonium-molybdenum alloys, in 6.5M nitric acid, uranium concentrations greater than 100 g/liter resulted in the formation of precipitates that contained uranium, plutonium, and molybdenum.

c. Codecontamination

The solvent-extraction processing of irradiated-fuel feeds containing high concentrations of plutonium has received little attention. The available information indicates that the permissible solvent loadings of plutonium is not unduly restrictive. A solvent loading of 80 g/liter of uranium and 40 g/liter of plutonium appears to be feasible.⁵ Other data show that a solvent containing 20% tributyl phosphate in equilibrium with 3M nitric acid containing 50 g/liter of plutonium will contain 35 g/liter of plutonium. Molybdenum is not extracted by the solvent.⁸

d. Partition of Uranium and Plutonium

The adjustment of the plutonium valency to Pu(III) is required to partition uranium and plutonium. In conventional Purex processing, ferrous ion is employed to reduce plutonium. However, because of the high concentration of plutonium in the fast-reactor fuels under consideration, the use of ferrous ion would result in unacceptably high contamination of the product with iron.

An alternate reductant, which might be acceptable, is uranous ion.⁹⁻¹¹ About two moles of hydrazine-stabilized uranium (IV) ion are required per mole of plutonium. Uranium contamination of the plutonium product to about 1% is not considered objectionable, because in a subsequent step the plutonium product is blended with a portion of the uranium product to reconstitute the core fuel composition. In the conceptual flowsheet, about 16% of the uranyl nitrate product is reduced electrolytically to provide uranium (IV) for the reduction of plutonium. The conceptual flowsheet has not been tested at these high plutonium concentrations. The concentrations and quantities of U(IV) nitrate required are much greater than those that have been tested previously.

e. Subsequent Solvent-extraction Steps

After the partition of uranium and plutonium, the plutonium product undergoes no further solvent-extraction processing in the conceptual flowsheet. The uranium product is subjected to further solvent-extraction processing to yield a product fully decontaminated from fission products. Conventional Purex processing is employed in subsequent uranium processing.

f. Conceptual Aqueous Process Flowsheets for Fast-reactor Fuel

All the Purex-type flowsheets that have been proposed require modification for the processing of the fast-reactor fuels considered in this study. The conceptual aqueous process, on which the costs developed in this report are based, is a flowsheet proposed for the Idaho small-plant concept¹² modified to accommodate criticality restrictions, uranium (IV) for plutonium reduction, fission-product solubility restrictions, and solvent-loading restrictions. Stream specifications for the conceptual aqueous-processing flowsheet are presented in Figs. A.1 through A.4. Core and blanket fuel are processed simultaneously. The numbers shown are for a plant serving a single reactor.

Combined core and blanket fuel is dissolved in batches in a continuous-circulation dissolver (Fig. A.1). The operational-criticality restriction is 5 kg of Pu (239 + 241) per batch, and the dissolution time cycle is 16 hr. Consequently, more than one dissolver is required in any plant serving two or more reactors. The composition of the dissolver solution is controlled by the operational-criticality restriction of 14 g/liter of total plutonium (10 g/liter of fissile plutonium). The restriction limiting the concentration of fission products to 8 g/liter is not controlling.

The codecontamination cycle (Fig. A.2) consists of an extraction column, a scrub column, and a stripping column. More than 99% of the fission products are removed in the aqueous high-activity waste (HAW) from the extraction column. Uranium and plutonium are extracted with 20% tributyl phosphate in kerosene and stripped with 0.01M nitric acid. The aqueous product is concentrated in an intercycle evaporator, and the concentrate is adjusted to provide a feed for the partition cycle.

Fig. A.1. Aqueous Processing: Dissolution

Fig. A.2. Aqueous Processing: Codecontamination Cycle

Fig. A.3. Aqueous Processing: Partition Cycle

Fig. A.4. Aqueous Processing: Uranium Third Cycle

The partition cycle (Fig. A.3) provides a second cycle of decontamination and the separation of uranium and plutonium. Decontamination is obtained in the 1A extraction and 1B scrub columns. The effluent rates and compositions are identical with those employed in the decontamination cycle. Uranium and plutonium are separated in the 1B partition column. The plutonium extractant, which contains 221 g of uranium (IV) per liter is introduced in the lower portion of the column just above the entry point for the feed. A 0.5M nitric acid scrub stream is introduced at the top of the partition column. The aqueous product from the partition column, which contains the plutonium, is scrubbed with 20% tributyl phosphate in the 1B scrub column. The plutonium product contains 30.5 g of fissile plutonium (239 + 241) per liter (43.1 g total plutonium per liter) and is collected in geometrically safe slab tanks. Uranium is stripped from the organic product in the 1C column.

Further decontamination of the uranium product is obtained in the third uranium cycle (Fig. A.4). The conditions of the standard Purex flowsheet¹³ are employed with appropriate modification to accommodate the use of 20% instead of 30% tributyl phosphate as the extractant. To avoid more than one solvent inventory, 20% tributyl phosphate is employed.

g. Solvent-radiation Damage

The three major effects of radiation damage to the TBP-diluent solvent are:

- 1) Poor stripping of uranium and plutonium from the organic stream.
- 2) Lowering of fission-product decontamination factors due to retention of fission products in the organic phase.
- 3) Formation of emulsions and extraneous solids, which decrease extraction efficiency and increase the likelihood of flooding.

A number of studies were made in the late 50's on radiation effects in solvent extraction processes. Although radiation doses employed in some studies were far in excess of those encountered in actual processing operations, the most pertinent observations seem to be those based on process experience. Rigg and Weld¹⁴ stated that plutonium and fission-product retention in the solvent may be expected to be a problem at radiation doses in excess of 0.7 W-hr/liter, and that above 10 W-hr/liter, the precipitation of solids leads to product losses, emulsification, and poor phase separation. Bruce¹⁵ stated that the solvent-extraction processes had been successfully demonstrated at exposure levels of 0.5 W-hr/liter and would probably be feasible at 3.4 W-hr/liter. At 1 W-hr/liter, a twofold decrease in fission-product decontamination occurred. No further decrease occurred as the exposure was increased to 6 W-hr/liter, but at 6 W-hr/liter, serious emulsion problems were encountered. Goode¹⁶ reported that noticeable drops in decontamination occurred at radiation doses of 0.5 W-hr/liter, and indicated that in future applications of solvent extraction, radiation doses would have to be held to less than 10 W-hr/liter.

More recent work has not changed this picture significantly. Ishihara, Tsujino, and Yamamoto showed that decontamination from zirconium and niobium is markedly decreased as solvent-exposure levels go above about 1 W-hr/liter.¹⁷ Emulsion formation increases at exposure above 1 W-hr/liter. Orth and Alcott¹⁸ reported on the effect of solvent exposure on the performance of the Purex process at the Savannah River Plant. Process performance was improved by substituting Adakane (a normal paraffin diluent consisting largely of dodecane) for Ultrasene (a kerosene-type diluent). Even with this improvement, 180-day cooling is still specified for fuels that have undergone 0.5% burnup of the heavy atoms (as compared to the 10% burnup of this study).

For the fuel burnups considered in this study, the heat dissipation of heavy elements is about 0.2 kW/kg. The heat generation in blanket elements is less than 10% of that of the core material. Therefore, the

energy-generation rate in the combined material would be about 0.1 kW/kg or 10,000 W/100 kg (10 to 20 W/liter of feed solution). These heat-generation rates would be reduced by about a factor of three by cooling 120 days, and only a fraction of the energy would be absorbed in the organic phase. Contact times of 1 hr or more could be allowed in the contactor without exceeding a solvent dose of 5 W-hr/liter. Thus, solvent damage due to beta-gamma radiation will probably not be a significant factor, except in the codecontamination cycle. Solvent management procedures to maintain the quality of the solvent were incorporated in the conceptual aqueous-plant operational philosophy. The required solvent treatment and other factors associated with the high radiation level have a significant effect on reprocessing costs.

References for Section 1 of Appendix A

1. C. D. Watson, B. C. Finney, G. A. West, and B. A. Hannaford, *The Shear-Leach Process for Spent Nuclear Fuels*, Trans. Am. Nucl. Soc. 6(2):408-409 (1963).
2. J. A. Buckham, R. S. P'Pool, and R. G. Pitkin, *Conceptual Study of a System for Radioactive Demonstration of the Chop-Leach Process*, IDO-14610 (Aug. 9, 1963).
3. A. L. Uriarte and R. H. Rainey, *Dissolution of High-Density UO₂, PuO₂, and UO₂-PuO₂ Pellets in Inorganic Acids*, ORNL-3695 (April 1965).
4. L. A. Hanson, *Removal of Irradiated UO₂ From the Cladding by Controlled Oxidation*, NAA-SR-3591 (Aug 1959).
5. D. M. Donaldson, K. Harley, P. Lees, and N. Parkinson, *Reprocessing Fast Reactor Fuels at Dounreay*, Trans. A.I.M.E. 227:191-202 (1963).
6. R. P. Larsen, R. S. Shor, H. M. Feder, and D. S. Flikkema, *A Study of the Explosive Properties of Uranium-Zirconium Alloys*, ANL-5135 (July 1954).
7. F. L. Culler, R. E. Blanco, L. M. Ferris, E. L. Nicholson, R. H. Rainey, and J. Ullman, *Proposed Aqueous Processes for Power Reactor Fuels*, Nucleonics 20(8):124-127 (1962).
8. J. F. Flagg, *Chemical Processing of Reactor Fuels*, Academic Press, New York and London, p. 40 (1961).
9. C. S. Shlea, M. R. Caverly, H. E. Henry, and W. J. Jenkins, *Uranium (IV) Nitrate as a Reducing Agent for Plutonium (IV) in the Purex Process*, DP-808 (April 1963).
10. H. A. C. McKay, R. J. W. Streeton, and A. G. Wain, *Mixer-Settler Runs to Study Uranium (IV) as a Reductant in Uranium/Plutonium Separation*, AERE-R-4381 (Sept 1963).
11. T. R. Bhat and B. S. Mathur, *Studies of the Preparation and Properties of Uranium (IV) Nitrate*, Indian Journal of Technology 1(1):14 (Jan 1963).

12. H. Schneider, R. D. Fletcher, J. W. Coddling, R. G. Bearsden, E. E. Erickson, S. J. Horn, M. E. Jacobson, R. F. Murray, and E. L. Rowe, *A Study of the Feasibility of a Small Scale Reprocessing Plant for the Dresden Nuclear Power Station*, IDO-14521 (April 28, 1961).
13. V. R. Cooper and M. T. Walling, Jr., *Aqueous Processes for Separation and Decontamination of Irradiated Fuels*, Prog. Nucl. Energy, Ser. III, 3:229-288 (1961).
14. T. Rigg and W. Weld, *Radiation Effects in Solvent Extraction Process*, Prog. Nucl. Energy, Ser. III, 2:320-331 (1958).
15. F. P. Bruce, *Chemical Processing of Aqueous Blanket and Fuel From Thermal Breeder Reactors*, Chem. Eng. Prog. 52(9):347-352 (1956).
16. J. H. Goode, *How Radiation Affects Organics in Solvent Extraction of Fuel*, Nucleonics 15(2):68-71 (1957).
17. T. Ishihara, T. Tsujino, and Y. Yamamoto, *Radiation Damage to TBP/Kerosene-Solvent Effect on Behavior of Fission Products and Nitric Acid*, AEC-tr-5071 (July 1962).
18. D. A. Orth and T. V. Olcott, *Purex Process Performance Versus Solvent Exposure and Treatment*, Nucl. Sci. Eng. 17(4):593-612 (1963).

2. Critique of Fluid-bed Fluoride-volatility Processing

Fluoride-volatility processing as a type of process is identified by the use of the volatile hexafluorides of uranium and/or plutonium to effect separations of these fuel actinides from one another and from fission products. Fluid-bed volatility processing employs fluid-bed techniques to effect heat transfer and the transport of materials as required during fluorination or other chemical operations involved in processing. The principal steps in the reference fluid-bed volatility process are the following: (1) chemical decladding; (2) fluid-bed fluorination; (3) volatility separations; (4) collection and disposal of fission-product fluorides; and (5) conversion of volatile hexafluorides of uranium and plutonium to forms suitable for fuel refabrication. Before considering these principal steps with reference to fast-reactor fuel processing, we will review briefly some important demonstrations of applications of fluoride-volatility processing to the nuclear fuel cycle.

a. Review of Applications of Fluoride-volatility Processing

Interest in fluoride-volatility processing began early in the U. S. Atomic Energy program because of the role of volatile uranium hexafluoride in the production of the fissionable isotope ^{235}U by means of gaseous diffusion. The aim of development of the fluoride-volatility process was to lower the cost of processing by eliminating aqueous processing steps.¹ Interest in fluid-bed techniques in the nuclear industry also originated several years ago, since they offered more economical means of conducting the principal steps of feed-materials production, namely, reduction of higher

oxides of uranium to uranium dioxide and conversion of UO_2 to uranium tetrafluoride (UF_4) by reaction with gaseous hydrogen fluoride.² The compound UF_4 served as an intermediate for the subsequent production of either UF_6 or uranium metal.

Fractional distillation as a means of purifying UF_6 was also investigated early. For example, in early pilot-scale experiments at ORNL and ANL, irradiated uranium was dissolved in liquid fluorinating agents such as chlorine trifluoride (ClF_3) and bromine trifluoride (BrF_3), and the resulting UF_6 was separated by distillation in packed towers to a high degree of purity from both fission products and the halogen constituents.³ These experiments demonstrated virtually complete fission-product decontamination by means of fractional distillation. They also demonstrated techniques for handling UF_6 and halogen fluorides (ClF_3 , BrF_3 , etc.) in both condensed and vapor phases. The vapor-liquid equilibrium of the system UF_6 - BrF_3 was determined by Liimatainen.⁴ Pilot-scale work at ANL demonstrating high-purification UF_6 from mixtures containing BrF_3 was reviewed by Ivins⁵ as part of his study of small-scale distillation over the full range of UF_6 - BrF_3 compositions.

Fluid-bed fluorination and fractional-distillation techniques have been successfully applied to the large-scale production of purified UF_6 from ore concentrates. Very stringent purity specifications were placed on the UF_6 product to make it acceptable as feed material for isotopic enrichment in the gaseous-diffusion plant. A commercial plant using this process for UF_6 purification was built and operated by the General Chemical Company (a subsidiary of Allied Chemical Company) at Metropolis, Illinois. Large-scale fluid beds and bubble-cap distillation towers were employed in this plant, which had a capacity of about 20 tons of UF_6 per day.^{6,7}

b. Chemical Decladding

Fluoride-volatility techniques are applicable to the chemical removal of cladding alloys of current interest, including stainless steel, Zircaloy, and vanadium-20% titanium. Of these alloys, stainless steel is of interest for both thermal and fast reactors, while Zircaloy is of current interest only for thermal reactors. The vanadium-titanium alloy is a high-temperature, high-strength alloy, representative of a type of refractory-metal alloy compatible with metal fast-reactor fuel.

Chemical decladding allows simple mechanical head-end operations. Specifically, the requirement of mechanical decanning or chopping of fuel rods into short segments is avoided. The only mechanical head-end operations required in connection with chemical decladding are dismantling of the fuel assemblies and trimming away the excess hardware from the

fuel rods; thus, the fuel is still in the clad state when charged to the fluorinator for decladding. The decladding operations for the two clads of interest in this study are reviewed below.

(1) Stainless-steel Decladding. Equimolar mixtures of oxygen and hydrogen fluoride at about 550°C have been demonstrated in pilot-scale tests to be effective for decladding Types 304 and 347 stainless steel-clad fuel. Cladding with 13-mil wall thickness was completely reacted in 3 hr in the presence of a fluid bed of granular alumina. The product of the decladding operation is usually in the form of small, thin pieces of stainless-steel oxides, which settle to the bottom of the fluid bed.⁸ During the subsequent fluorinations, in which the uranium and plutonium are removed from the bed as volatile hexafluorides, the decladding debris disintegrates to a powder. The waste bed is free-flowing and can be drained from the fluid-bed reactor. This cladding waste consists of nonvolatile fluorides of iron, nickel, and chromium, mixed with the alumina and nonvolatile fission-product fluorides. Some of the chromium forms volatile fluorides in the fluorination step. These volatile fluorides leave the fluorinator in the gaseous effluent with the hexafluorides of uranium and/or plutonium.⁹ Although the fate of these volatile chromium fluorides has not been established in experiment, volatile chromium fluorides and oxyfluorides have volatilities substantially different from UF_6 , and it was assumed in this study that adequate separation from the UF_6 and PuF_6 products can be achieved by the volatility-separations steps incorporated in the reference process. The requirements for decladding stainless steel with HF -oxygen mixtures under fully reliable conditions are still under development. The work that has been completed indicates that the process is feasible.

(2) Vanadium-Titanium Alloy Decladding. The vanadium-20% titanium alloy specified for metal-fuel cladding in this study is not readily attacked by oxygen-hydrogen fluoride mixtures, and consequently, a decladding step was adopted that is different from that used for stainless steel. Both vanadium and titanium form volatile fluorides with either BrF_5 or fluorine. In the reference process for volatility processing of metal fast-reactor fuel, the vanadium-titanium decladding is accomplished in the early stages of the fluid-bed fluorination using BrF_5 as the fluorinating agent. The volatile decladding products (VF_5 , bp 48°C ; TiF_4 , bp 284°C) are separated from product UF_6 (bp 56°C) in the subsequent volatility-separation steps.

The metal fuel rods contain sodium metal as a thermal bond between fuel and cladding. The sodium is assumed to be present in an amount equal to 1.5 and 1.0 w/o of the total amount of uranium, plutonium, and fission products in the core and blanket fuel, respectively. As individual rods are declad, the sodium is converted to the fluoride, which disperses in the fluid bed. Not all the sodium will react at one time, and the presence of the fluid bed is expected to minimize both the movement of the sodium as liquid and the buildup of local hot spots due to the fluorination reaction.

This decladding operation has not been tested experimentally, and a possibility exists for problems, such as bed caking, due to the presence of the sodium metal.

(3) Decladding and Subsequent Oxidation of Carbide Fuel. The carbide fuel is assumed to be clad in stainless steel. No sodium metal is present in the fuel rod as a thermal bond. The same decladding procedure is used for the cladding of the carbide fuel as with that of the oxide fuel. Carbide fuel is exposed to the oxygen-hydrogen fluoride mixture, and the carbide reacts with oxygen. In the reference process, the carbon leaves the reactor as gaseous carbon dioxide, and the uranium and plutonium remain in the form of solid oxides and fluoride. The uranium and plutonium-bearing residues are subsequently fluorinated in the same manner as the residues of oxide fuel following decladding.

The fluid-bed oxidation of pyrolytic carbon-coated, enriched UC_2 granules in a graphite matrix has been conducted on a pilot scale at Brookhaven National Laboratory. Granular alumina was used as an inert fluid-bed material. The process, carried out satisfactorily at $750^\circ C$, was deemed promising for continuous operation.¹⁰ Preliminary experiments at Mol, Belgium, suggest that complete oxidation of dense UC pellets with air may be difficult to achieve.¹¹ However, recent results at Mol show equimolar O_2 -HF to be a promising reagent; in these experiments, dense UC was converted to UF_4 and UO_2F_2 at $500^\circ C$.¹²

The oxidation behavior of dense monocarbides corresponding to core and blanket fuel of a fast reactor has not been sufficiently investigated experimentally to define optimum reaction conditions. It was assumed that the batches of fuel rods could be reacted virtually completely with oxygen-hydrogen fluoride mixtures in about 14 hr. It was assumed further that temperatures employed in this reaction would be low enough to allow operation in a nickel reactor.

The possibility of exposing unreacted carbide to BrF_5 vapor in the subsequent fluorination does not appear to have any serious practical difficulties. In such a reaction, the uranium would be converted to UF_6 vapor, and the carbon would be converted to CF_4 or to a mixture of CF_4 and CO_2 , if oxygen as well as BrF_5 were present. The reaction would readily go to completion, and the subsequent condensation system would recover the UF_6 and send the noncondensable gases (CF_4 or CO_2) to waste.

c. Fluid-bed Fluorination

The conceptual fluid-bed fluorinations considered in this report are discussed below in terms of the types of the fuel undergoing fluorination.

(1) Metal Fuel. In the case of metal fuel, the fluorination of the metal is performed directly to yield hexafluoride product. Only exploratory experimental work has been carried out on the direct fluorination of uranium metal with fluorine. The fluorination reaction of uranium as metal is known to proceed rapidly at about 120°C in liquid-phase studies.³ Analogous fluid-bed processes converting aluminum and zirconium to volatile chlorides have shown satisfactory operation.^{13,14} In the direct fluorination of metal fuel, it was assumed that practical fluorination behavior in fluid beds was similar to that for oxide fuel, except that the difference in the heat released in the chemical reaction was considered. For example, in the specification of equipment size and of the time required in batch processing, the rate of heat release rather than the chemical kinetics was rate controlling for the UF_6 production step.

(2) Oxide and Carbide Fuel. Since carbide fuel is oxidized before fluorination starts, the fluid-bed fluorination step for oxide fuel applies to both types of ceramic fuel. Experimental investigations of the fluid-bed volatility processing have been directed toward oxide fuel of compositions typical of that discharged from thermal power reactors. Such thermal fuel approximates the composition of blanket fuel discharged from a fast reactor, but the plutonium and fission-product content of thermal fuel is substantially less than that present in the core fuel of the reference fast reactor adopted in this study. Only very limited experimental work has been carried out on fuel compositions approximating that of the reference core fuel. In these preliminary experiments oxidation and fluorination reactions of dense PuO_2 - UO_2 pellets are slower with higher contents of plutonium or fission products.¹⁵ Since such a large fraction of the fuel forms volatile fluoride products, no serious difficulty was assumed in carrying out the steps of the reference process. In the balance of this section the basic features of the fluid-bed fluorination will be briefly reviewed in terms of experimental results to date.

Recent experimental work has been conducted at ANL on the fluid-bed volatility processing of simulated dense oxide pellet fuel with a composition of UO_2 -0.5% PuO_2 . The simulated fuel also contained fission products corresponding to fuel irradiations of about 15,000 MWD/metric ton. In experiments employing the BrF_5 fluorination, uranium hexafluoride production rates of about 100 lb of UF_6 per (hr) (sq ft of reactor cross-sectional area) were obtained at 300 to 400°C. In this reaction with BrF_5 , plutonium remains as a nonvolatile residue and does not accompany the volatile UF_6 product. In a subsequent processing period, the fluorination of plutonium and recovery of PuF_6 were performed. In this PuF_6 production step, the recycle of fluorine gas for 5 hr at no higher than 550°C reduced the residual plutonium content of the fluid bed to about 7×10^{-5} g of plutonium per gram of alumina fluid bed.¹⁶ For the processing batch size assumed in this study for fast-reactor fuel, namely, about 250 kg of alumina and 11 kg of plutonium (total plutonium) the above residual concentration of plutonium in the bed is equivalent to a loss of about 0.15% of the

plutonium in the solid waste stream from the fluid bed. For fast-reactor fuel, higher initial concentrations of plutonium and fission products in the fuel are expected to result in a slightly higher concentration of residual plutonium than the value cited above. The plutonium losses to the solid waste stream would be correspondingly higher in this case. However, it was assumed that a plutonium loss of about 1/2% would be allowable for processing the fuel, and that process losses would be held to this amount in the reference process.

The reaction of bromine pentafluoride with the oxide fuel has been shown to yield bromine as the principal product.¹⁷ However, the effluent from the fluorinator may also contain some unreacted BrF_5 and some of the intermediate fluoride BrF_3 . In the reference process, the bromine and BrF_3 products are refluorinated with elemental fluorine to regenerate BrF_5 , which is separated and reused in the process. The reactions are well known since they are used in the commercial production of bromine fluorides. The refluorination may be presented by the equations

The scheme of refluorination adopted in this study entails a partial refluorination (i.e., of bromine to BrF_3) in the fluorinator itself and a completing of the regeneration of BrF_5 in a separate, gas-phase, fluorination reactor. By dividing the refluorination (to form BrF_5) between the fluid-bed fluorinator and the separate gas-phase reactor (not fluidized), the size and cooling requirement of the separate gas-phase reactor can be substantially reduced without impairing the performance of the overall process.

The separate gas-phase reactor is supplied with reagent fluorine in slight excess of the stoichiometric requirements and is operated at about 400°C. The thermodynamic properties and the chemical kinetic behavior of the system have been determined in laboratory investigations.¹⁸ The reaction of Eq. (1) proceeds faster than the reaction of Eq. (2); therefore, the rate of fluorination of BrF_3 (Eq. 2) is rate-controlling for BrF_5 production. The net products of the fluorination appear in the following two streams:

(a) A UF_6 product stream containing UF_6 , BrF_5 , oxygen, and nitrogen. This stream is completely free from plutonium.

(b) A PuF_6 product stream, containing PuF_6 , fluorine, some UF_6 , and a comparatively small amount of oxygen or nitrogen diluent. The UF_6 is produced by fluorination of residual uranium from the previous

fluorination step. Substantial amounts of UF_6 are allowable in the plutonium stream since, in the reference process, the plutonium product may contain as much uranium as the reference uranium-plutonium composition of recycled core fuel.

d. Volatility Separations

For this critique, volatility separations are considered to include all process steps that follow fluorination and to contribute to the separation of uranium, plutonium, and fission products. These steps comprise the condensation of uranium and plutonium hexafluorides from the gaseous effluent from the fluorinator and all distillation operations. Also included in the metal-fuel process is the step for the recovery of plutonium as nonvolatile PuF_4 from a volatile liquid mixture initially containing PuF_6 .

(1) Condensation of Uranium Hexafluoride from Fluorinator Gaseous Effluent. The uranium hexafluoride product from the fluorinator is condensed as a liquid UF_6 - BrF_5 mixture and is thereby separated from the noncondensable diluent gases. For this separation to be carried out with low losses of UF_6 and BrF_5 to the noncondensable oxygen-nitrogen waste-gas stream, condenser temperatures of 0 to -30°C are desired. The requirement of maintaining a liquid condensate also imposes a restriction, namely, the minimum ratio of BrF_5/UF_6 in the condenser feed. For the above condenser temperature limits, the minimum BrF_5/UF_6 mole ratios are 4 at 0°C and 10 at -30°C . The physical properties of the UF_6 - BrF_5 system have been determined.¹⁹ It is possible, however, that UF_6 can be condensed as a solid, forming a slurry in BrF_5 . In this case, the composition restriction would be relaxed.

This scheme for liquid condensation of UF_6 was selected in preference to solid condensation of UF_6 because of the advantages of smaller equipment and greater ease of handling the condensate. Liquid condensers can be smaller than solid condensers of the same heat duty, because (in the case of liquid condensation) the product is not retained on the heat-transfer surface, but is simply collected in a reservoir as it is formed. The liquid product may readily be transferred to subsequent processing steps. In the case of the solid condenser (or cold trap), the removal of solid entails shutdown of the condenser and a separate heating step to melt or vaporize the condensate for transfer.

For the PuF_6 condensation, different schemes were employed for the oxide fuel than for the metal and carbide fuel. For the oxide fuel, PuF_6 was condensed as a solid. For the metal and carbide fuel, PuF_6 was condensed as a liquid PuF_6 - BrF_5 mixture, in a manner similar to that described above for UF_6 - BrF_5 condensation.

The condensation of solid PuF_6 has somewhat less disadvantage than for UF_6 (as described above). This is so because PuF_6 is handled in relatively smaller batches (about 10 kg of plutonium) with correspondingly small condenser capacities. By employing solid PuF_6 condensation, subsequent equipment for $\text{PuF}_6\text{-BrF}_5$ separation (as entailed in the liquid condensation system) could be avoided with the oxide fuel. On the other hand, in the processing of metal and carbide, the liquid $\text{PuF}_6\text{-BrF}_5$ condensation scheme was selected to facilitate transfer of PuF_6 from the condenser and to provide a suitable feed material for the subsequent step for reduction of PuF_6 to PuF_4 . (This reduction step is required for both metal and carbide fuel. Metal is an intermediate product in the carbide synthesis.)

Condensation of hexafluorides, both as solid and liquid, is a well-developed operation. Equipment for condensing UF_6 as a solid has been operated in a wide range of sizes for uranium of all enrichments. Rather bulky designs are required to accommodate the deposited solid product, and provisions for heating are required to remove the product by melting or vaporization during shutdown periods. In general, special provisions are required in cold traps to prevent "snow" formation and its undesirable consequences, such as plugging of the gas exit or loss of product. Condensers of baffled design have been used extensively at ORNL,²⁰ and an annular hot-and-cold wall design was used in pilot-scale work at ANL.²¹

The decomposition of PuF_6 to form PuF_4 is a special consideration throughout the volatility-separations portion of the process. This decomposition is governed by the equilibrium.

The equilibrium constant and other properties of PuF_6 have been summarized by Steindler.²¹

In the fluorinator, high temperature and excess fluorine drive the equilibrium to the left and thereby produce PuF_6 . But when the PuF_6 is separated from fluorine (as it is in all condensed phases), the PuF_6 tends to decompose to reach equilibrium. However, below about 90°C the rate of approach to this chemical equilibrium is very slow. This is not to say that the actual rate of decomposition is unimportant at low temperatures, because under these conditions there is an appreciable decomposition rate due to alpha radiation. In the condensed phase, the alpha radiation is responsible for a decomposition rate of about 2% per day.²² This radiation-induced decomposition rate (unlike that of chemical decomposition) is independent of temperature. This decomposition will occur throughout the volatility-separations portion of the plant; the total rate of decomposition

thus depends on the inventory of PuF_6 in this system. The deposited plutonium (as PuF_4) can be recovered from the equipment by fluorinating the equipment and lines at about 350°C (or higher) by converting the PuF_4 to PuF_6 , which is then collected in a cold trap. It is clearly desirable to minimize the number of pieces of equipment, the quantity of PuF_6 holdup, and the processing time, in order to minimize the PuF_6 decomposition and the associated plutonium recovery operations.

(2) Distillation of UF_6 . In the reference process, fractional distillation in a packed tower is used to prepare a UF_6 product to serve as feed to the subsequent step for fluid-bed conversion to UO_2 . This distillation separates UF_6 from the bulk of the BrF_5 and from virtually all the fission-product radioactivity carried with the UF_6 from the fluorination step. The physical properties, distillation performance, and fission-product decontamination have been demonstrated in previous pilot-scale work.²⁻⁵

In the present study a single batch-fractionation tower was specified with a diameter of 8.0 in. and a packed height of 9 ft, and an equivalent of about 16 theoretical plates. This number of plates was selected for the UF_6 - BrF_5 separation; fewer plates would be required if the objective were only to provide decontamination of radioactive fission products.

One fission product, ^{99}Tc , forms a fluoride (TcF_6) with a volatility so close to that of UF_6 that separation by fractional distillation would be difficult. An alternative separation has been proposed for the reference process, namely, selective sorption of technetium fluoride on a bed of magnesium fluoride. Experimental results have been reported for the removal of technetium in the effluent from the fluorination of uranium oxide containing technetium. However, the technetium compound absorbed on magnesium fluoride has not been identified specifically as TcF_6 .²³ In the present process application, technetium (because of its very low specific activity) is regarded primarily as a chemical impurity rather than a contributor to radiation.

(3) Distillation of PuF_6 . In oxide-fuel processing, the volatility-separations operations include three instances of the simple batch distillation of PuF_6 : (1) the removal of PuF_6 from the condenser; (2) the subsequent transfer of this PuF_6 into a small cold trap, and the revaporization into a feed vessel for the subsequent fluid-bed conversion step; and (3) the vaporization of the PuF_6 as feed for the conversion of PuF_6 to PuO_2 .

Although each of these distillations is essentially a simple batch transfer, the steps nevertheless afford opportunity to remove volatile impurities as "heads" and "tails." Because of their high plutonium content, these heads and tails are not considered waste streams, but are recycled to the fluorinator to be processed in the next complete batch cycle. Since the PuF_6 is purified by the above steps, a separate fractional distillation for PuF_6 is not specified in the reference process for the oxide fuel.

In metal and carbide-fuel processing, the PuF_6 from the fluorination is collected as a liquid $\text{PuF}_6\text{-BrF}_5$ mixture. A packed fractionation tower is provided to enrich the mixture in PuF_6 to serve as feed to the subsequent PuF_6 reduction step. The fractionation provides additional decontamination from fission products. This tower is approximately 3 in. in diameter and 5 ft high.

Fractional distillation has not been demonstrated for mixtures containing PuF_6 . Although PuF_6 has physical properties similar to those of UF_6 , the consequences of PuF_6 decomposition have not been evaluated. If difficulties are encountered with the liquid $\text{PuF}_6\text{-BrF}_5$ system described here, the solid PuF_6 scheme described above for the oxide fuel case could be used, with about the same equipment cost.

(4) Reduction of PuF_6 to PuF_4 . In processing metal and carbide fuel, one of the volatility-separation operations involves the separation and recovery of PuF_4 from a $\text{PuF}_6\text{-BrF}_5$ liquid mixture. This operation includes the reduction of PuF_6 with bromine or bromine trifluoride as follows:

Excess reductant insures complete conversion of the PuF_6 to PuF_4 . All products except PuF_4 are volatile and are distilled off and reused in the process. The PuF_4 product is mechanically removed and charged as feed to the subsequent step, the pyrochemical reduction to metal.

The above batch reduction of PuF_6 is done in a closed system. The $\text{PuF}_6\text{-BrF}_5$ mixture is added to bromine at room temperature or lower. Cooling is provided for temperature control. The temperature of the system may be raised to increase the rate of reaction. The pressure is not expected to go above 1 atm. This reduction has not been conducted experimentally. However, from knowledge of the thermodynamic properties and of the kinetics of the reaction of bromine with other strong fluorinating agents (such as fluorine^{18,22}), the reaction should proceed readily.

e. Collection and Disposal of Fission-product Fluorides

High-efficiency removal of halogen compounds from gas streams can be attained by reaction with commercial soda lime or activated alumina.²⁴ The desirable characteristics of such beds are their active chemical nature as basic oxides, their granular form, which allows passage of gas through packed beds, and their porosity and high surface area. Thus, a high degree of chemical conversion can be obtained in these systems.

Such beds can be employed with practical capacities of 30 to 90% of theoretical conversions. Both soda lime and activated alumina are used to take advantage of their combined effectiveness for removal of volatile fluorides from waste-gas streams. The absorbents are expected to be effective for all volatile fission products, including iodine and tellurium (as TeF_6).²⁴ In plant practice, a series of beds is used to provide efficiency and safety.

f. Conversion of Volatile Hexafluorides of Uranium and Plutonium to Forms Suitable for Fuel Fabrication

The fluid-bed process adopted in this study for the direct production of uranium dioxide from hexafluoride has been demonstrated on the pilot scale. The procedure consists of a period in which hexafluoride vapor is fed into the fluid bed, followed by a period during which residual traces of fluoride are removed from the fluidized solid product. Mixtures of steam and hydrogen serve as the reagent gases in both steps. Product is removed semicontinuously (that is, after each fluoride cleanup period) so that the bed weight is approximately constant. The formation of seed particles is required to compensate for particle-size growth, and this is accomplished by periodically conducting a partial oxidation of the UO_2 bed to U_3O_8 powder. Particles with densities of 8.5 to 9.5 g/cc have been produced at 650°C by this method.^{25,26}

The above fluid-bed conversion was utilized in the reference process for blanket fuel of all three types. In the metal and carbide fuel, the UO_2 product served as a feed for the subsequent step for reduction to metal. In the oxide fuel, the UO_2 product is used directly for fuel fabrication. A high-temperature firing (1600 to 1725°C for about $2\frac{1}{2}$ hr) can be used to densify the particles. Final particle densities of about 10.4 to 10.6 g/cc were produced in this way.²⁶

In the reference process, some BrF_5 remains with the UF_6 feed to the converter. It was assumed that the BrF_5 would be reacted to form HF and bromine without appreciable effect on the UO_2 product.

It was assumed that the same basic fluid-bed conversion could be used to produce PuO_2 - UO_2 solid-solution product for core fuel. In this case, the feed would consist of a PuF_6 - UF_6 vapor mixture. The conversion and characterization of the product have not yet been demonstrated experimentally.

g. Engineering Design and Scale-up

The plant design and cost estimates for the fluid-bed volatility process were made in detail for the case of processing oxide fuel for a single 1000-MWe reactor. This basic plant was modified for the other fuels and plant sizes. The general plant-processing scheme was based on batch

charging of 2-day reference loads of fuel (combined core and blanket). This batch size was chosen primarily because it was reasonably large, but well within restrictions imposed by nuclear safety. The 2-day batch (as based on Table I) consists of 8.0 kg of ^{239}Pu and ^{241}Pu and 11.2 kg of total plutonium. This restriction on batch size was applied to all the plutonium-processing steps, but not to other portions of the processing plant.

The main functional units of the processing plant can be grouped as follows:

1. A fluorinator system in which decladding, oxidation, and primary and secondary fluorinations were performed. This system included equipment for collection of hexafluoride products. The fluorinator was 13 in. in diameter, and the overall height of the fluorinator, including the disengaging and filter section, was about 12 ft. The required cooling load was 120,000 Btu/hr, maximum.

2. A fractional distillation system, in which uranium hexafluoride, bromine fluorides, and volatile fission-product fluorides were separated. The fractionation tower was 8 in. in diameter and contained a 9-ft section packed with nickel packing (perforated saddles). The required heat duty was estimated to be 34,000 Btu/hr.

3. Equipment for the fluid-bed conversion of uranium and plutonium hexafluorides to dioxide products for refabrication into fuel elements for the core and blanket. The converter was 7 in. in diameter and 7 ft in overall height.

The operations in these three major divisions of the plant, including material transfers, were coordinated on 48-hr cycles.

For estimating the costs of the volatility plants of larger sizes, the plan of scale-up was determined by nuclear safety considerations. For the larger throughput plants, those portions of the plant that contained plutonium were not increased in size; throughput increase was provided by increasing the number of units of equipment. The portions of the plant affected by this restriction were the fluorinator, the PuF_6 collection system, and the converter for UO_2 - PuO_2 core fuel. For the portions of the plant not containing plutonium, the items of equipment for which increased throughput was obtained by increasing the size of single units were the UF_6 collection system, the distillation tower, auxiliary equipment for handling bromine fluorides, and the converter for blanket oxide fuel.

The estimated time schedules for the decladding and fluorination steps are as follows:

<u>Process Step</u>	<u>Reagent Gas</u>	<u>Process Time, hr</u>		
		<u>Oxide Fuel</u>	<u>Carbide Fuel</u>	<u>Metal Fuel</u>
Chemical decladding	O ₂ -HF	5	14	-
Fluorination to form UF ₆	BrF ₅	5	5	19
Fluorination to form PuF ₆	F ₂	15	15	15

Note that for metal fuel, the chemical decladding was included in the fluorination with BrF₅.

In addition to the above time requirements, an allowance of 2 to 4 hr was estimated for required solid-charging, gas-purging, and solid-discharging operations. For all fuels, the fluorinator process cycle was less than the 48-hr reference cycle time for the single-reactor processing case.

For the three- and ten-reactor cases, the actual estimated cycle time, not the nominal 48-hr period, was used in determining the number of separate lines of equipment required. Superficial velocities of the fluidizing gas (at 1 atm and the process temperature) were 1.0 ft/sec for the decladding and conversion steps and 0.75 ft/sec for the fluorination steps.

The tower for the fractional distillation of UF₆ was designed to operate on a 48-hr cycle and was scaled up in size for the various plant capacities. The superficial vapor velocity was 0.35 ft/sec at 2.5 atm pressure in the conceptual design. This velocity was employed in pilot-scale distillation operations.

The size and shape of the disposable waste containers were limited by maximum allowable temperature; therefore the same-size units were employed for all throughputs.

Appendix I contains a detailed flowsheet for the fluid-bed volatility process for the processing of the oxide fuel of a single 1000-MWe reactor.

References for Section 2 of Appendix A

1. S. M. Stoller and R. B. Richards, editors, *Reactor Handbook, Volume II, Fuel Reprocessing (2nd ed.)*, Interscience Publishers, Inc., New York, New York, p. 250 (1961).
2. *Reactor Handbook, Volume II*, op. cit., p. 347.
3. *Reactor Handbook, Volume II*, op. cit., pp. 285-286.

4. R. C. Liimatainen, *Vapor-liquid Equilibrium in the System Bromine Pentafluoride-Uranium Hexafluoride*, ANL-6003 (May 1959).
5. R. O. Ivins, *The Distillation of Uranium Hexafluoride and Bromine Pentafluoride in a 0.5-inch-diameter Packed Column*, ANL-6362 (May 1962).
6. *Reactor Fuel Processing* 4(3):58 (July 1961).
7. *Reactor Fuel Processing* 6(4):3 (Fall 1963).
8. R. C. Vogel, M. Levenson, and F. R. Masten, *Chemical Engineering Division Research Highlights, May 1964-April 1965*, ANL-7020, pp. 123-127 (1965).
9. D. Ramaswami, N. M. Levitz, and A. A. Jonke, *Engineering Development of Fluid-bed Fluoride Volatility Processes. Part 2. Bench-scale Investigation of a Process for Aluminum-Uranium Alloy and Stainless Steel-Cermet Fuels*, ANL-6830 (Dec 1965).
10. *Reactor Fuel Processing* 8(2):90-91 (Spring 1965).
11. *Reactor Fuel Processing* 9(2):44-49 (draft) (Spring 1966).
12. *Reactor Fuel Processing* 9(3):63 (draft) (Summer 1966).
13. *Reactor Fuel Processing* 8(2):88-90 (Spring 1965).
14. John T. Holmes, Howard Stethers, and John J. Barghusen, *Engineering Development of Fluid-bed Fluoride Volatility Processes. Part 8. Pilot-plant Development of a Process for Uranium Alloy Fuels*, ANL-6973 (Aug 1965).
15. *Reactor Fuel Processing* 8(2):93 (Spring 1965).
16. R. C. Vogel, M. Levenson, J. H. Schraidt, and J. Royal, *Chemical Engineering Division Research Highlights, May 1965-April 1966*, ANL-7175, pp. 49-53 (1966).
17. R. M. Adams and A. Glassner (Comps.), *Reactor Development Program Progress Report, March 1966*, ANL-7193, pp. 75-76 (April 22, 1966).
18. H. E. Kluksdahl and G. H. Cady, *J. Am. Chem. Soc.* 81, 5285 (1959).
19. *Reactor Handbook, Volume II*, op. cit., pp. 250-265.
20. *Reactor Handbook, Volume II*, op. cit., pp. 301-302.
21. S. Lawroski, R. C. Vogel, M. Levenson, and V. H. Munnecke, *Chemical Engineering Division Summary Report, October, November, December 1962*, ANL-6648, pp. 144-148 (May 1963).
22. M. J. Steindler, *Laboratory Investigations in Support of Fluid-bed Fluoride Volatility Processes. Part II. The Properties of Plutonium Hexafluoride*, ANL-6753 (Aug 1963).
23. W. R. Golliher, U. S. Pat. 3,165,376 (Jan. 12, 1965).
24. R. C. Vogel, M. Levenson, J. H. Schraidt, and J. Royal, *Chemical Engineering Division Research Highlights, May 1965-April 1966*, ANL-7175, p. 96 (1966).

25. I. E. Knudsen, H. E. Hootman, and N. M. Levitz, *A Fluid-bed Process for the Direct Conversion of Uranium Hexafluoride to Uranium Dioxide*, ANL-6606 (Feb 1963).
26. I. E. Knudsen, N. M. Levitz, and A. A. Jonke, *Engineering Development of Fluid-bed Fluoride Volatility Processes. Part 6. Preparation of Dense Uranium Dioxide Particles from Uranium Hexafluoride in a Fluidized Bed*, ANL-6902 (Dec 1964).

3. Pyrochemical Processing Critique

The overall objective in the reprocessing of spent reactor fuels is to restore the nuclear and physical properties of the fuel so it can be returned to the reactor. The specific technical objectives, in the case of a fast breeder reactor, are: (a) to remove fission products from the discharged fuel and provide for their disposal; (b) to recover bred plutonium from the irradiated blanket material; (c) to re-enrich the core fuel with plutonium from the blanket, and (d) to recover the reprocessed core and blanket material in a form that is suitable for refabrication and return to the reactor. The purpose of this critique is to review briefly the technological status of pyrochemical processes for uranium-plutonium fuel cycles employing oxide, carbide, and metallic fuels.

Complete removal of the fission products is not needed to restore the nuclear properties of reactor fuels. Therefore, pyrochemical processes, which in their present state of development provide only partial fission-product decontamination, are suitable for use with power-reactor fuels. Because only partial decontamination is achieved, remote refabrication of fuel elements and assemblies is required. However, the buildup of radioactive uranium and plutonium isotopes in extensively recycled fuels may necessitate semiremote or possible fully remote refabrication, regardless of the fission-product decontamination achieved during processing. Somewhat higher fission-product decontamination factors in the range of 10^3 to 10^4 are obtained in the more advanced salt-transport process described in Appendix B. However, these advanced processes were not sufficiently developed to be included in the formal cost comparisons of the present study.

One advantage of pyrochemical processes, which is shared to a large extent by the fluoride-volatility processes, is the use of reagents and solvents that are not subjected to radiation damage. This feature permits high-burnup, short-cooled fuels to be processed and recycled to the reactor in a short time, thereby minimizing the out-of-reactor fuel inventory. A 15-day cooling period is considered adequate for pyrochemical processes.

Other advantages of pyrochemical processes are: (a) the use of compact equipment, (b) less severe critical-mass restrictions because of the absence of neutron-moderating materials, and (c) the direct collection

of radioactive wastes as solids. The main disadvantages are: (a) low fission-product decontamination, (b) the use of high temperatures, and (c) the limited availability of suitable container materials.

Although the earlier pyrochemical processes were developed specifically for metallic fuels, the more recent processes have been directed mainly toward ceramic fuels.¹⁻³ In these newer processes, uranium and plutonium are separated and decontaminated by procedures that employ liquid-metal and molten-salt solvents at 500 to 1400°C. Both physical and chemical procedures are used for the process separations. The physical separations include liquid-metal distillation and retorting, and the fractional crystallization of solutes from liquid-metal solutions. The chemical separations generally make use of differences in the partition behavior of solutes between liquid-metal and salt phases.

This critique is restricted to pyrochemical processes that employ cadmium, zinc, magnesium, or combinations thereof as liquid-metal solvents. An important consideration underlying the choice of these metals as solvents is that they are readily separated from the metallic uranium and plutonium products of the process by volatilization. The molten-salt solvents used in these processes usually consist primarily of magnesium chloride, which may be diluted with alkali-metal or other alkaline-earth chlorides.

In the pyrochemical processes, it is convenient to class the fission-product elements that exhibit similar behavior in the following five groups:

Volatiles:	Xe, Kr, I, Te
Alkali metals and alkaline earths:	Rb, Cs, Sr, Ba
Group III elements:	Y, rare earths
Refractory metals:	Nb, Zr
Noble metals:	Mo, Tc, Ru, Rh, Pd, Ag, Cd, In, Sn, Pb.

The volatile fission products are released when the mechanical integrity of the fuel is destroyed, e.g., during the oxidative decladding of ceramic fuels, and during the melting or dissolution of metal fuels. The fission products that are included in this class depend upon the conditions. For example, xenon and krypton are gases that will be released as indicated above. However, the release and collection of iodine, tellurium, and cesium will depend upon the temperature and the chemical environment. These latter elements can be trapped either by condensation or by suitable chemical traps. The handling of volatile fission products is discussed in greater detail in Section III-C in the body of this report.

The alkali-metal and alkaline-earth fission products, because of their strongly electropositive character, appear in the salt wastes. Yttrium

and the rare earths can be directed either to a salt waste or a metal waste, as desired, by the appropriate choice of metal and salt solvents. The refractory-metal fission products in the reference pyrochemical processes must be removed by a special procedure, such as volatilization of their chlorides.* The noble-metal fission products appear in a metal-waste stream. Thus, the fission products other than the volatiles behave as would be expected on the basis of their position in the electrochemical series.

In the following portions of this critique, the status of the individual separations steps of the pyrochemical processes is discussed briefly.

a. Decladding

(1) Metal Fuels. In the EBR-II Fuel Cycle Facility, mechanical decladding is used routinely for the removal of sodium-bonded, stainless-steel jackets from the enriched uranium alloy fuel.⁴ Although some difficulty was encountered in the early operation of this step, modification of the equipment has overcome these initial problems.⁵ The release of volatile fission products to the argon atmosphere of the process cell during this operation has been sufficiently small to cause no significant problem.

Chemical procedures for the separation of metal fuel and its cladding have been considered as an alternative possibility. These procedures consist of (a) dissolution of the cladding fuel elements as a whole, followed by a chemical separation of the cladding constituents at some point in the process, (b) selective dissolution of either the cladding or the fuel in a liquid-metal or molten salt, and (c) oxidative decladding, in which the metallic fuel is converted to a mixture of oxides. Insufficient information is available at present on items (a) and (b) to warrant their selection for the reference processes, although scouting studies have been started on this possibility. The oxidative decladding of metal fuels would probably be technically feasible, but it would require an additional reduction step in the process to convert the resulting oxides back to the metals.

(2) Ceramic Fuels. It is not practical to declad ceramic fuels mechanically, because of the lack of structural integrity of the irradiated fuel. The most attractive alternative procedure is oxidative decladding. In this operation, the stainless-steel cladding is punctured at intervals, followed by exposure to air or oxygen at 400 to 600°C. The resulting oxidation of uranium dioxide to U_3O_8 causes a volume increase of about 30% that pulverizes the fuel and splits the cladding.^{6,7} Vibration techniques may be used to assist in the removal of the powdered fuel from the cladding hulls. If necessary, further pulverization of the fuel is possible through alternate

*In the salt-transport process (Appendix B), zirconium and niobium appear in the metal-waste streams.

oxidation with oxygen at 400 to 600°C and reduction with hydrogen at 600°C. The oxidative decladding of stainless steel-clad uranium dioxide fuel, irradiated to burnups of up to 20,000 MWD/metric ton, has been demonstrated at Atomics International.^{3,8,9} The oxidative decladding of Zircaloy-clad uranium dioxide and mixed uranium and plutonium oxides, irradiated to a burnup of 5,000 MWD/metric ton, has been demonstrated at Hanford.¹⁰ In general, the recoveries were greater than 99.5% and usually greater than 99.9%. However, no work has been reported on the oxidative decladding of mixed uranium-plutonium dioxide fuel having a high plutonium content (15-20%) characteristic of a fast-reactor fuel, nor has the operation been fully developed for remote, plant-scale operation. The indications are that the oxidative decladding of oxide blanket material would offer little difficulty, but further information is needed on the oxidative decladding of mixed uranium and plutonium oxide having a plutonium content typical of an oxide core fuel.

The oxidative decladding of carbide fuel involves much the same procedure as for oxide fuel. However, the operation has not been developed as fully for carbides as for oxides. Essentially all the investigations in this area have been conducted at Atomics International, where the oxidative decladding of stainless steel-clad uranium monocarbide was demonstrated on a 750-g scale, using flowing air at 400°C as the oxidizing agent.^{9,11} Some additional experiments showed that sodium used as a thermal bond in stainless steel-clad uranium carbide fuel can be removed by vacuum evaporation at 600°C after the cladding has been punctured.¹² A substantial amount of more general information on the oxidation of uranium monocarbide has been reported. For example, it was found that the oxidation rate depends markedly upon the carbon-uranium ratio of the material, whether it has been "activated" by prior exposure to a moist atmosphere, and whether it has been irradiated.¹¹ The oxidation of uranium and thorium carbides in fixed and fluid beds has been investigated at Oak Ridge and Brookhaven National Laboratories.^{13,14}

Some uncertainty exists as to whether adequate oxidation rates can be achieved with highly compacted and sintered oxide and carbide fuels. Additional work is also needed on the effect of plutonium at concentrations up to about 20%, the influence of irradiation, and the development of remote, plant-scale operations.

b. Extraction of Noble-metal Fission Products from Oxide Mixtures

In the pyrochemical process for ceramic fuels, the oxide mixture from the oxidative decladding step is suspended in a molten chloride salt, which is then contacted with liquid zinc at 750°C. The noble-metal fission products are selectively reduced and extracted into the zinc phase.

This operation has been developed and demonstrated in pilot-plant investigations of the EBR-II skull-reclamation process.¹⁵ These investigations, which employed mixtures of U_3O_8 and inactive oxides of fission-product elements, showed noble-metal fission-product removals in the range of 80 to 90% and uranium losses of less than 0.1%. Similar demonstrations with inactive material, conducted in remotely-operable equipment designed for installation in the EBR-II Fuel Cycle Facility, gave approximately the same results.¹⁶ However, the noble-metal extraction step for oxide and carbide fuels has not yet been demonstrated with uranium-plutonium combinations, nor with irradiated material.

The equipment required for this operation includes a container, an agitator, a transfer tube, and a bell jar with heaters.¹⁷ The only fully satisfactory container material is tungsten. Various techniques for the fabrication of large tungsten containers are currently being developed with considerable success.

c. Reduction of Oxides by Liquid Magnesium-Zinc Alloys

In the pyrochemical process for oxide and carbide fuels, the molten salt from the noble-metal extraction step is contacted with liquid magnesium-zinc alloy at 750°C in the vessel that was used for the noble-metal extraction. Uranium oxides have also been reduced on a pilot-plant scale,^{15,18} and in the remotely-operable plant-scale equipment in the EBR-II Fuel Cycle Facility.¹⁶ With a magnesium chloride-based salt of the proper composition and adequate mixing of the liquid-metal and salt phases, over 99% reduction of uranium oxides is achieved consistently. On a 5-kg scale, the reduction can be completed in 2 hr or less. The solid magnesium oxide formed as a by-product of the reduction is readily suspended in a salt phase, where it can be removed as a slurry. Reduction studies with oxides of the major fission-product elements show that they behave as indicated in the process diagram in Fig. 8. The presence of plutonium dioxide in mixed uranium-plutonium oxide fuels is not expected to hinder the reduction, since the laboratory investigations showed plutonium dioxide to be more easily reduced than the uranium oxides.^{18,19} Although the reduction step has not been demonstrated with highly irradiated materials, it is believed to be feasible in remotely operated, plant-scale equipment.

d. Fission-product Separations by Liquid-metal-salt Extraction

In the pyrochemical process for metal fuels (Fig. 7) the core fuel is melted and contacted at 1250°C with molten calcium chloride containing a small, controlled amount of magnesium chloride, which preferentially oxidizes the alkali-metal, alkaline-earth, and rare-earth fission products. This halide-slugging operation results in the extraction of these fission products into the salt phase. The magnesium metal formed as a by-product of the reaction is volatilized as it is formed, thereby preserving

the driving force of the reaction. Halide slagging has been investigated at Los Alamos²⁰ for the processing of LAMPRE fuel (a liquid plutonium alloy), and at Argonne for the separation of the above-mentioned fission-product elements from molten uranium-plutonium alloys.^{21,22} In the latter studies, which utilized beryllia crucibles as the container, cerium removals were over 99.5% when a small amount of magnesium chloride was added to a calcium chloride-barium chloride salt phase. The plutonium loss to the salt was less than 0.5%, and the volatilization of plutonium was negligible. The pouring yields for the molten-metal phase were 98% when new crucibles were used and 100% for reused crucibles.

There appears to be little doubt as to the chemical feasibility of the halide-slagging step, and much of the engineering development has been in connection with the EBR-II melt-refining process. However, the halide-slagging operation requires careful control of the magnesium chloride concentration in the salt phase in order to avoid excessive plutonium losses in the salt. It may prove difficult to ascertain the fission-product content of the fuel in order to determine the amount of magnesium chloride to be used. Another problem may arise in that high-integrity, plant-scale crucibles are not readily available, although there has been considerable progress recently in fabrication techniques. The halide-slagging procedure still awaits remote, plant-scale demonstration with irradiated fuel.

The pyrochemical process for oxide and carbide fuels also employs a liquid-metal, molten-salt extraction procedure for the separation of rare-earth fission products from uranium and plutonium. However, in this instance, the liquid-metal phase is a cadmium-zinc-magnesium solution of uranium and plutonium, and the molten-salt phase consists mainly of magnesium chloride. A substantial body of chemical information on the distribution behavior of pertinent elements between magnesium alloys and magnesium chloride-based salts is available as a result of studies at Brookhaven,²³⁻²⁵ Ames,²⁶ Hanford,²⁷ and Argonne.²⁸⁻³¹ Nevertheless, additional chemical information is needed on the distribution coefficients of uranium, plutonium, and rare earths in the particular liquid-metal-salt combinations used in this process.

The extraction is carried out at about 750°C in a continuous-flow column with a 9-ft packed section. The column provides two scrub and four extraction stages, which are needed for adequate removal of the rare earths and satisfactory plutonium recovery. Although some preliminary work has been done on liquid-metal, molten-salt extraction kinetics, a substantial additional effort is needed both on fundamental engineering studies and on equipment design.³²⁻³⁵ Initial results from investigations that are currently in progress at Argonne indicate that the height of a theoretical stage is about 1.5 ft for liquid-metal-salt systems. For the process solvents indicated, stainless steel is expected to be a satisfactory containment material.

The rare-earth elements in the molten-salt effluent from the column are extracted by aluminum-magnesium alloy to permit recycle of the salt. This extraction is a relatively simple, single-stage operation. This operation has been demonstrated on a laboratory scale,³⁶ but has not been conducted in larger-scale equipment.

In the process for metal fuels, uranium and plutonium are chlorinated by cadmium chloride. This reaction produces cadmium metal. The oxidizing power of cadmium chloride is such that uranium and plutonium form the chlorides, which are extracted into the molten-salt phase, whereas the noble-metal fission products remain in the liquid cadmium as the metals. Zinc chloride can also be used for this purpose, but it has the disadvantages of a higher vapor pressure, which leads to fuming, and the fact that the zinc metal formed by the reaction is more corrosive than cadmium. The chlorination of metallic uranium by cadmium chloride in a molten-salt phase has been used occasionally in laboratory studies,²² but it has not yet been developed as a process operation. However, the engineering development of this step appears to be relatively straightforward. Some provision may be necessary to confine the cadmium chloride vapor, but suitable equipment for this purpose has been developed in connection with the EBR-II skull-reclamation process.⁵

e. Separation of Refractory-metal Fission Products

No provision is made for the routine removal of zirconium and niobium in the pyrochemical processes. Instead, they are removed periodically by chlorination with cadmium chloride at 650°C, in the presence of a molten chloride salt. If a molten chloride salt of the proper composition is selected, its complexing tendency will be such that uranium and plutonium chlorides will remain in the salt solution while the volatile chlorides of the refractory metals are vaporized.³¹ This procedure has a rather unattractive aspect in that provision must be made to condense the chlorides of the refractory-metal fission products and convert them into a form that is suitable for disposal. Essentially no development work has been done on this procedure, since the more advanced salt-transport processes presently under development remove niobium and zirconium with the noble-metal fission products (see Appendix B).

f. Separation of Uranium and Plutonium

Uranium and plutonium have relatively high solubilities in cadmium and magnesium alloys of low magnesium content. However, at high magnesium concentrations (above about 30%) the solubility of uranium becomes very small. For example, at 650°C the solubility of uranium is about 6.0% in a zinc-22% magnesium alloy and 0.03% in a zinc-70% magnesium alloy. Consequently, uranium can be separated from plutonium by fractional crystallization. The plutonium is separated from the precipitated uranium by transferring off the supernatant liquid-metal solution.

In the pyrochemical process for metal fuels, a magnesium-rich solution is produced by vacuum evaporation of cadmium and zinc from a cadmium-zinc-magnesium solution of uranium and plutonium. The plutonium in the magnesium-rich supernatant solution is separated from the precipitated uranium by pressure siphoning.

Since a magnesium-rich zinc-magnesium alloy is used for the reduction of the oxidized fuel in the process for ceramic fuels, the uranium is precipitated as it is reduced, while the reduced plutonium remains in the liquid-metal solution. The supernatant solution is removed by pressure siphoning, as indicated above.

The fractional crystallization method for separating uranium and plutonium has been used in the proposed liquid-metal process for the recovery of bred plutonium from the EBR-II blanket material. The chemical and engineering aspects of this separation have been investigated on a pilot-plant scale,²⁹ but the procedure has not been developed for remote, plant-scale application.

For a metal blanket, halide slagging is a possible alternative method for the recovery of plutonium. The blanket material is melted under a molten chloride salt containing a small amount of magnesium chloride to extract the electropositive fission-product elements as indicated above. The plutonium is then extracted by a second molten chloride salt containing uranium trichloride as the oxidant. Laboratory-scale studies have shown that this procedure results in a satisfactory extraction of plutonium from molten uranium.^{31,38} However, the engineering development of this application of halide slagging has been inhibited by the lack of fully satisfactory container materials.

g. Liquid-metal Distillation and Retorting

In the pyrochemical processes for both metallic and ceramic fuels, the metallic uranium and plutonium products from the separation steps are recovered from the liquid-metal solvents by retorting. The retorting procedure involves vacuum evaporation of the solvent metals, followed by a melting step to consolidate the product. Retorting equipment has been developed for remote operation in the EBR-II Fuel Cycle Facility.³⁹ However, retorting equipment of much larger capacity is required for the pyrochemical processes used in this study. Although the feasibility of large-scale retorting has been demonstrated in industrial applications, a substantial amount of development work is needed to adapt the techniques to remote operation in a fuel-reprocessing plant. An attractive feature of the salt-transport processes is that they do not require large-capacity retorts (see Appendix B).

References for Section 3 of Appendix A

1. S. Lawroski and L. Burris, Jr., *Processing of Reactor Fuel Materials by Pyrometallurgical Methods*, Atomic Energy Rev. 2(3), 3 (1964).
2. R. D. Pierce and L. Burris, Jr., "Pyroprocessing of Reactor Fuels," in *Reactor Technology, Selected Reviews - 1964*, L. Link (Ed.), p. 441, USAEC Division of Technical Information Extension, Oak Ridge, Tenn., (July 1964).
3. L. Burris, Jr., K. M. Harmon, G. E. Brand, E. W. Murbach, and R. K. Steunenberg, *Pyrometallurgical and Pyrochemical Fuel Processing*, presented at the Third International Conference on the Peaceful Uses of Atomic Energy, Geneva, Aug. 31-Sept. 9, 1964, Paper A/CONF. 28/P/251.
4. J. C. Hesson, M. J. Feldman, and L. Burris, Jr., *Description and Proposed Operation of the Fuel Cycle Facility for the Second Experimental Breeder Reactor (EBR-II)*, ANL-6605 (April 1963).
5. R. M. Adams and A. Glassner (Comps.), *Reactor Development Program Progress Report, December 1965*, ANL-7132, p. 9 (Jan. 25, 1966).
6. L. A. Hanson, *Removal of Irradiated UO₂ Fuel from the Cladding by Controlled Oxidation*, NAA-SR-3591 (Aug 1959).
7. L. J. Colby, Jr., K. L. Mattern, H. Pearlman, E. W. Murbach, and G. E. Brand, *Comparative Fuel Cycle Evaluations. Low Decontamination Pyroprocessing vs. Aqueous Reprocessing. Part I. UO₂ Fuel in a Thermal Reactor*, NAA-SR-8036 (Feb. 1, 1963).
8. Reactor Fuel Processing 7(1), 47 (1964).
9. J. E. Bodine, I. J. Groce, J. Guon, and L. A. Hanson, *Oxidative Decladding of Uranium Dioxide Fuels*, Nucl. Sci. Eng. 19, 1 (1964).
10. Reactor Fuel Processing 7(2), 122 (1964); 8(1), 32 (1965); 8(2), 103 (1965).
11. E. W. Murbach and G. E. Brand, *Pyrochemical Reprocessing of Uranium Carbide - Summary Report*, NAA-SR-11340 (Aug. 31, 1965).
12. I. J. Groce, *Decladding of UC Fuel Rods*, NAA-SR-TDR-7883 (Nov 1962).
13. C. D. Scott, L. E. McNeese, and J. C. Suddath, *Processing of Graphite Fuels for Uranium Recovery by Oxidation and Fluorination*, ORNL-TM-185 (March 20, 1962).
14. Reactor Fuel Processing 7(3), 181 (1964).
15. L. Burris, Jr., I. G. Dillon, and R. K. Steunenberg, *The EBR-II Skull Reclamation Process. Part I. General Process Description and Performance*, ANL-6818 (Jan 1964).
16. R. M. Adams and A. Glassner (Comps.), *Reactor Development Program Progress Report, January 1966*, ANL-7152, p. 42 (Feb. 24, 1966).
17. Reactor Fuel Processing 7(4), 256 (1964).
18. J. B. Knighton and R. K. Steunenberg, *Preparation of Metals by Magnesium-Zinc Reduction. Part I. Reduction of Uranium Oxides*, ANL-7057 (June 1965).

19. J. B. Knighton and R. K. Steunenberg, *Preparation of Metals by Magnesium-Zinc Reduction. Part III. Reduction of Plutonium Dioxide*, ANL-7059 (June 1965).
20. L. J. Mullins, J. A. Leary, and W. J. Maraman, *Removal of Fission Product Elements by Slagging*, Ind. Eng. Chem. 52, 227 (1960).
21. G. A. Bennett, L. Burris, Jr., and R. C. Vogel, *Halide Slagging of Uranium-Plutonium Alloys*, ANL-6918 (July 1964).
22. R. C. Vogel, M. Levenson, and F. R. Masten, *Chemical Engineering Division Semiannual Report, January-June 1965*, ANL-7055 (Oct 1965).
23. O. E. Dwyer, *Process for Fission Product Removal from Uranium-Bismuth Reactor Fuels by Use of Fused Salt Extraction*, AIChE Journal 2, 1963 (1956).
24. W. S. Ginell, *Oxidation Extraction of Lanthanide Metals from Molten Bismuth by Fused Salts*, Ind. Eng. Chem. 51, 185 (1959).
25. F. J. Salzano and F. B. Hill, *Distribution of Uranium and Cerium Between Liquid Bismuth-Magnesium Alloy and Liquid NaCl-KCl-MgCl₂ Eutectic*, BNL-639 (June 1962).
26. P. Chiotti and S. J. S. Parry, *Separations of Various Components from Uranium by Oxidation-Reduction Reactions in a Liquid Potassium Chloride-Lithium Chloride/Zinc System*, J. Less-Common Metals 4, 315 (1962).
27. R. H. Moore and W. L. Lyon, *Distribution of Actinide Elements in the Molten System KCl-AlCl₃-Al*, HW-59147 (Oct 1959).
28. J. B. Knighton and R. K. Steunenberg, *Distribution of Selected Elements Between Magnesium Chloride and Magnesium/Zinc Alloy*, Trans. Am. Nucl. Soc. 5(2), 460 (1962).
29. S. Lawroski, R. C. Vogel, M. Levenson, and V. H. Munnecke, *Chemical Engineering Division Research Highlights, May 1962-April 1963*, ANL-6766 (1963).
30. R. C. Vogel, M. Levenson, and V. H. Munnecke, *Chemical Engineering Division Research Highlights, May 1963-April 1964*, ANL-6875 (1964).
31. R. C. Vogel, M. Levenson, and F. R. Masten, *Chemical Engineering Division Research Highlights, May 1964-April 1965*, ANL-7020 (1965).
32. H. M. Katz, F. B. Hill, and J. L. Spiers, *Kinetics of Exchange Reactions Between Liquid Bismuth-Rare Earth Alloys and Fused Salts*, Trans. Met. Sec. AIME 218, 770 (1960).
33. F. B. Hill and L. E. Kukacka, Jr., *Axial Mixing and Mass Transfer in Fused Salt-Liquid Metal Extraction Columns*, BNL-791 (Jan 1963).
34. P. R. Josephson and L. Burkhard, *Development of Countercurrent Multi-stage, Fused Salt-Molten Metal Extractor*, IS-420 (July 1961).
35. P. M. C. Lacey, G. F. Hewitt, R. Murdock, B. A. Partridge, J. A. Lacey, and M. E. Darlington, *Studies of Fused Salt/Liquid Metal Techniques Applicable to Uranium and Other Purifications*, Trans. Inst. Chem. Eng. 41, 352 (1963).
36. J. B. Knighton, R. A. Mancini, and R. K. Steunenberg, *Consolidation of Lanthanum Fission Products in Alloys*, ANL-7064 (Aug 1965).

37. H. M. Feder and R. J. Teitel, *Purification of Reactor Fuels and Blankets by Crystallization from Liquid Metal Solvents*, Prog. Nucl. Energy, Ser. III, 3, p. 355 (1960).
38. D. E. McKenzie, W. L. Elsdon, and J. W. Fletcher, *The Extraction of Plutonium from Neutron-Irradiated Uranium by Uranium Trichloride and by Magnesium Chloride*, Can. J. Chem. 36, 1233 (1958).
39. R. C. Vogel, M. Levenson, and V. H. Munnecke, *Chemical Engineering Division Semiannual Report, July-December 1963*, ANL-6800 (May 1964).

4. Product-conversion Procedures

In the aqueous processes, the blanket product is uranium dioxide and the core product is a uranium dioxide-plutonium dioxide mixture. The fluoride-volatility process for oxide fuels also produces uranium dioxide and a uranium dioxide-plutonium dioxide mixture. However, for carbide and metal fuels, the fluoride-volatility process yields uranium dioxide and plutonium tetrafluoride as separate products. The pyrochemical processes for oxide, carbide, and metal fuels all produce metallic uranium as the blanket product and a uranium-plutonium alloy as the core product.

It is therefore necessary to provide for the conversion of uranium dioxide, uranium-plutonium oxide mixtures, and plutonium tetrafluoride to both the metals and the carbides. The conversion of uranium and uranium-plutonium alloy to the corresponding oxides is also required.

a. Conversion of Oxides and Fluorides to Metals

The reduction of uranium dioxide and mixed uranium and plutonium oxides to their respective metals is an integral part of the reference flowsheets for the pyrochemical processing of oxide and carbide fuels. Since this step is included in the critique of pyrochemical process in Section 3 of this appendix, no further discussion is presented here. The same general procedure has been demonstrated on a laboratory scale for the reduction of uranium tetrafluoride,¹ and it would be expected to offer no difficulty in the reduction of plutonium tetrafluoride.²

In the fluoride-volatility process, plutonium metal is produced directly from the plutonium tetrafluoride, whereas the uranium hexafluoride product is first converted to uranium dioxide, which is reduced by the pyrochemical method. As an alternative procedure, the uranium hexafluoride could be reduced with hydrogen, for example, to form uranium tetrafluoride, which could be converted to the metal by the pyrochemical method.

In the pyrochemical conversions of oxide or fluoride to metal, the products are retorted to remove the liquid-metal solvent. Since retorting is also an integral part of the pyrochemical processes, the critique of this operation appears in Section 3 of this appendix.

b. Conversion of Metals to Carbides

In all three types of processing (aqueous, volatility, and pyrochemical), carbide fuels are regenerated by the conversion of metallic products to carbides, the oxide or fluoride products of the aqueous and volatility processes having first being converted to the metals.* Several procedures are available for the conversion of actinide metals to the carbides, e.g., arc-melting the metal with carbon,^{3,4} the direct reaction of the metal with graphite,⁵ carburization of the metal with hydrocarbon gases,⁶⁻⁸ reaction of the metal with charcoal in liquid-metal solutions.⁶

The method of carbide synthesis employed in this study is a fluid-bed carburization procedure, which appears to be suitable for the preparation of uranium monocarbide blanket material and mixed uranium-plutonium monocarbide core fuel.^{7,8} The respective feeds to the process are metallic ingots of uranium and uranium-plutonium alloy. The ingot is charged to a hydriding vessel, where it is exposed to a flowing hydrogen stream at a temperature of about 250°C. These conditions result in the formation of finely-divided metal hydride. The exit stream of hydrogen serves as a gas lift, which conveys the hydrided material into a fluid-bed reactor. A mixture of hydrogen with methane or propane is then passed through the fluid bed at 750°C to convert the hydride to carbide. By maintaining an appropriate methane-hydrogen ratio and operating at about 5 atm pressure, adequate conversion rates and the proper carbon-to-metal ratio of the product are achieved and the deposition of free carbon is avoided. The exit-gas stream from the fluid-bed reactor passes through filters to remove entrained solids. Material collected on the filters is returned to the bed periodically by a momentary reversal of the gas flow through the filters. The exit gas from the fluid-bed reactor is recycled by means of a compressor.

The preparation of uranium monocarbide by this procedure has been demonstrated in laboratory and pilot-plant studies. Thermobalance studies have indicated the chemical feasibility of preparing uranium-plutonium monocarbide mixtures by this technique, and a fluid-bed unit for the synthesis of mixed carbides on a pilot-plant scale is under construction. Although this process has not yet been fully developed for plant-scale use, it appears to be well suited to remote, automated-process applications. The carbide product from this process has suitable properties for fuel refabrication by pelletizing and sintering.

c. Direct Conversion of Oxides to Carbides

For this study, it is assumed that the oxides and fluorides of uranium and plutonium are converted first to the metals, then to the carbides, as indicated above. Since aqueous and fluoride-volatility processes can both produce oxide products, however, direct carbothermic reduction

*The direct conversion of oxides to carbides is discussed in Section c below.

of the oxides is an alternative possibility for carbide preparation. The carbothermic reduction procedure involves the reaction of uranium dioxide or U_3O_8 with carbon at about 1600 to 2000°C.⁹ Although this procedure is relatively well developed for the preparation of uranium monocarbide, considerably less experience is available on the preparation of mixed uranium-plutonium carbides. In the mixed carbide, the temperature of the reduction is limited to about 1625°C to avoid volatilization of the plutonium. The preparation of uranium-plutonium carbide mixtures are currently being investigated at the United Nuclear Corporation and at the Carborundum Company.¹⁰

d. Conversion of Metals to Oxides

In the pyrochemical process for oxide fuels, the uranium metal and the uranium-plutonium alloy resulting from the retorting operations are converted to uranium dioxide and a uranium-plutonium dioxide mixture, respectively. This procedure entails an oxidation of the metallic ingot with oxygen at 700°C, followed by reduction with hydrogen at 600°C. These operations are conducted in a small, resistance-heated furnace.

Other gases, such as steam or carbon dioxide might be utilized for the oxidation step. Oxygen was selected on the basis of experience with the skull oxidation step of the EBR-II skull-reclamation process¹¹ and the results obtained by Atomics International on oxidation-reduction cycles for uranium oxides.¹² In the development program on the EBR-II skull-reclamation process, laboratory and pilot-plant studies were conducted on the oxidation of melt-refining skulls, which consist largely of metallic uranium. These results were used in the design of a remotely operated, plant-scale, skull-oxidation furnace, which is being used routinely with highly irradiated fuel in the EBR-II Fuel Cycle Facility.¹³

A substantial background of chemical information on cyclic oxidation and reduction of uranium oxides has been developed at Atomics International in connection with the AIROX Process.¹² However, little information is available at present on the conversion of uranium-plutonium alloys to mixed oxides and their behavior during cyclic oxidation and reduction. Laboratory-scale investigations of these procedures have been initiated, but are still in an early stage.

References for Section 4 of Appendix A

1. J. B. Knighton and R. K. Steunenberg, *Preparation of Metals by Magnesium-Zinc Reduction. Part I. Reduction of Uranium Oxides*, ANL-7057 (June 1965).
2. J. B. Knighton and R. K. Steunenberg, *Preparation of Metals by Magnesium-Zinc Reduction. Part III. Reduction of Plutonium Dioxide*, ANL-7059 (June 1965).

3. O. L. Kruger, *Preparation and Some Properties of Arc-Cast Plutonium Monocarbide*, J. Nucl. Mater. 7(2), 142 (1962).
4. J. H. Kittel, L. A. Neimark, R. Carlander, O. L. Kruger, and R. C. Lied, *Preliminary Irradiation of PuC and UC-PuC*, ANL-6678 (Sept 1963).
5. *Annual Progress Report for 1964, Metallurgy Division*, ANL-7000 (1964).
6. J. Crane and E. Grodon, *The Development of Uranium Carbides as a Nuclear Fuel, Final Report*, UNC-5080 (Feb 1964).
7. R. C. Vogel, M. Levenson, and F. R. Masten, *Chemical Engineering Division Research Highlights, May 1964-April 1965*, ANL-7020 (1965).
8. R. C. Vogel, M. Levenson, J. H. Schraidt, and J. Royal, *Chemical Engineering Division Research Highlights, May 1965-April 1966*, ANL-7175 (1966).
9. Symposium on Carbides in Nuclear Energy, held at Harwell (Nov 1963):
 - a) R. Ainsley, P. C. Wood, and R. G. Sowden, *Some Aspects of the Preparation of Uranium and Plutonium Carbides from Oxides*, p. 540.
 - b) J. D. L. Harrison and J. W. Isaacs, *The Preparation of UC and (U,Pu)C by the Reduction of Oxides with Carbon in Flowing Argon*, p. 556.
 - c) J. M. Horspool, J. A. S. Mowat, and S. Pickles, *Development of Mixed Carbide Fuel Preparation and Fabrication Routes at DERE*, p. 572.
10. A. Strasser, *Carbide Fuel Development, Phase V Report*, UNC-5081 (June 1964).
11. T. R. Johnson, R. D. Pierce, L. Burris, Jr., and R. K. Steunenberg, *The EBR-II Skull Reclamation Process. Part II. Oxidation of Melt-refining Skulls*, ANL-6874 (April 1964).
12. L. J. Colby, Jr., K. L. Mattern, H. Pearlman, E. W. Murbach, and G. E. Brand, *Comparative Fuel Cycle Evaluations. Low Decontamination Pyroprocessing vs. Aqueous Reprocessing. Part I. UO₂ Fuel in a Thermal Reactor*, NAA-SR-8036 (Feb. 1, 1963).
13. R. C. Vogel, M. Levenson, and F. R. Masten, *Chemical Engineering Division Semiannual Report, January-June 1965*, ANL-7055 (Oct 1965).

APPENDIX B

Advanced Pyrochemical Processing Concept
and Preliminary Cost Estimate

In the body of this report, the cost estimates for the pyrochemical processing of oxide and carbide fuels were based upon the use of a modified version of the EBR-II skull-reclamation process, for which the technology has been developed. The more advanced pyrochemical processes, which are currently in the laboratory and development stage, employ a salt-transport procedure for the separation of uranium, plutonium, and fission products.¹⁻³

Salt-transport separations depend upon the selective transfer of solutes between two liquid-metal solutions that are equilibrated with the same molten salt. When a liquid-metal solution, saturated with a particular solute, is contacted with a molten salt, the equilibrium concentration of the solute in the salt phase is the product of its solubility in the metal phase and its distribution coefficient. Thus, upon equilibration of the salt phase with two liquid-metal phases of different composition, the solute will transfer to that metal phase over which the equilibrium concentration of the solute in the salt is lower, and the transfer will continue as long as the concentration gradient exists in the salt. The salt, in effect, serves only as a carrier for the solute transfer between the liquid-metal phases. To achieve rapid transfer of the solute, the salt phase is circulated between the two liquid-metal phases.

A salt-transport separation of uranium and plutonium is illustrated schematically in Fig. B.1. Uranium and plutonium are added initially to a Cu-6.5 w/o Mg alloy, say by reduction of the mixed oxides. Since uranium has a relatively low solubility (~4 w/o) in this alloy at 800°C, it is largely precipitated as finely divided metal. In the system indicated, the distribution coefficient and solubility relationships are such that uranium will transfer to the Zn-65 w/o Mg alloy and plutonium to the Zn-3 w/o Mg alloy. This transfer will continue until the solid uranium in the copper-magnesium alloy has all disappeared and the concentrations of both uranium and plutonium in this alloy have reached low values. Because of the low distribution coefficients of uranium and plutonium in the zinc-magnesium alloys, their concentrations in the salt phase are also small, with the result that the process losses are low (<1%).

Fig. B.1

Salt-transport Separation
of Uranium and Plutonium

The salt-transport process has several technological aspects, which could lead to a substantial economic advantage over the skull-reclamation process described in Section III.C. For example, the fact that only a small fraction of the uranium need be in solution at any one time permits the use of small solution volumes and compact equipment. Salt-transport processes also offer a marked improvement in product yields and in the separations of uranium, plutonium, and fission products. However, these processes are still in an early stage of development. Studies on a laboratory and small engineering scale have shown that the transfer of uranium takes place according to theory, and that good removals (>99%) of cerium and zirconium from uranium can be achieved in single-stage operations. Plutonium transfer has been demonstrated on a laboratory scale. At present, work is only beginning on the equipment engineering and conceptual plant design for salt-transport processes.

Figure B.2 illustrates a conceptual salt-transport process for discharged core and blanket material consisting of UO_2 - PuO_2 and fission products. This flowsheet involves the following steps:

1. The fuel is separated from the cladding by oxidation of the UO_2 . This procedure has not been demonstrated with UO_2 containing a substantial amount (15-20%) of PuO_2 , but it is believed to be feasible on the basis of work with UO_2 alone.
2. The uranium, plutonium, and fission-product oxides are charged to a vessel containing immiscible liquid phases of Cu-6.5 w/o Mg and NaCl-10 w/o MgCl_2 at 800°C. Essentially complete (>99%) reduction of the uranium and plutonium oxides takes place in this system, the bulk of the uranium precipitating as the finely divided metal in the copper-manganese phase. The solid MgO formed as by-product of the reaction remains suspended in the salt phase. At the completion of the reduction, the alkali-metal, alkaline-earth, and rare-earth fission products are also distributed to the salt phase, which is discarded as a waste. An additional extraction stage can be used both on the effluent salt stream and on the effluent metal stream to enhance rare-earth removal and to reduce plutonium losses.
3. The plutonium is then transported from the copper-magnesium alloy to a Zn-3 w/o Mg alloy by cycling a molten MgCl_2 phase between the two alloys.
4. After the plutonium transfer is completed, the uranium is transferred to a Zn-65 w/o Mg alloy by the same procedure.* The noble- and refractory-metal fission products remain behind in the copper-magnesium alloy, which is discarded as waste.
5. The zinc and magnesium are removed from the uranium and plutonium products by evaporation.

* The uranium and plutonium can be transferred to the zinc-manganese alloys simultaneously in theory, but sequential transfers appear preferable from an operational viewpoint.

6. The metallic uranium and plutonium products are reoxidized to regenerate oxides for fuel refabrication. The oxidation of uranium has been demonstrated, but additional information is needed on the oxidation of uranium-plutonium alloys. If core fuel is being processed, the metallic products may be combined along with make-up plutonium before the oxidation step. For blanket material, an appropriate amount of the plutonium product would be diverted to the core process and the remainder would be available as surplus. The uranium is converted to UO_2 and returned to the blanket.

Fig. B.2 Conceptual Salt-transport Process

With the appropriate head-end and tail-end modifications, this type of process is also applicable to carbide and metal fuels. For example, oxidative decladding might be used for carbide fuels, and the metallic products of the process might be reconverted to carbides by treatment with hydrogen and methane, or by oxidation followed by carbothermic reduction of the oxide. Metal fuels may be declad mechanically and hydrided to form a metallic powder suitable for introduction into the process.

A preliminary estimate was made of the savings that might be realized by adopting the salt-transport process in place of the reference pyrochemical process for ceramic fuel. (The reference process, a modified version of the skull-reclamation process, was the one for which comparative costs were developed in the body of this report.) The estimated savings of the salt-transport process for the processing of an oxide fuel

at a plant throughput of 22,700 kg/yr (actinides and fission products) in dollars/kg was 8.5, 2.3 and 28.9, respectively, for total labor, direct materials, and investment in plant equipment. The cost components for the salt-transport processing of oxide fuel are compared with those for alternative processes in Table B.I. The processing charge, which was developed by application of the economic ground rules of this study, was the lowest for volatility and pyrochemical salt-transport processing. Under the rules employed in this study, the difference in the processing charges for the volatility and salt-transport processes is not significant.

TABLE B.I. Cost Comparison of Pyrochemical Salt-transport Processing with Alternative Processes for Oxide Fuel

Plant Throughput: 22,700 kg/yr (actinides and fission products)

Cost Component	Unit Costs, \$/kg of Fuel Processed			
	Fluoride Volatility	Pyrochemical (Salt Transport)	Pyrochemical (Skull Reclamation)	Aqueous
Total labor	32.9	37.4	45.9	67.0
Direct materials	6.3	9.9	12.2	3.3
Investment in building and equipment	178.5	210.6	239.5	261.1
Processing charge	106.7	114.8	136.3	163.4

REFERENCES FOR APPENDIX B

1. R. M. Adams and A. Glassner (Comps.), *Reactor Development Program Progress Report, September 1965*, ANL-7105, p. 34 (Oct. 20, 1965).
2. R. M. Adams and A. Glassner (Comps.), *Reactor Development Program Progress Report, October 1965*, ANL-7115, pp. 38-40 (Nov. 23, 1965).
3. R. M. Adams and A. Glassner, (Comps.), *Reactor Development Program Progress Report, December 1965*, ANL-7132, pp. 24-28 (Jan. 25, 1966).

APPENDIX C

Estimated Investment Requirements and Operating Costs for Reference Plants

Investment requirements for buildings and equipment and operating costs were estimated for the cases of this study on the basis of specifications of processing load and reference flowsheets. The plant designs were discussed in Section IV and the cost models employed were cited in Section V of the main body of this report. Tables C.I through C.VI exhibit a breakdown of the estimated investment requirements of the plant components for the three processes. Table C.VII exhibits the investment costs of the basic waste disposal facilities.

Tables C.VIII, C.IX, and C.X summarize the operating costs for the three processes. The total operating costs were calculated from the total labor and direct-material cost estimates shown in the tables, with certain additions as specified in the ground rules of this study.

This appendix exhibits the cost estimates developed in the reference cases of this study. For both aqueous and volatility processing, fewer steps are required for oxide fuel than for carbide or metal fuel. For the basic oxide fuel plants, detailed estimates were made of the reprocessing costs for each of the three sizes of reference plants. Detailed estimates were also made for processing of carbide and metal fuel for the one-reactor plant. The costs for processing carbide and metal fuel for the three- and ten-reactor cases were calculated by scaling factors that were derived from the oxide processing case and individually applied to appropriately categorized cost components of the one-reactor plants for carbide and metal-fuel processing. This cost categorization for investment costs corresponds to the depreciation schedule. The cost categorization for operating costs corresponds to labor, direct materials, and other items.

A similar procedure was employed for pyrochemical processing, with the exception that the metal-fuel case was the one selected for the primary cost estimation of plants of the three reference sizes.

The objective of this study was to develop comparative costs, and no implication is intended that the estimate of costs in this appendix are predictions of actual plant costs. Appendix D provides additional information on unit-cost details.

TABLE C.I. Estimated Investment Requirements for Building and
Equipment for Aqueous Processing of Oxide Fuel
(80% plant factor).

	Investment Requirements in Thousands of Dollars		
	Annual Throughput: 22,700 kg/yr	Annual Throughput: 68,100 kg/yr	Annual Throughput: 227,000 kg/yr
A. General Facilities			
1. Offices, storerooms, and personnel convenience areas	278	393	600
2. Warehouse	50	75	123
3. Analytical laboratory	338	471	675
4. Maintenance facilities	550	814	1,322
5. Reagent-preparation facilities	341	586	1,079
6. Equipment disposal facilities	836	1,034	1,298
B. Remote-handling Cell			
1. Building	467	686	1,600
2. Transfer locks	167	167	167
3. Windows	150	150	150
4. Cranes and manipulators	122	143	170
5. Installed processing equipment	123	201	702
6. Instrumentation	19	45	117
7. Welding station	15	15	15
C. Processing Cell			
1. Building	748	1,524	3,243
2. Installed equipment	507	1,031	2,191
3. Instrumentation	214	339	592
D. Intermediate-activity-level Processing Facilities			
1. Building	129	249	515
2. Installed equipment	28	54	111
3. Instrumentation	3	4	6
E. Low-activity-level Processing Facilities			
1. Building	52	101	209
2. Installed equipment	194	374	775
3. Instrumentation	76	104	152
F. Auxiliary Equipment			
1. Argon purification and cooling	63	75	114
2. Electrical substation and emergency generator	72	108	176
3. Spare equipment	349	705	1,494
4. Mobile equipment	35	73	161
Total	5,926	9,521	17,757

TABLE C.II. Estimated Investment Requirements for Building and Equipment
for Aqueous Processing of Metal and Carbide Fuel
(80% plant factor)

	Investment Requirements in Thousands of Dollars	
	Metal Annual Throughput: 22,700 kg/yr	Carbide Annual Throughput: 22,700 kg/yr
A. General Facilities		
1. Office, storerooms, and personnel convenience areas	291	293
2. Warehouse	63	66
3. Analytical laboratory	338	338
4. Maintenance facilities	724	770
5. Reagent-preparation facilities	383	394
6. Equipment disposal facilities	836	836
B. Remote-handling Cell		
1. Building	690	850
2. Transfer locks	166	166
3. Windows	125	125
4. Cranes and manipulators	122	122
5. Installed processing equipment	211	286
6. Instrumentation	41	87
7. Welding station	15	15
C. Processing Cell		
1. Building	748	748
2. Installed equipment	507	507
3. Instrumentation	214	214
D. Intermediate-activity-level Processing Facilities		
1. Building	129	129
2. Installed equipment	28	28
3. Instrumentation	3	3
E. Low-activity-level Processing Facilities		
1. Building	75	86
2. Installed equipment	492	551
3. Instrumentation	151	181
4. Inert atmosphere glovebox and auxiliaries	-	44
F. Auxiliary Equipment		
1. Argon purification and cooling	75	85
2. Electrical substation and emergency generator	97	97
3. Spare equipment	349	349
4. Mobile equipment	35	35
Total	6,908	7,405

TABLE C.III. Estimated Investment Requirements for Building and Equipment for Volatility Processing of Oxide Fuel (80% plant factor)

	Investment Requirements in Thousands of Dollars		
	Annual Throughput: 22,700 kg/yr	Annual Throughput: 68,100 kg/yr	Annual Throughput: 227,000 kg/yr
A. General Facilities			
1. Offices, storerooms, and personnel convenience areas	156	254	396
2. Warehouse	40	58	97
3. Analytical laboratory	340	472	680
4. Maintenance facilities	241	308	530
5. Equipment disposal facilities	640	779	990
B. Remote-handling Cell			
1. Building	309	470	1,050
2. Transfer locks	101	101	202
3. Windows	150	150	400
4. Cranes and manipulators	122	122	286
5. Equipment - processing	188	262	617
6. Equipment - nonprocessing	68	79	129
7. Instrumentation	43	51	144
C. Processing Cell			
1. Building	140	269	620
2. Installed equipment	583	1,126	2,530
3. Installed instrumentation	152	306	719
D. Intermediate-activity-level Pro- cessing Facilities (UF ₆ Storage)			
1. Building	12	24	49
2. Equipment	38	92	210
3. Instrumentation	18	54	180
E. Low-activity-level Processing Facilities			
1. Building	16	37	150
2. Equipment	51	250	525
3. Instrumentation	58	140	305
F. Auxiliary Equipment			
1. Argon purification and cooling	55	66	97
2. Refrigeration	24	58	151
3. Electrical substation	80	116	193
4. Spare equipment	496	1,013	2,263
5. Mobile equipment	35	73	161
Total	4,150	6,730	13,704

TABLE C.IV. Estimated Investment Requirements for Building and Equipment
for Volatility Processing of Metal and Carbide Fuel
(80% plant factor)

	Investment Requirements in Thousands of Dollars	
	Carbide; Annual Throughput: 22,700 kg/yr	Metal; Annual Throughput: 22,700 kg/yr
A. General Facilities		
1. Offices, storerooms, and personnel convenience areas	156	156
2. Warehouse	40	40
3. Analytical laboratory	340	340
4. Maintenance facilities	241	241
5. Equipment disposal facilities	640	640
B. Remote-handling Cell		
1. Building	584	526
2. Transfer locks	68	68
3. Windows	150	150
4. Cranes and manipulators	122	122
5. Equipment - processing	224	186
6. Equipment - nonprocessing	101	101
7. Instrumentation	66	36
C. Processing Cell		
1. Building	107	85
2. Installed equipment	537	518
3. Installed instrumentation	141	141
D. Intermediate-activity-level Processing Facilities (UF₆ Storage)		
1. Building	151	127
2. Equipment	218	180
3. Instrumentation	70	40
E. Low-activity-level Processing Facilities		
1. Building	14	14
2. Equipment	129	129
3. Instrumentation	78	78
F. Auxiliary Equipment		
1. Argon purification and cooling	72	70
2. Refrigeration	24	24
3. Electrical substation	80	80
4. Spare equipment	474	456
5. Mobile equipment	35	35
Total	4,862	4,583

TABLE C.VI. Estimated Investment Requirements for Building and Equipment for Pyrochemical Processing of Oxide and Carbide Fuel (80% plant factor)

	Investment Requirements in Thousands of Dollars	
	Oxide; Annual Throughput: 22,700 kg/yr	Carbide; Annual Throughput: 22,700 kg/yr
A. General Facilities		
1. Offices, utility rooms, and personnel convenience areas	205	208
2. Warehouse	40	42
3. Analytical laboratory	370	370
4. Maintenance facilities	322	318
5. Reagent-preparation facilities	53	54
B. Processing Cell		
1. Building	2,373	2,425
2. Transfer locks	151	151
3. Windows	200	200
4. Cranes and manipulators	307	329
C. Processing Equipment and Instrumentation		
1. Processing equipment	700	768
2. Instrumentation	192	248
3. Weighing stations	10	10
4. Service lead-ins and disconnects	94	97
5. Welding stations	15	15
D. Auxiliary Equipment		
1. Argon cooling and purification	140	142
2. Off-gas handling equipment	46	47
3. Electrical substation and emergency generator	167	175
4. Mobile equipment	35	35
Total	5,420	5,634

TABLE C.V. Estimated Investment Requirements for Building and Equipment for Pyrochemical Processing of Metal Fuel (80% plant factor)

	Investment Requirements in Thousands of Dollars		
	Annual Throughput: 22,700 kg/yr	Annual Throughput: 68,100 kg/yr	Annual Throughput: 227,000 kg/yr
A. General Facilities			
1. Office, utility rooms, and personnel convenience areas	150	236	469
2. Warehouse	31	39	63
3. Analytical laboratory	370	515	738
4. Maintenance facilities	249	317	506
5. Reagent-preparation facilities	47	74	135
B. Processing Cell			
1. Building	1,398	1,817	4,065
2. Transfer locks	151	151	151
3. Windows	125	150	325
4. Cranes and manipulators	206	206	514
C. Processing Equipment and Instrumentation			
1. Processing equipment	452	705	1,604
2. Instrumentation	123	170	245
3. Weighing stations	10	10	23
4. Service lead-ins and disconnects	47	65	94
5. Welding stations	15	15	30
D. Auxiliary Equipment			
1. Argon cooling and purification	105	122	187
2. Off-gas handling equipment	39	73	151
3. Electrical substation and emergency generator	127	164	264
4. High-frequency power generation	27	52	76
5. Mobile equipment	35	73	161
Total	3,507	4,954	9,801

TABLE C.VII. Estimated Investment in Facilities for Disposal of Waste from Processing of Oxide Fuel from a 1000-MWe Fast Reactor

Item	Investment (Thousands of Dollars)		
	Aqueous	Volatility	Pyrochemical
Exhaust stack and service building	458	458	458
Service building and tanks for interim storage of noble gas effluent	25	25	25
Waste-neutralization facilities	64	38	-
Waste casks and transporters	27	27	27
Tanks and evaporators for miscellaneous active aqueous waste	-	335	335
Canal and building for interim storage of high-level activity solid wastes	-	483	393
Two 200,000-gal mild-steel storage tanks with reflux condenser; installed underground in concrete vaults	950	-	-
Service building and incinerator for disposal of waste solvent	59	-	-
Evaporators for aqueous waste	566	-	-
Spares for evaporators	150	-	-
Lagoon for evaporator overheads and/or other very low-activity wastes	117	30	30
Total	2,416	1,396	1,268

TABLE C.VIII. Estimated Operating Costs for Aqueous Processing
(in thousands of dollars)

	Oxide			Carbide	Metal
	1000 MWe, 22,700 kg/yr	3000 MWe, 68,100 kg/yr	10,000 MWe, 227,000 kg/yr	1000 MWe, 22,700 kg/yr	1000 MWe, 22,700 kg/yr
Processing					
Total labor	1,522	2,354	4,030	1,641	1,615
Direct materials	76	226	755	145	145
Total annual operating costs ^a	2,256	3,630	6,677	2,516	2,474
Waste Disposal					
Total labor	238	484	1,260	238	238
Direct materials	8	22	75	8	8
Total annual operating costs ^{a,b}	373	797	2,131	373	373

^aCalculated from the labor and direct-material costs, according to the ground rules of this study.^bAll costs associated with the tank replacement and perpetual care of stored liquid wastes are included in these operating costs.TABLE C.IX. Estimated Operating Costs for Pyrochemical Processing
(in thousands of dollars)

	Metal			Oxide	Carbide
	1000 MWe, 22,700 kg/yr	3000 MWe, 68,100 kg/yr	10,000 MWe, 227,000 kg/yr	1000 MWe, 22,700 kg/yr	1000 MWe, 22,700 kg/yr
Processing					
Total labor	696	1,232	2,931	1,042	1,065
Direct materials	164	493	1,643	276	276
Total annual operating costs ^a	1,188	2,324	6,046	1,811	1,846
Waste Disposal					
Total labor	62	188	625	62	62
Direct materials	14	42	140	60	60
Total annual operating costs ^a	186	554	1,842	352	352

^aCalculated from the labor and direct material costs according to the ground rules of this study.TABLE C.X. Estimated Operating Costs for Volatility Processing
(in thousands of dollars)

	Oxide			Carbide	Metal
	1000 MWe, 22,700 kg/yr	3000 MWe, 68,100 kg/yr	10,000 MWe, 227,100 kg/yr	1000 MWe, 22,700 kg/yr	1000 MWe, 22,700 kg/yr
Processing					
Total labor	747	1,377	2,395	868	840
Direct materials	144	432	1,440	292	218
Total annual operating costs ^a	1,281	2,548	5,300	1,630	1,499
Waste Disposal					
Total labor	62	188	625	62	62
Direct materials	48	144	480	48	48
Total annual operating costs ^a	318	950	3,161	318	318

^aCalculated from labor and direct materials cost according to the ground rules of this study.

APPENDIX D

Unit-cost Details

Tables D.I through D.VII presents the cost estimates developed in this study as unit costs in dollars per kilogram of uranium and plutonium in the fuel before irradiation. These unit costs are displayed under columnar headings of processing-plant throughput in kilograms per year. The annual throughputs are also in terms of the kilograms of uranium and plutonium before irradiation.

TABLE D.I. Unit-cost Details: Investment in Buildings and Structures

	Unit Cost, ^a \$/kg		
	Annual Throughput: 22,700 kg	Annual Throughput: 68,100 kg	Annual Throughput: 227,000 kg
<u>Processing</u>			
Aqueous, Metal	136	74	40
Aqueous, Oxide	124	67	36
Aqueous, Carbide	146	79	43
Volatility, Metal	81	42	27
Volatility, Oxide	73	38	24
Volatility, Carbide	86	44	29
Pyrochemical, Metal	72	31	20
Pyrochemical, Oxide	115	50	31
Pyrochemical, Carbide	116	50	32
<u>Waste Disposal</u>			
Aqueous, Metal	43 ^b + 41 ^c	29 ^b + 41 ^c	21 ^b + 41 ^c
Aqueous, Oxide	43 ^b + 41 ^c	29 ^b + 41 ^c	21 ^b + 41 ^c
Aqueous, Carbide	43 ^b + 41 ^c	29 ^b + 41 ^c	21 ^b + 41 ^c
Volatility, Metal	49	40	36
Volatility, Oxide	49	40	36
Volatility, Carbide	49	40	36
Pyrochemical, Metal	44	36	32
Pyrochemical, Oxide	46	37	33
Pyrochemical, Carbide	46	37	33

^aThe total investment amount is expressed as a cost per unit of annual throughput of actinides and fission products.

^bInvestment amounts are for items provided at time of construction, not including original waste-storage tanks. Tanks installed after startup, perpetual-care charges, and charges for other items required in the future are treated as annual expenses.

^cThis item is for the investment in initial waste-storage tanks at time of startup.

TABLE D.II. Unit-cost Details: Investment in
In-cell Processing Equipment

	Unit Cost, ^a \$/kg		
	Annual Throughput: 22,700 kg	Annual Throughput: 68,100 kg	Annual Throughput: 227,000 kg
<u>Repairable Equipment</u>			
Aqueous, Metal	28	17	12
Aqueous, Oxide	23	15	11
Aqueous, Carbide	31	19	15
Volatility, Metal	28	12	9.2
Volatility, Oxide	13	7	5.4
Volatility, Carbide	29	15	12
Pyrochemical, Metal	21	11	7.2
Pyrochemical, Oxide	32	16	11
Pyrochemical, Carbide	34	18	12
<u>Replaceable But Nonrepairable^b</u>			
Aqueous, Metal	6.9	4.6	2.9
Aqueous, Oxide	6.9	4.6	2.9
Aqueous, Carbide	6.9	4.6	2.9
Volatility, Metal	19	12	7.5
Volatility, Oxide	21	14	9.1
Volatility, Carbide	19	13	7.8
Pyrochemical, Metal	0	0	0
Pyrochemical, Oxide	0	0	0
Pyrochemical, Carbide	0	0	0

^aThe total investment amount is expressed as a cost per unit of annual throughput of actinides and fission products.

^bProcessing equipment subject to very high levels of radioactivity was considered not able to be decontaminated sufficiently for repair. Pyrochemical equipment, however, can be repaired in its remote-operation cell without decontamination.

TABLE D.III. Unit-cost Details: Investment in
Nonprocessing and Out-of-cell Processing Equipment

	Unit Cost, ^a \$/kg		
	Annual Throughput: 22,700 kg	Annual Throughput: 68,100 kg	Annual Throughput: 227,000 kg
<u>Processing</u>			
Aqueous, Metal	133	67	36
Aqueous, Oxide	107	54	29
Aqueous, Carbide	142	71	38
Volatility, Metal	77	39	23
Volatility, Oxide	71	36	22
Volatility, Carbide	79	39	24
Pyrochemical, Metal	70	31	16
Pyrochemical, Oxide	93	41	21
Pyrochemical, Carbide	98	43	23

^aThe total investment amount is expressed as a cost per unit of annual throughput of actinides and fission products.

TABLE D.IV. Unit-cost Details: Annual Total Labor Cost

	Unit Cost, ^a \$/kg		
	Annual Throughput: 22,700 kg	Annual Throughput: 68,100 kg	Annual Throughput: 227,000 kg
<u>Processing</u>			
Aqueous, Metal	71	37	19
Aqueous, Oxide	67	35	18
Aqueous, Carbide	72	32	19
Volatility, Metal	37	23	12
Volatility, Oxide	33	20	11
Volatility, Carbide	38	23	12
Pyrochemical, Metal	31	18	13
Pyrochemical, Oxide	46	27	19
Pyrochemical, Carbide	47	28	20
<u>Waste Disposal</u>			
Aqueous, Metal	10	7.1	5.6
Aqueous, Oxide	10	7.1	5.6
Aqueous, Carbide	10	7.1	5.6
Volatility, Metal	2.8	2.8	2.8
Volatility, Oxide	2.8	2.8	2.8
Volatility, Carbide	2.8	2.8	2.8
Pyrochemical, Metal	2.8	2.8	2.8
Pyrochemical, Oxide	2.8	2.8	2.8
Pyrochemical, Carbide	2.8	2.8	2.8

^aThe annual total labor cost is expressed as a cost per unit of annual throughput of actinides and fission products.

TABLE D.V. Unit-cost Details: Annual Direct-materials Cost

	Unit Cost, ^a \$/kg		
	Annual Throughput: 22,700 kg	Annual Throughput: 68,100 kg	Annual Throughput: 227,000 kg
<u>Processing</u>			
Aqueous, Metal	6.4	6.4	6.4
Aqueous, Oxide	3.3	3.3	3.3
Aqueous, Carbide	6.4	6.4	6.4
Volatility, Metal	9.6	9.6	9.6
Volatility, Oxide	6.3	6.3	6.3
Volatility, Carbide	13	13	13
Pyrochemical, Metal	7.2	7.2	7.2
Pyrochemical, Oxide	12	12	12
Pyrochemical, Carbide	12	12	12
<u>Waste Disposal</u>			
Aqueous, Metal	0.33	0.33	0.33
Aqueous, Oxide	0.33	0.33	0.33
Aqueous, Carbide	0.33	0.33	0.33
Volatility, Metal	2.1	2.1	2.1
Volatility, Oxide	2.1	2.1	2.1
Volatility, Carbide	2.1	2.1	2.1
Pyrochemical, Metal	0.61	0.61	0.61
Pyrochemical, Oxide	2.6	2.6	2.6
Pyrochemical, Carbide	2.6	2.6	2.6

^aThe annual direct-materials cost is expressed as a cost per unit of annual throughput of actinides and fission products.

TABLE D.VI. Unit-cost Details: Investment in Waste-disposal Equipment

	Unit Cost, ^a \$/kg		
	Annual Throughput: 22,700 kg	Annual Throughput: 68,100 kg	Annual Throughput: 227,000 kg
Aqueous, Metal	25	17	12
Aqueous, Oxide	25	17	12
Aqueous, Carbide	25	17	12
Volatility, Metal	14	12	11
Volatility, Oxide	14	12	11
Volatility, Carbide	14	12	11
Pyrochemical, Metal	10	8.0	6.9
Pyrochemical, Oxide	11	8.9	7.7
Pyrochemical, Carbide	11	8.9	7.7

^aThe investment amount is expressed as a cost per unit of annual throughput of actinides and fission products.

TABLE D.VII. Unit-Cost Details: Total Annual Charges for Processing and Waste Disposal^a

	Unit Cost, ^b \$/kg		
	Annual Throughput: 22,700 kg	Annual Throughput: 68,100 kg	Annual Throughput: 227,000 kg
<u>Processing Charge^b</u>			
Aqueous, Metal	184	101	58
Aqueous, Oxide	163	88	49
Aqueous, Carbide	190	99	59
Volatility, Metal	121	75	49
Volatility, Oxide	107	65	42
Volatility, Carbide	131	83	55
Pyrochemical, Metal	91	52	38
Pyrochemical, Oxide	136	79	58
Pyrochemical, Carbide	140	81	59
<u>Waste Disposal Charge^b</u>			
Aqueous, Metal	47	38	32
Aqueous, Oxide	47	38	32
Aqueous, Carbide	47	38	32
Volatility, Metal	27	25	23
Volatility, Oxide	27	25	23
Volatility, Carbide	27	25	23
Pyrochemical, Metal	19	17	16
Pyrochemical, Oxide	27	24	23
Pyrochemical, Carbide	27	24	23

^aTotal charges are calculated by the ground rules of this study.

^bAnnual charges are expressed as a cost per unit of annual throughput of actinides and fission products.

Unit costs incurred in processing and waste disposal are presented for total labor (the sum of direct and indirect labor), direct materials, investment required in building and equipment, and the charges developed under the economic ground rules of this study for processing and waste disposal. The product of processing is a feed material for refabrication which can be utilized without further chemical treatment. The waste-disposal charge includes the cost of ultimate disposal in perpetual-care facilities. By application of the economic ground rules and assumptions of this study to the unit costs, the charges appearing in the tables can be developed. The presentation of the basic unit-cost information facilitates the application of alternative economic ground rules and assumptions.

The scope of this report did not include the economic effect of burnup. However, over a limited range of about 7 to 13%, it should be possible to obtain meaningful comparisons of processing and waste-disposal charges. The following is suggested as estimation procedures:

1. To compute the charge for processing, consider the throughput to be the only significant variable and ignore burnup.
2. To compute the waste-disposal charge in aqueous processing, employ the average of the annual charges based on total throughput and on fission-product throughput. (Fission-product throughput is the product of total throughput and burnup.)
3. To compute the waste-disposal charge in pyrochemical processing of metal fuel and in the volatility processing of all fuels, employ the unit charge based on the throughput of fission products.
4. To compute the annual charge for waste disposal in the pyrochemical processing of oxide and carbide fuels, consider the cost of storage of fission products, which is largely a function of the heat-generation rate, and the cost of storage of magnesium oxide formed in the reduction of ceramic fuel. The sum of the following provides a reasonable approximation of the annual charge:
 - a. The annual charge based on the throughput of fission products computed from the metal-fuel estimates.
 - b. The difference in the annual charges for ceramic and metal fuels based on total throughput.

APPENDIX E

Shipping and Inventory Costs for Central Processing Plants1. General Discussion

That shipment of material as hazardous as high-burnup fuel from a fast reactor will be practical is an implicit assumption of this analysis; this assumption may prove to be false in the future. A single incident might result in the passage of legislation that makes spent-fuel shipping prohibitively expensive. In that case, on-site fuel processing would be a necessity.

Shipping and inventory costs are analyzed together in this study because of the interrelationship between the two costs. The value of the fuel affects both the inventory cost and property-insurance cost during shipping. The shipping costs affect the value of the fuel before shipping and, thus, the inventory cost before shipping. Finally, for pyrochemical and volatility processing, the combined cost of shipping plus inventory has a minimum value for an optimum cooling period, which is the time at which benefits of further reduction in fission-product cost are balanced by increasing inventory charges. This optimum cooling period varies with burnup of the fuel. Other variables, including plutonium concentration in the discharged fuel and the cost of plutonium, have only minor effects.

Various estimates are available in the literature for the cost of shipping thermal-reactor fuel ranging from \$1.00/kg to more than \$20.00/kg, depending primarily on the distance shipped and the shielding required. For example, the cost of shipping 7600 lb of uranium in a 70-ton cask for 3000 miles was estimated at \$6.40/kg to \$10.90/kg in "Costs of Nuclear Power."¹ The same source stated that the main uncertainties in the transportation costs for thermal fuels are the insurance rates, the choice of cask size and weight, cooling time before shipment, and effect of negotiations on carrier rates.

Shipping costs for fast-reactor fuels cannot be conveniently expressed in terms of cost per kilogram of material shipped because the high rate of heat dissipation of the fuel makes it impossible to utilize the full volume or weight capacity of casks designed for thermal reactor fuel. The cask-cooling requirement is more limiting in determining the cask capacity than the volume or weight of fuel. The shipping cost, therefore, is more directly a function of the amount of heat that must be dissipated than of the weight of fuel shipped.

In this study of fuel shipping costs, the cost estimates were made for rail shipments of rented casks for delivering spent fuel to the reprocessing plant and separate shipments for delivering new fuel to the reactor.

An empty cask would be shipped to the reactor, loaded with spent fuel, and shipped back to the reprocessing plant. New fuel (or refabricated fuel) would be shipped from the fuel-fabricating plant, which was assumed to be on the site of the reprocessing plant, to the reactor, and the empty container would be shipped back to the reprocessing site.

Alternate shipping schemes were found to be more expensive than the one adopted. If new fuel is delivered to the reactor in the cask that is sent to collect the spent fuel, the new fuel arrives long before it is needed, and the inventory charge is greater than the cost of a special shipment of new fuel. Another alternative, that of retaining at the reactor the refabricated-fuel shipping cask until spent fuel is discharged, is uneconomical because the cask rental charge is too high.

The annual cost of shipping spent fuel was determined from the estimated cost of a single trip and the number of trips required per year. The annual cost of shipping refabricated fuel to the reactor was obtained by similar means. The estimated annual property-insurance cost, which varies with the length of the trip, and the annual value of the fuel shipped were added to the annual cost of shipments. To determine the approximate effect of distance on the overall cost of shipping, distances of 500 and 1000 miles were considered.

The main hazards in shipping spent-fuel elements are: 1) a criticality incident, 2) dispersion of radioactive materials, 3) direct gamma radiation from the cask, and 4) damage to or loss of the fuel. The cask (together with the surrounding cask shield if one is provided) must be constructed to prevent the above incidents from occurring, not only during normal conditions of shipping, but also during accidents that might subject the cask to damage by impact against a massive object or a sharp projection, to fire, to immersion in water, or to loss of power for coolant circulation.² Shipment of spent fast-reactor fuel is usually more hazardous than shipment of thermal-reactor fuels because of high plutonium content and high heat-dissipation rates. For these reasons, unusual care must be taken to guard against a criticality incident in case of fuel meltdown or flooding of the cask with water. The basket containing the fuel subassemblies must have thick dividers containing neutron poisons to separate the subassemblies. Little appears to be gained from the use of a forced-circulation coolant since provision must be made to dissipate the heat in case of loss of coolant flow if forced coolant circulation is utilized.² For this reason many recent cask designs do not call for forced coolant circulation.³⁻⁶

The characteristics of the cask for shipping spent reactor fuel for this study are shown in Part A of Table E.I. These characteristics conform in many respects to the specifications that would be calculated by the computer code, designated MYRA, developed at Oak Ridge National Laboratory.³

The fuel subassemblies in such a cask would radiate heat to the dividers separating the fuel elements, and the dividers would conduct the heat to the cask wall. No coolant other than naturally circulating air or other gas would be utilized. The cask would be heavier than the maximum cask size used in formulating the MYRA code, 100 versus 70 tons, because the larger size is believed feasible for loading and transporting a short time in the future. A shock buffer or shield, as provided in some recent designs,^{7,8} is also included to improve safety during shipping. Cooling air would be forced through the shock buffer to cool the finned cask under normal conditions. In case of power failure, enough airflow would be provided by natural convection to limit the temperature rise of the fuel to 100°F above the normal shipping temperature. The combined shipping weight of the cask and buffer is 125 tons. This weight is within the capability of special flatcars, which would transport the burden at the cost-per-ton rates that would apply for casks weighing 20 to 70 tons, which are shipped on standard flatcars.

TABLE E.I. Cost Basis of Fuel-shipping Cask for Spent Fuel

<u>A. Cask Characteristics</u>	
Weight, loaded, tons	100
Weight of cask shock buffer, tons	25
Capacity	
Full loading, actinides plus fission products, kg	2,000
Heat dissipation, kW	50
 <u>B. Interest, Depreciation, and Insurance</u>	
	<u>% of Cask Cost</u>
Return on investment	6.75
Sinking-fund factor (10 years at 6.75%)	7.33
Income tax	3.40
State and local taxes	2.45
Property insurance on cask	1.00
	<u>20.93</u>
 <u>C. Rental Cost</u>	
Cost of cask and buffer,	
total \$250,000 annual	\$52,000
Annual cost of repairs and supplies	3,000
Total annual cost	\$55,000
Rental cost per day, 60% use factor	250

The maximum weight-carrying capacity of casks for this service was estimated to be about 2000 kg of actinides and fission products, the equivalent of about 15 six-inch hexagonal fuel subassemblies. In most cases, the heat-dissipation capacity of the cask would limit the weight of

fuel shipped to much less than 2000 kg of actinides and fission products. For fuel cooled for long periods, however, the bulk of fuel that would dissipate 50 kW is too great for provision of radiation shielding and neutronic isolation of the subassemblies within the cask weight limitation of 100 tons.

It is assumed that spent-fuel casks would be designed to suit the fuel to be shipped. Therefore, to specify the same heat-dissipating capacity and the same weight for the many different casks that would be designed to ship the entire spectrum of spent fuels in this study, is a simplification of a complex problem, which introduces error into the results. But the error is not as serious as it would first appear because the shielding thickness and the external heat-transfer area required of the cask for fuel developing 50 kW of heat would not vary greatly with the type of fuel or its bulk within the fuel-volume restriction corresponding to the cask load limit of 2000 kg of actinides and fission products. Consequently, the weight of the representative cask of Table E.I is approximately correct for any cask designed to transport spent fast-reactor fuel dissipating 50 kW of heat.

For shipping refabricated fuel from the fabrication site to the reactor site, heat-dissipation and shielding requirements are not significant problems. However, in case of a transportation accident, the high plutonium content of the refabricated fuel constitutes a hazard, with respect to criticality incidents and to dispersion of alpha-active materials, that is as severe as the hazard for spent fuel. As a result, the container for shipping refabricated fuel will be a substantial structure even though it does not contain significant amounts of shielding. It has been estimated for this study that a shipment of fuel containing three tons of uranium and plutonium, when structurally protected, neutronically shielded, and fireproofed, would weigh 30 tons.

2. Cask Rental Costs

The cost of the spent-fuel shipping cask and shock buffer was estimated at \$1.00/lb or \$250,000 for the entire 100-ton cask and 25-ton buffer assembly, a cost in agreement with other recent estimates.^{3,4,9} The interest and depreciation on the cask, which are presented in Table E.I, were calculated for a 10-yr life and at rates applicable to privately owned utilities,¹⁰ rather than at rates for privately owned, chemical-processing companies or municipally owned utilities. It is assumed that the casks would be rented from a company specializing in cask rentals, which might be, for example, a subsidiary owned by one or more utility companies. Since the rental company would need spare casks to meet peak demands, and casks would be out of service during repair or maintenance, a use factor of 60% was applied in calculating the daily rental cost of \$250.00/day for the spent-fuel shipping cask. By the same ground rules, the rental cost of the 30-ton refabricated-fuel shipping cask was calculated to be \$60.00/day.

3. Cost of Round Trip for Shipping Fuel

Several casks would be required to ship spent fuel from one reactor unloading. It was assumed that at least three casks would be shipped on one train under the care of a crew of escorts.

The estimated costs for round-trip shipments of the spent-fuel cask and the refabricated-fuel cask (each loaded one way only) for distances of 500 and 1000 miles each way are presented in Table E.II. The estimates of the trip durations (which were applied to calculate the cask rental cost and the cost of escorts) were obtained from Argonne experience for the actual shipping time for MTR fuel elements between Scoville, Idaho, and Argonne (5 to 6 days each way for 1500 miles) with an allowance for loading time. The freight costs are based on current rates for shipment of casks weighing 40,000 lb or more from Chicago or Argonne to Kansas City, Missouri (~460 miles, \$39.80/ton), to Laramie, Wyoming (~1030 miles, \$39.80/ton), and to Scoville, Idaho (~1460 miles, \$58.60/ton). The cost of labor for loading and unloading the casks was assumed to be \$600 per trip. Whether

TABLE E.II. Cost of Single Trips for Shipping Spent Fuel to Central Processing Plant and Refabricated Fuel to Reactor

(Costs do not include property insurance on the fuel during shipping.)

	Spent-fuel Cask		Refabricated-fuel Cask	
	500	1,000	500	1,000
One-way Distance, miles	500	1,000	500	1,000
Round-trip Duration, days	10	15	10	15
Total Cask Weight, tons	125	125	30	30
Rental of Cask, \$/trip	2,500	3,750	600	900
Freight Cost (round trip):				
@ \$50/ton for 500-mile distance	6,250		500	
@ \$80/ton for 1000-mile distance		10,000		2,400
Total Labor, Including:				
Escort during shipping ^a	1,300	1,650	1,300	1,650
Liability insurance ^b	1,500	2,000	750	1,000
Administrative overhead	1,000	1,000	500	500
Total Shipping cost (less property insurance)	\$12,550	\$18,400	\$4,650	\$6,450

^aCost includes train fare in caboose.

^bCalculated at rate of \$1,000 + \$1.00/mile for spent fuel, and \$500 + \$0.50/mile for refabricated fuel.

escorts will be required in the future is debatable. Since several casks will be shipped on the same train, the cost of escorts is inexpensive and may result in lower insurance rates.

Insurance rates for shipment of fast-reactor fuel are difficult to assess at present because there is little experience on which to base actuarial data. Eventually, insurance rates will probably be based on accurate assessments of the risks involved, but at present, rates do not distinguish between shipping distances, heat-dissipation rates, or the fissile content of the fuel. In this study, liability insurance was estimated to cost \$1,000/trip plus \$1.00/mile for spent fuel and \$500/trip plus \$0.50/mile for refabricated fuel. Property insurance, which is a function of the value of the material shipped as well as the risk of accident, is discussed below with the value of the fuel and the inventory costs.

In this study an administrative cost of \$1,000/trip was allowed for spent-fuel shipments and \$500/trip for refabricated-fuel shipments. Thus, with round-trip shipment of 25 or 30 spent-fuel casks per year, enough would be allowed for administrative overhead to pay for a full-time shipping administrator, secretarial help, and the required office space and equipment. For fewer shipments, the part-time services of a supervisor would be sufficient.

The number of spent-fuel cask shipments per year is determined by the cask capacity limitations provided by the maximum heat-dissipation rate of 50 kW and the maximum fuel loading of 2,000 kg of actinides plus fission products. The rate of heat dissipation from the spent fuel was calculated from the data of Burris and Dillon.¹¹ The heat dissipations were calculated for a range of fuel burnups and cooling times for a reference zone loading of a hypothetical 1,000-MWe fast breeder reactor; these results are summarized in Table E.III. In the present study of comparative fuel costs, only

TABLE E.III. Heat Dissipation from Core and Blanket Discharged from 1,000-MWe Reactor

Conditions: 2,500-MWt capacity, 80% reactor load factor, 85% of power developed in core, 12,000 kg of actinides in core charge, 50,000 kg of actinides in blanket charge.

Burnup, %	Processing Rate, kg/yr	Irradiation Time, days	Heat-dissipation Rate ^a				
			20 days	40 days	80 days	100 days	180 days
<u>Core</u>							
3	21,770	201	0.197	0.126	0.0834	0.0705	0.0417
5	13,060	335	0.218	0.145	0.0995	0.0852	0.0530
10	6,530	670	0.238	0.165	0.118	0.104	0.0672
20	3,260	1,340	0.254	0.183	0.135	0.119	0.0783
<u>Blanket</u>							
0.36 (1.0) ^b	32,350	564	0.00988	0.00681	0.00480	0.00419	0.00270
0.71 (2.0)	16,180	1,130	0.0106	0.00756	0.00553	0.00488	0.00329
1.07 (3.0)	10,780	1,690	0.0109	0.00796	0.00592	0.00523	0.00362
1.42 (4.0)	8,090	2,260	0.0112	0.00824	0.00618	0.00552	0.00386
1.78 (5.0)	6,470	2,820	0.0113	0.00843	0.00638	0.00573	0.00403

^aHeat-dissipation rate is expressed as kW/kg of actinides and fission products.

^bPlutonium concentration at discharge is given in percent within the parentheses.

the 10% core burnup and the 0.71% blanket burnup (2.0% plutonium) specifications were used. The minimum number of required trips per year was determined from the fuel-processing rate, the heat dissipation (Table E.III), and the cask-capacity specifications (Table E.I). Core and blanket fuel were mixed in a cask to achieve this minimum number of shipments.

The charges for the out-of-reactor fuel inventory is determined by the out-of-reactor fuel-holdup times, the value of the fuel in the several stages of fuel handling, and the fixed-charge rate. The holdup times for the central processing plants* were based on the schedule for fuel handling outlined in Table E.IV. For nonaqueous processing, cooling times were chosen to minimize the total costs of inventory and shipping.

TABLE E.IV. Residence Times in Out-of-reactor Fuel-handling Operations

Operation	On-site Processing, days				Central-plant Processing, days			
	Aqueous		Volatility and Pyrochemical		Aqueous		Volatility and Pyrochemical	
	Min	Max	Min	Max	Min ^a	Max ^b	Min ^a	Max ^b
Cooling	120 ^c	280 ^c	20 ^c	100 ^c	100	100	35 ^d	65 ^d
Shipping to Processing	0	0	0	0	5	8	5	8
Storage at Processing Plant	0	0	0	0	15	15	15	15
Processing	20	20	20	20	15	15	10	10
Storage at Fabricating Plant	10	10	10	10	10	10	10	10
Fabrication	20	20	20	20	10	10	10	10
Shipping to Reactor	0	0	0	0	5	7	5	7
Storage at Reactor	190	30	110	30	30	30	30	30
	360 ^e	360 ^e	180 ^f	180 ^f	190 ^g	195 ^g	120 ^h	155 ^h

^a500-mile shipping distance.

^b1000-mile shipping distance.

^cThe end of the minimum cooling period coincides with the beginning of processing. At the end of the maximum cooling period, all fuel in the batch has been started through processing.

^dThe optimum cooling time varies with fuel composition, reactor conditions, and shipping costs and is presented in Table E.VI below.

^eTwo reactor-unloading cycles.

^fOne reactor-unloading cycle.

^gReturn to reactor other than one of origin is assumed; otherwise value is 360 days (see Note e).

^hReturn to reactor other than one of origin is assumed; otherwise value is 180 days (see Note f).

The basic fuel value was assumed to be \$10/g for fissionable plutonium and \$8/kg for other actinides. This basic fuel value was taken to apply to fuel material ready for fabrication, and adjustments in fuel value were required for fuel in other stages of the fuel-cycle operations. These adjustments depend on costs incurred in reprocessing, shipping, insurance, and refabrication, together with costs due to losses. The fixed-charge rate for fuel inventory was 13%/yr, the same as used throughout this study of comparative costs. Property insurance during shipping was estimated at 0.15% of the basic fuel value for the 500-mile distance and 0.20% of the basic fuel value for the 1000-mile distance.

*Holdup times and inventory charges for the close-coupled plants (on-site processing) used the inventory model described in Appendix H.

Liability insurance (Table E.II was estimated at \$1000/trip, plus \$1/mile for spent fuel, and \$500/trip, plus \$0.50/mile for refabricated fuel.

Shipping and inventory costs for central aqueous plants are summarized in Table E.V and for central nonaqueous plants in Table E.VI.

TABLE E.V. Shipping and Inventory Costs for Central Aqueous-processing Plants
(1,000 MWe, 80% load factor, fissile plutonium valued at \$10/g)

% Pu (total) ^a		% Burnup	Processing Rate per Reactor, kg/yr	Costs for 500-mile Shipping, mills/kWh		Costs for 1,000-mile Shipping, mills/kWh	
Core	Blanket			Shipping	Inventory	Shipping	Inventory
15	2	3	37,950	0.077	0.22	0.11	0.23
		5	29,220	0.057	0.15	0.082	0.15
		10	22,710	0.038	0.086	0.054	0.088
		20	19,440	0.025	0.055	0.036	0.057
	4	3	29,860	0.073	0.23	0.11	0.23
		5	21,150	0.052	0.15	0.076	0.15
		10	14,620	0.033	0.085	0.048	0.087
		20	11,350	0.021	0.054	0.030	0.056
20	2	3	37,950	0.078	0.30	0.11	0.30
		5	29,220	0.058	0.19	0.084	0.19
		10	22,710	0.038	0.11	0.050	0.11
		20	19,440	0.025	0.066	0.037	0.067
	4	3	29,860	0.075	0.30	0.11	0.30
		5	21,150	0.054	0.20	0.078	0.19
		10	14,620	0.034	0.11	0.049	0.11
		20	11,350	0.021	0.065	0.031	0.067
30	2	3	37,950	0.081	0.43	0.12	0.44
		5	29,220	0.059	0.27	0.086	0.28
		10	22,710	0.038	0.15	0.056	0.15
		20	19,440	0.026	0.086	0.037	0.088
	4	3	29,860	0.078	0.43	0.11	0.44
		5	21,150	0.055	0.27	0.080	0.28
		10	14,620	0.036	0.15	0.050	0.15
		20	11,350	0.022	0.085	0.031	0.088

^aThe values listed are for all plutonium isotopes in the fuels, which contain 65 w/o fissile isotopes in discharged core plutonium and 95 w/o fissile isotopes in discharged blanket plutonium.

TABLE E.VI. Shipping and Inventory Costs for Central Pyrometallurgical and Fluoride Volatility Processing Plants
(1,000-MWe, 80% load factor, fissile plutonium valued at \$10/g)

% Pu (total) ^a			Processing Rate, kg/yr	500-mile Shipping			1,000-mile Shipping		
Core	Blanket	% Burnup		Optimum Cooling Time, days	Cost, mills/kWh		Optimum Cooling Time, days	Cost, mills/kWh	
					Shipping	Inventory		Shipping	Inventory
15	2	3	37,950	45	0.11	0.16	65	0.14	0.19
		5	29,220	45	0.082	0.10	65	0.10	0.12
		10	22,710	45	0.050	0.060	60	0.065	0.068
		20	19,440	40	0.034	0.038	55	0.045	0.042
	4	3	29,860	45	0.11	0.16	65	0.14	0.19
		5	21,150	45	0.077	0.10	65	0.063	0.12
		10	14,620	40	0.048	0.057	60	0.060	0.068
		20	11,350	35	0.031	0.036	55	0.038	0.043
20	2	3	37,950	35	0.13	0.18	50	0.16	0.22
		5	29,220	35	0.092	0.12	50	0.12	0.14
		10	22,710	35	0.057	0.069	50	0.072	0.080
		20	19,440	35	0.036	0.043	45	0.045	0.048
	4	3	29,860	35	0.13	0.19	50	0.15	0.22
		5	21,150	35	0.087	0.12	50	0.11	0.14
		10	14,620	35	0.052	0.068	50	0.065	0.079
		20	11,350	35	0.031	0.043	45	0.041	0.048
30	2	3	37,950	30	0.15	0.26	35	0.20	0.29
		5	29,220	30	0.10	0.17	35	0.14	0.18
		10	22,710	30	0.062	0.091	35	0.084	0.099
		20	19,440	30	0.041	0.054	35	0.053	0.058
	4	3	29,860	30	0.14	0.26	35	0.19	0.29
		5	21,150	30	0.096	0.16	35	0.13	0.18
		10	14,620	30	0.056	0.090	35	0.076	0.098
		20	11,350	30	0.035	0.054	35	0.046	0.058

^aThe values listed are for all plutonium isotopes in the fuels, which contain 65 w/o fissile isotopes in discharged core plutonium, and 95 w/o fissile isotopes in discharged blanket plutonium.

For comparison, the values are given for 500- and 1000-mile shipping distances and for a range of core and blanket burnups. However, the reference case for this comparative study of processing costs is the one for 20% total plutonium in the core, 2% blanket burnup, 10% core burnup, and a 1000-mile shipping distance. It is assumed that the type of fuel (oxide, metal, or carbide) is a matter of indifference as to inventory and shipping costs.

References for Appendix E

1. *Costs of Nuclear Power*, TID-8531, (1961).
2. *Regulation to Protect Against Radiation in the Shipment of Irradiated Fuel Elements*, U. S. Code of Federal Regulations, Title 10, Part 72 (Proposed).
3. R. Salmon, *A Computer Code (CDC 1604A or IBM 7090) for Calculating the Cost of Shipping Spent Reactor Fuels as a Function of Burnup, Specific Power, Cooling Time, Fuel Composition, and Other Variables*, ORNL-3648 (1964).
4. Lead Industries Association, Inc., *Radioactive Fuel - A Packaging Problem*, Lead, 27(3):2-3 (1963).
5. E. C. Lusk, H. M. Epstein, R. J. Eiber, and J. E. Gates, *Containers for the Shipment of Spent Reactor Fuel Elements*, Trans. Am. Nucl. Soc. 5, pp. 361-362 (1962).
6. E. C. Lusk and J. E. Gates, *New Developments in the Design of Shipping Casks for Irradiated Fuel Elements*, Nuclear News 6(6):11-14 (1963).
7. D. W. McLenegan, C. W. Smith, E. A. Ripperger, and J. E. Breen, *Applications of External Energy Absorbers to Large Fission Product Shipping Casks*, Summary Report of AEC Symposium on Packaging and Regulatory Standards for Shipping Radioactive Material, TID-7651, pp. 265-302 (1962).
8. C. W. Smith, *Development and Design of Off-Site Fission Product Shipping Casks at Hanford*, HW-8A-2295 (1962).
9. L. B. Shappert, *An Economic Analysis of the Domestic and Overseas Shipment of Spent Reactor Fuel*, ORNL-TM-590 (1963).
10. Kaiser Engineers, *Guide to Nuclear Power Cost Evaluation*, Vol. 5, TID-7025 (1962).
11. L. Burris, Jr., and I. G. Dillon, *Estimation of Fission Product Spectra in Discharged Fuel from Fast Reactors*, ANL-5742 (July 1957).

APPENDIX F

Radiations in Recycle Core Fuel, An Outline of the Problem

The nuclides produced by long neutron irradiation of uranium and plutonium have very complex decay chains. These nuclides emit various amounts of alpha, beta, and gamma radiation in the course of their decay. Also, fast neutrons are produced by those nuclides undergoing spontaneous fission and by those nuclides undergoing alpha decay, whereby energetic alpha radiations interact with light elements to emit neutrons. The radiation hazard to operating personnel varies greatly, depending on the concentration of the nuclides that may be present. For example, the radiations of unirradiated natural uranium are such as to present a minimal hazard, which can be dealt with by conventional use of protective clothing and ventilated equipment spaces. Plutonium-239 has only a small amount of hard radiation, but its alpha emission is the source of a very high toxicity if ingested. It is customary to conduct operations on ^{239}Pu in gloveboxes, whereby complete physical containment is provided in an enclosure with glass windows and direct handling is allowed with gloves of rubber-like materials. While a short thermal irradiation of, say, 1/10% burnup of natural uranium generates plutonium that is nearly pure ^{239}Pu , longer irradiation and higher burnups produce substantial amounts of other isotopes. With increased concentration of the other isotopes, the radiation level increases. It is common to supplement gloveboxes with local shielding around the equipment, with extra thicknesses of lead glass on the windows, and with gloves containing extra layers of leaded cloth for shielding the hands. Finally, for substances emitting substantial amounts of hard radiation, massive shielding must be interposed between the equipment and the human operator. Under these conditions, glovebox designs are not practical and remote-control techniques must be used.

To evaluate the shielding requirement for the fabrication of recycled fast-reactor fuel, the following reference conditions must be established:

1. The radiations emitted per unit of recycled fuel.
2. The quantity and spatial distribution of fuel in the operating area of the plant.

Experimental data are not available on the radiation emitted from fuel exposed in a fast reactor to the specifications adopted in this study, nor can the radiation be predicted accurately because of the lack of basic cross-section data of the nuclides in question.

Among the types of radiation and their sources that are likely to be of significance are the following:

1. Neutrons from spontaneous fission of ^{238}Pu , ^{240}Pu , and ^{242}Pu .

2. Neutrons from alpha-n reactions due to decay of ^{236}Pu , ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu , and ^{242}Pu .
3. Gamma rays from the various plutonium isotopes.
4. Gamma rays from the ^{241}Am daughter of ^{241}Pu .
5. Gamma rays from the ^{237}U daughter of ^{241}Pu .
6. Gamma rays from the daughters of ^{232}U , itself a daughter of ^{236}Pu .

Neutrons from alpha-n reactions will be much more serious in the case of oxide fuels and carbide fuels than in the case of metal fuels because of the presence of the light elements oxygen and carbon in the fuel. The absolute amounts of actinide activity will be a function of the isotopic ratio and cooling time before and after reprocessing. While existing data do not permit accurate estimates of the actual amounts of radiation that will be associated with the actinide elements in recycled fuel, it will be significant, and at least semiremote if not fully remote fabrication will be required in all cases. (Since the 10,000-MWe oxide plant requires the fabrication of 100,000 pellets/day, automation would be required in any case and "glove-box handling" is a nominal term.)

APPENDIX G

Economic Penalty for Low-decontamination Pyrochemical Processing

Since the aqueous and volatility processes are high-decontamination processes and the pyrochemical process selected for this study is a low-decontamination process, there is a difference in the radioactive fission-product content of refabrication feed material produced by pyrochemical processing and the other processes. This difference has little effect on the worth of the product as a core or blanket element in the reactor because all the processes satisfy the processing requirements of fast-breeder-reactor fuels. However, it does increase the cost of those fuel-cycle operations subsequent to processing, including refabrication, storage of finished fuel elements, and the loading of fuel elements into the reactor.

To compare the economics of alternate processes, the categories of cost allotted to processing should be sufficiently inclusive to render the other fuel-cycle costs matters of economic indifference. Thus a monetary valuation, which constitutes an economic penalty to be levied against pyrochemical processing, must be associated with low decontamination.

Of the fuel-cycle operations subsequent to processing, the only one that has a significant effect on the economic penalty is refabrication. In an integrated reactor-fuel cycle complex, the inventory of finished-fuel assemblies is kept at a minimum and the additional annual charge for shielded facilities in which to store these assemblies is negligible. The annual charge for the additional cost of a fuel-handling machine, which serves both the loading and unloading of the reactor, is also negligible and would probably be done in any case. Therefore, the economic penalty is essentially a measure of the increased cost of refabrication attributable to the use of a processing product that has been only partially decontaminated. Since in this study, remote handling of fully decontaminated core fuel was specified because of the presence of plutonium isotopes in recycled plutonium, only the separable costs of blanket fabrication are relevant.

The present study did not include the estimation of refabrication costs, and no studies have been published that provide a definitive comparison of remote and contact refabrication costs for uranium-plutonium fuels. Therefore, complete analysis of the economic penalty is not possible. Separable costs of the remote refabrication of metal fuel in an integrated processing-refabrication fuel-cycle facility have been estimated by Argonne National Laboratory.¹ These data, together with other unit costs based on the EBR-II Fuel Cycle Facility and other installations at Argonne National Laboratory were employed to estimate the economic penalty for metal fuel. Using this estimate as a reference and with the application of heuristics, estimates were also developed for the economic penalty for oxide and carbide fuels.

The following estimated unit costs based on the annual throughput in kilograms per year at 80% plant factor for remote fabrication were developed from the data in ANL-7001:¹

1. Remote-equipment cost	\$21.9/kg
2. Direct-labor cost	\$15.4/kg
3. Remote cell space required	0.039 sq ft/kg
4. Cost of remote cell space	\$775/sq ft
5. Contact-processing area cost	\$ 70/sq ft

The following assumptions are employed:

1. The throughput in a contact plant is 25% greater than in a remotely operated plant, when the two types of plants are compared on the basis of equal physical amounts of labor or of equipment.
2. The average cost of a unit of equipment is 25% lower in a contact plant than in a remotely operated plant.

Based on the foregoing specifications, the following unit costs are obtained for building, equipment, and direct labor:

	Unit Cost, \$/kg	
	<u>Remote</u>	<u>Contact</u>
Building	30.2	2.2
Equipment	21.9	13.1
Direct labor	15.4	12.3
(annual cost)		

These unit costs can be converted to annual unit charges by application of the capital-replacement schedules of this study, i.e., 30 yr for building, 15 yr for contact processing equipment, and 5 yr for remote-cell processing equipment. The application of these schedules at the 6.75% interest rate and 13% return on investment of this study yields the following statistics for the difference in cost between remote and contact refabrication of blanket material:

	Annual Unit Charge Difference between Remote and Contact Refabrication, \$/kg of Blanket
Building	
a. Capital replacement	0.3
b. Return on investment	3.6
Equipment	3.3
Labor	3.0
Total difference	10.2

The penalty of \$10/kg applies only to the blanket elements and not to the fuel elements. This differential is the estimate for a plant serving three reference reactors.

The effect of the economic penalty is apparent from the data in Table G.I. This does not alter the conclusion that pyrochemical processing is the economic choice for the processing of metal fuel.

TABLE G.I. Comparison of Costs of Alternative Processes, Including an Economic Penalty in Pyrochemical Processing for Low Decontamination (Plant with annual capacity of 68,100 kg for service of three reactors)

	Unit Costs, \$/kg		
	Aqueous	Volatility	Pyrochemical
<u>Metal Fuel</u>			
Processing	101	75	52
Waste disposal	38	25	17
Inventory	33	18	18
Low-decontamination penalty ^a	-	-	7
Total	172	118	94
<u>Oxide Fuel</u>			
Processing	88	65	79
Waste disposal	38	25	24
Inventory	33	18	18
Low-decontamination penalty ^a	-	-	5
Total	159	108	126
<u>Carbide Fuel</u>			
Processing	99	83	81
Waste disposal	38	24	24
Inventory	33	18	18
Low-decontamination penalty ^a	-	-	4
Total	170	125	127

^aBlanket fabrication penalty in \$/kg is applied to the 70% of the loading that is blanket.

The differences so calculated may be high, since it is likely that some equipment used for fuel-element assembly and testing would also be used for blanket elements, and the realizable savings may actually be no more than one-third or one-half of the \$10/kg estimated for completely separate facilities.

Adequate data are not available for the estimation of the economic penalty in the pyrochemical processing of oxide and carbide fuels. The following are some factors that should be considered:

1. Uranium carbide should be handled in inert atmosphere to avoid contamination with oxygen. In this study, a unit cost of \$350/sq ft was applied for inert-atmosphere gloveboxes, including the building to house the gloveboxes. More time is required to perform an operation in a glovebox than by direct contact, and the unit cost for glovebox direct labor is about the same as it is for fully remote operations.

2. Uranium dioxide can be handled in air in hooded facilities.

These considerations and the assumption that all other factors are matters of economic indifference, and that the metal-fuel unit costs are applicable, afford a guess as to the economic penalty in the pyrochemical processing of oxides and carbides. This estimate of the cost penalty is:

Oxide	\$7/kg
Carbide	\$5/kg.

As for the metal fuel, the realizable savings may be no more than one-third to one-half of the calculated value for separate facilities.

An estimate of about \$15/kg for the difference in cost between remote and contact fabrication of oxide fuel was developed by McKee.²

References for Appendix G

1. L. E. Link, J. E. Ayer, D. K. Butler, K. A. Hub, W. B. Loewenstein, W. J. Mecham, D. Meneghetti, D. H. Thompson, V. G. Trice, Jr., and J. T. Weills, *1000 MW(e) Metal-fueled Fast Breeder Reactor Concept*, ANL-7001 (to be published).
2. R. W. McKee, *Economic Evaluation of Close-coupled Fuel Processing with Plutonium Recycle*, BNWL-28 (Jan 1965).

APPENDIX H

Model for Out-of-reactor Fuel Inventory

The charges for out-of-reactor fuel inventory in this report were based on a simple method of inventory valuation of \$10/g of ^{239}Pu and ^{241}Pu for the quantities of these materials that were estimated to be held up in the course of all out-of-reactor operations. This appendix compares the out-of-reactor inventory for close-coupled and central plants of the types and sizes considered in this study. The inventory quantities presented here were used directly for the inventory charges of the close-coupled plants. In the case of the central plant, the inventory charges were subject to the further requirement of minimization of the combined costs of inventory and shipping.*

The basis of determining the out-of-reactor fuel inventory is the schedule of reactor refueling and the holdup times in the several out-of-reactor fuel-cycle operations. These time schedules are shown in Table H.I for a close-coupled plant serving a single reactor.

TABLE H.I. Determination of Quantity of Out-of-reactor Fuel Inventory
for a Close-coupled Plant Serving One Reactor
(Reactor refueling period: 180 days)

Item	Pyrochemical or Volatility Process	Aqueous Process
1. Holdup time for cooling of fuel discharged from reactor, days	≥ 15	≥ 120
2. Holdup in processing, days	10	10
3. Analysis and batch preparation, days	5	5
4. Holdup in fabrication, days	15	15
5. Holdup storage of fabricated fuel at reactor, days	≥ 15	≥ 15
6. Sum of above holdup times, days	≥ 60	≥ 165
7. Total time interval between refuelings = time required to process entire batch, days	180	180
8. Maximum net processing time available in a single reloading interval to process fuel discharged at the start of that interval, days	$180 - 60 = 120$	$180 - 165 = 15$
9. Maximum fraction of fuel discharged at start of a reloading interval that can be processed within that interval	$\frac{120}{180}$	$\frac{15}{180}$
10. Additional fraction of fuel required for a reload, above that amount processed within the single interval terminating with that reload	$120 - \frac{120}{180} = 0.33$	$1 - \frac{15}{180} = 0.917$
11. Total amount of out-of-reactor fuel inventory as a factor times the reload quantity W, kg	1.33W	1.917W

*Inventory and shipping charges for central plants were presented in Appendix E.

If W is the quantity of core and blanket fuel discharged from a reactor every 180 days, the average daily process rate for a matched, close-coupled fuel cycle facility is $W/180$. As shown in Table H.I for the aqueous process, none of the fuel discharged at time t can enter processing before time $t + 120$, and none of it will be available for the next refueling unless it entered the process no later than $t + 135$. Only $(15/180)W$ of the batch W discharged at time t is available for refueling at time $t + 180$. Therefore an additional quantity, $(165/180)W$, must be in inventory to provide for the refueling. Hence, the minimum inventory factor for a close-coupled aqueous plant serving one reactor is $(1 + 165/180)$ and the minimum inventory is $(1 + 165/180)W$. By the same reasoning, the inventory factor for the non-aqueous plant schedule is 1.33. The inventory quantities calculated by this basic model are summarized in Table H.II for the plant sizes of this study. A further explanation of the table with respect to the three- and ten-reactor cases is given in the following paragraphs.

TABLE H.II. Comparison of Minimum Inventory Factors
for Plants Serving One to Ten Reactors

Fuel and Plant	Minimum Inventory Factor ^a	
	Aqueous	Volatility or Pyrochemical
Close-coupled plant serving one reactor	1.92	1.33
Close-coupled plant serving three reactors	$3.75/3 = 1.25$	$2.00/3 = 0.667$
Central plant serving ten reactors	$10.44/10 = 1.04$	$4.89/10 = 0.49$

^aThe minimum inventory factor is the factor which, when multiplied by the quantity of fuel discharged per reactor per cycle, equals the minimum out-of-reactor inventory.

For a processing plant serving three reactors, it was assumed that the 180-day reactor refueling periods were staggered at 60-day intervals to allow minimum out-of-reactor fuel inventory by transfer of fuels between reactors rather than waiting to insert the fuel in the same reactor. In effect, the time between refuelings is 60 days, rather than 180 days. The same holdup time schedules applied as shown in Table H.I. For this three-reactor processing plant, 60 days is the time required to process one reactor batch. For the nonaqueous plant schedule, the maximum net processing time available is zero (60 days holdup - 60 days processing = 0), corresponding to Item 8 of Table H.I. Thus, in addition to the fuel batch

awaiting processing, a second fuel batch, already processed, must be on hand for the refueling. Therefore, two reactor batches constitute the out-of-reactor inventory of the nonaqueous plants. The inventory per reactor is 0.667, as shown in Table H.II.

For the corresponding aqueous plant for processing three reactors, the 165-day holdup corresponds to an inventory comprising the initial batch, an additional batch discharged on each of the two successive 60-day periods, plus an additional 45-day period. Thus, only 15 days of four 60-day periods was available for processing, and the total out-of-reactor inventory is 3.75 reactor batches or 1.25 batches per reactor, as shown in Table H.II.

For the central plant serving ten reactors, uniform spacing of the 180-day reactor-loading intervals was again assumed, resulting in 18-day refueling intervals between reactors. Because of shipping fuel to and from the reactor, the holdup times were slightly longer than shown in Table H.I for the close-coupled plants. Central-plant holdup times were taken as 165 days for the aqueous and 70 days for the nonaqueous plants. By use of the reasoning employed in the previous cases, the inventory quantities calculated for the central plants were those shown in Table H.II.

The comparison of out-of-reactor inventory quantities in Table H.II for the case of this study shows that inventory can be reduced substantially both by shorter holdup time (especially shorter cooling) and by multiple reactor service, wherein fuel may be shuffled between reactors. Those effects combine in the concept of a reactor park serviced by a close-coupled fuel cycle facility employing volatility or pyrochemical processing.

The above discussion did not deal with differences in inventory quantities between pyrochemical and volatility processes, nor with differences between metal, oxide, or carbide fuels. Such differences, arising in differences in holdup times for the alternative operations of processing and fabrication, cannot in general be ignored. However, under the rules and purposes of this study, such differences appeared small compared to the major sources of differences in inventory quantities cited above.

APPENDIX I

Exhibit of Detailed Flowsheets of Conceptual Fluid-bed Volatility Process

Detailed flowsheets of the reference process for the fluid-bed volatility plant were prepared as a basis for the cost estimates. These flowsheets are shown in Figs. I.1 through I.9 for the processing of the oxide core and blanket of the reference 1000-MWe fast breeder reactor. The overall process concept, the plant-design philosophy, and the adaption to other fuels and plant sizes were pointed out in the preceding appendixes. The stream quantities, flow rates, and processing times are not to be regarded as optimum or final, but are indicative of a process believed feasible on the basis of present knowledge.

The application to the single oxide-fueled 1000-MWe reactor constitutes the basic volatility-processing plant. No major item of processing equipment needs to be furnished in multiple quantity to achieve the specified plant capacity of this case. However, the condenser in the primary fluorination step (for collecting the UF_6 and bromine fluorides) may have to be a two-stage unit operating at two temperatures (say, 0 and -55°C) in order to allow (a) condensation of UF_6 as a liquid solution in BrF_5 and (b) the low BrF_5 losses indicated.

Fig. I.1. Volatility Flowsheet: Decladding Operation

Fig. I.2. Volatility Flowsheet: Decladding Off-gas Treatment

Fig. I.3. Volatility Flowsheet: Primary Fluorination

Fig. I.4. Volatility Flowsheet: Secondary Fluorination

Fig. I.5. Volatility Flowsheet: Fractional Distillation

Fig. I.6. Volatility Flowsheet: Converter Reagent and Hexafluoride Feed Operations

Fig. I.7. Volatility Flowsheet: Converter Operations

Fig. I.8. Volatility Flowsheet: Fluorination Waste-gas Reactor.

Fig. I.9. Volatility Flowsheet: Rare-gas Concentration and Storage

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Cost data taken from sources other than Argonne National Laboratory are referenced in this report. However, such data, principally on aqueous processing and on radioactive waste storage, have been reinterpreted in this study, and the final results do not necessarily reflect the judgment of the original authors.