



OAI 3 4456 0513325 4 . LABORATORY

operated by

UNION CARBIDE CORPORATION
NUCLEAR DIVISION



for the

U.S. ATOMIC ENERGY COMMISSION

ORNL - TM - 1784

PRELIMINARY INVESTIGATION OF PROCESSING FAST-REACTOR FUEL
IN AN EXISTING PLANT

E. L. Nicholson



LEGAL NOTICE

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

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

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

ORNL-TM-1784

Contract No. W-7405-eng-26

CHEMICAL TECHNOLOGY DIVISION

PRELIMINARY INVESTIGATION OF PROCESSING FAST-REACTOR FUEL
IN AN EXISTING PLANT

E. L. Nicholson

MAY 1967

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
operated by
UNION CARBIDE CORPORATION
for the
U. S. ATOMIC ENERGY COMMISSION

LOCKHEED MARTIN ENERGY RESEARCH LIBRARIES



3 4456 0513325 4

PRELIMINARY INVESTIGATION OF PROCESSING FAST-REACTOR FUEL
IN AN EXISTING PLANT

E. L. Nicholson

ABSTRACT

A preliminary investigation was made of the aqueous processing of fast-breeder-reactor fuel in the Nuclear Fuel Services plant (NFS). The spent fuel consisted of stainless-steel-clad uranium-plutonium oxide, containing about 10% plutonium (heavy-metal basis), that had been irradiated to an average burnup of 38,500 Mwd/ton (core plus blanket). A processing capacity of 0.52 ton/day was selected to be compatible with the existing NFS operating license, although a daily throughput of up to 1.4 tons of fast-reactor fuel is technically feasible, assuming that some additions are made to the plant. The solvent extraction flowsheet is based on the use of 15% TBP and feeds of sub-critical concentrations (≤ 5.0 g of fissile plutonium per liter) in existing geometrically unrestricted equipment. A shielded, high-capacity, continuous anion exchange facility for purifying and packaging plutonium was specified to provide sufficient plutonium capacity. The irradiation dose that would be received by the solvent in the extraction section of the HA column was calculated to be 0.017 whr/liter for fuel cooled 150 days and only 0.043 whr/liter for fuel cooled 30 days. Processing after 30 days, or at even shorter cooling periods, was shown to be technically feasible. A brief cost study indicated that shipping and processing the spent fuel from ten 1000-Mw (electrical) fast reactors in the NFS plant would cost 0.14 mill/kwhr (electrical). Improvements in processing procedure that should significantly reduce the preceding cost and simplify operations are also discussed.

1. INTRODUCTION

Research and development of aqueous methods for processing fast-reactor fuels was started at the Laboratory in July 1966. This work should eventually yield optimum processing procedures and more realistic cost estimates based on conceptual designs of both small on-site plants and central processing facilities. In order to guide the research program and to establish a present-day unit processing cost, the capability of the Nuclear Fuel Services plant (which was originally designed for processing water-cooled reactor fuels) for processing fast-reactor fuels was

investigated, assuming present-day technology and a minimum of plant changes, most of which are concerned with increasing the existing plutonium capacity. Possible restrictions imposed by fuel shipping, the shear-leach head-end process, criticality, solvent irradiation, plutonium and uranium product activity, iodine, etc.--that is, most of the factors that might make aqueous processing of fast-reactor fuels unfavorable--were also examined. None appeared to cause serious problems, even though the NFS plant was not designed for this type of fuel. Thus, processing should be even more efficient and considerably cheaper in a plant specifically designed for this fuel. Since only a short time was available for this study, the conclusions presented here may have to be modified later by research and development results. Also, since this study was done completely independently of NFS, opinions expressed by their staff on fast-reactor fuel processing are not included; realistically, their interest in the latter may only be academic.

Results obtained to date indicate that processing fast-reactor fuel at a daily throughput of 0.52 ton (48.8 kg of plutonium) of fuel irradiated to a burnup of 38,500 Mwd/ton (core plus blanket) and cooled 150 days, is feasible at NFS if additional plutonium tail-end and packaging facilities are added. The capacity specified above was chosen for this study because it is roughly equivalent in fission product input (to the plant) to the standard daily throughput of 1 ton of water-cooled reactor fuel that has been irradiated to a burnup of 20,000 Mwd/ton. A maximum throughput of about 1.4 tons of fast-reactor fuel per day appears possible if the existing iodine removal system performs at a higher efficiency than that assumed for safety analysis purposes and if tritium and krypton disposal to the environment is not limiting. If NFS had taken full advantage of disposal limits permitted by existing safety regulations, a much larger daily throughput (perhaps as high as 8 to 10 tons of fast-reactor fuel) would be theoretically possible at this site. A throughput of about 3 tons/day would be possible if the extraction section of the HA column were the only capacity-limiting item.

The irradiation dose received by the solvent was investigated in detail in pulsed columns for a variety of designs and operating conditions and was found to be negligible in well-designed equipment. Finally, processing at NFS at ≤ 30 days' cooling is reasonable from the standpoint of solvent irradiation but would require upgrading the off-gas cleanup system and adding tanks for storage of the uranium product to permit decay of ^{237}U .

2. REFERENCE FAST-REACTOR FUEL

The liquid-metal-cooled, oxide-fueled fast-breeder type of reactor was chosen as the reference for this study because it appears to be nearer present-day technical capability than either carbide- or metal-fueled fast breeders. This reactor is based on GEAP-4418 (ref. 1) as extensively modified in a subsequent study.² Its core³ consists of $\text{UO}_2\text{-PuO}_2$ with an effective initial enrichment of 17.9%; the axial blankets are depleted UO_2 . It does not have a radial blanket; its core and axial

blanket are contained in the same pin and are thus processed simultaneously. Burnups of the core, the axial blanket, and the mixed core-plus-blanket are 110,000, 6600, and 38,500 Mwd/ton respectively. The reactor is refueled every six months and discharges 18.9 tons of fuel annually.

An average subassembly contains 429 pins, 0.265 in. OD and ~52 in. long; after disassembly at the reactor to remove BeO pins, sheath, grids, and the fission-gas vent chamber, it has the following approximate composition:

U	124 kg
$^{240}\text{Pu} + ^{242}\text{Pu}$	3.7 kg
$^{239}\text{Pu} + ^{241}\text{Pu}$	9.7 kg
Fission products	5.8 kg
Type 316 stainless steel	~54.5 kg

The spent fuel contains approximately 10% Pu (heavy-metal basis), of which approximately 72% is fissionable. The fission product activity of the core-plus-blanket is given below.

<u>Cooling Time (days)</u>	<u>Watts per kg of Initial (U + Pu)</u>	
	<u>Beta</u>	<u>Gamma</u>
30	38.2	42.4
150	17.4	11.3

The irradiation dose received by the solvent is so low that cooling times of less than 30 days would be feasible in a plant designed to cope with the increased amounts of iodine.

Each fuel pin is vented in such a manner that sodium does not enter the fueled portion of the pin; 35% of the krypton produced during irradiation is estimated to escape while the pin is in the reactor. Removal of the vent chamber and the cleaning of the external surfaces should eliminate any sodium hazard associated with processing normal pins.

3. FUEL SHIPPING, RECEIPT, AND STORAGE

The complete fuel subassembly is approximately 24 ft long. The reactor designers favor removal of the hardware from the fuel at the reactor to simplify shipment; therefore, additional disassembly to individual pins at the reactor before shipment may not be unreasonable.³ No cost was assigned to this operation since NFS

has a disassembly system that could be used for fast-reactor fuel processing at the standard throughput rate assumed for this study. In the cost study (Sect. 10.0), additional head-end costs are added to take care of disassembly, shearing, and leaching at high throughputs.

R. Salmon has studied the shipping cost for the reference fast-breeder fuel.⁴ His results represent a feasible, but nonoptimized, design because the configuration was not susceptible to computer optimization by the MYRA code. The abstract of his report states:

"Irradiated fuel shipping costs of \$11 per kg U + Pu at 20 days' cooling and \$10 per kg U + Pu at 90 days' cooling were calculated for the reference fast oxide breeder. This was based on rail shipment, using a distance of 1000 miles each way.

The fuel elements were assumed to be disassembled and packed into 2-in. steel pipe 'cans,' each can being 54 in. long. The cask carries 184 cans. A cooling system is required.

The cost of disassembly is not included in the figures given.

Inventory costs are about \$0.20 per kg U + Pu per day, valuing fissile plutonium at \$9 per gram."

The shipping cost, about 0.03 mill/kwhr (electrical), is reasonable. We can conclude that short-cooled fuel shipment is feasible and within the intent of the latest shipping regulations. The above results indicate that cooling times even shorter than 20 days are possible if inventory charges are considered; however, the problems in dismantling the fuel at the reactor and the effect of very short cooling on processing costs would have to be investigated. The latter effect is probably minor in a plant specifically designed for fast-reactor fuel.

Receipt of the fuel carrier, unloading the pin containers, and holdup in the NFS storage canal would be a routine operation that is not significantly different from present methods.

4. PROCESSING PLANT CAPACITY

The NFS standard throughput for oxide fuel irradiated to a burnup of 20,000 Mwd/ton and cooled 150 days⁵ is 1 ton/day. Using previously published data,⁵ I have estimated that the maximum instantaneous capacity of the plutonium anion exchange system, with no delays between operational steps, is ≤ 14.6 kg of plutonium per day. However, because of batch processing and quite limited column feed conditioning and waste handling facilities, the actual capacity is probably no more than 8 kg of plutonium per day. Thus, obtaining a reasonable throughput of fast-reactor

fuel with existing NFS facilities is impossible without some changes, which must include the addition of an adequately shielded packaging facility for handling large amounts of high-burnup plutonium.

A daily throughput of 0.52 ton of fast-reactor fuel that had been irradiated to 38,500 Mwd/ton and cooled 150 days was selected as being approximately equivalent to the standard throughput. This requires a capability for processing 48.8 kg of plutonium per day. While this capacity could probably be attained by upgrading the present batch ion exchange system, I thought it more interesting and conservative from the standpoint of cost to assume the addition of a high-capacity continuous processing plutonium facility in a new building adjacent to the main plant. The maximum daily throughput of this facility would permit processing up to 1.4 tons of fast-reactor fuel, which is equivalent to 131 kg of plutonium. This upper limit is set by my estimate of the capacity of the existing NFS acid recovery system and it would be subject to revisions of the site disposal limits, or possibly to improved fission product containment, depending on what the plant experience actually discloses.

The NFS safety analysis report covering disposal of ^{85}Kr , ^{131}I , and ^3H was conservative, and full credit was not taken for all dilution factors, etc., that would have permitted higher throughput at this site.⁵ For example, the license⁶ permits the daily release of the amount of ^{85}Kr that is equivalent to 2 tons of "standard" fuel; dilution factors ten times greater than those assumed in the calculations can normally be expected. For ^{131}I , the off-gas treatment system is assumed to remove 99.5% of the total daily ^{131}I input (1.8 curies per ton of fuel), which is a rather low efficiency for an iodine cleanup system consisting of a caustic scrubber and a silver tower in series. Also, not all the iodine will be volatilized from the process solutions in the plant. Additional safety factors external to the plant but not used would permit a 15-fold increase in iodine release.⁶ Finally, a daily release to the creeks of 130 curies of ^3H (from 1 ton of standard fuel per day) in low-level waste water is only about 1.3% of the permissible on-site limit and 5% of the permissible off-site concentration in the surface waters, even though the rivers and creeks at this particular site are rather small. Thus, it appears that upward revision of the plant capacity would not be unreasonable after monitoring has verified the safety analysis calculations.

5. SHEAR-LEACH HEAD-END PROCESS

At NFS, pins would be removed from the shipping tubes and sheared into geometrically safe fuel baskets (6.3 in. ID) having fuel capacities of about 150 kg. B. C. Finney,⁷ who has investigated the shearing of loose bundles of fuel pins, finds that it presents no unusual problems and can probably be done at the rate of 0.5 ton/day. Calculations of fuel temperatures in this process showed that the maximum temperature ($< 1230^\circ\text{F}$) occurred at the center line of the basket, assuming that only core material is present. This is quite acceptable, and the inclusion of the blanket material will further reduce the fuel temperatures.

Failed fuel elements will be segregated and sheared into baskets equipped with a purge tube extending to the bottom of the liner. Humidified inert gas, steam, and, finally, water will be introduced into the chopped fuel via the purge tube to effect slow and safe reaction of any sodium before charging to the dissolver. This probably could even be done with routine charges if desired, since storage space is provided in the general purpose cell for filled fuel baskets.

With respect to heat transfer and criticality control, the NFS dissolver is adequate for dissolution of the mixed oxide fuel without any modification.

6. CRITICALITY CONTROL

A conservative approach to criticality control, approximately equivalent to NFS standards, is assumed. Concentration control is specified for nonpoisoned or geometrically unrestricted vessels, thus limiting the plutonium concentration to ≤ 7.0 g of plutonium per liter⁸ (fissile plus nonfissile plutonium) in the head-end process. No allowance is made for the poisoning effect of ^{240}Pu , ^{238}U , or NO_3^- . Soluble nuclear poisons are technically feasible (they are added routinely at the ICPP as a primary criticality control) and could be used, in conjunction with more-concentrated feeds, to obtain the maximum possible throughput. They are not required at NFS, however, since the columns are oversized. In any case, use of dilute feeds and $< 30\%$ TBP extractant would not reduce the capacity prohibitively. For example, at comparable feed concentrations (20 to 100 g of uranium per liter), the flooding rate for 20% TBP extraction is roughly twice that for 30% TBP.⁹

The 10-in.-diam H cycle extraction, partitioning, and uranium stripping columns are safe up to concentrations in excess of 40 g of plutonium per liter; they have poisoned disengaging sections that are safe at concentrations up to twice the critical concentration for the 10-in.-diam columns.⁵

If additional safety is desired for the 10-in.-diam columns or if even larger pulsed columns were desired for a multiton/day plant, the pulse plates could be made of poisoned stainless steel, which would permit substantially higher plutonium concentrations and/or column diameters. The existing 10-in.-diam HA column extraction section at NFS has a potential daily capacity of 3 tons of fast-reactor fuel at 75% of flooding with the suggested flowsheet!

Both the HBS column (4 in. in diameter) and the plutonium cycle columns are geometrically safe.

Fifteen percent TBP is specified as the solvent in the H Cycle and the plutonium cycle as an additional (and probably unnecessary) safeguard. If uranium and plutonium are extracted in the same ratio as that of their concentrations in the HA column feed (plutonium is less extractable), then the organic phase in the H cycle equipment will contain less than 7 g of plutonium per liter. Fifteen percent TBP is used in the

geometrically safe plutonium cycle, since the throughput obtained with this solvent is adequate and since it is desirable to use the NFS standard backcycle procedure of sending the 2BW to the H cycle to minimize plutonium losses. No criticality or capacity limitations exist in the final uranium cycles. An always-safe design is specified for the equipment for the new high-capacity plutonium tail-end facility.

7. SOLVENT EXTRACTION AND ANION EXCHANGE FLOWSHEETS

The solvent extraction flowsheets (Figs. 1 and 2) were prepared by visual inspection of tabular data¹⁰ for a 20% TBP system, since data for the 15% TBP system are not available. These are not optimized flowsheets but are quite adequate for estimating purposes and for use as a starting point for flowsheet development. Since flooding data for 20% TBP were used,⁹ estimates of flooding rate and solvent irradiation are conservative. Prior to design of an optimized plant, these points and other questions such as the optimum TBP concentration and performance of the partitioning column at high plutonium concentrations should be studied. Plutonium reduction and partitioning could be avoided by using anion exchange resin to effect the uranium-plutonium separation.¹¹

The H Cycle (Fig. 1) is designed to give a high plutonium concentration in the HBP stream to minimize the volumetric throughput in the existing 2A and 2B columns. The 2A column (Fig. 2) should be operated at a slightly elevated temperature to avoid any possibility of third-phase formation. Sulfuric acid is used in the 2BX column to complex plutonium and to minimize backcycling losses in the 2BW.¹² Operating the 2B column as a partitioning column to reduce uranium carryover with plutonium is also possible but is unnecessary since good uranium separation¹¹ can be obtained in the anion exchange facility.

The 2BP stream containing about 60 g of plutonium per liter, is pumped to the plutonium tail-end facility (Fig. 3) and continuously adjusted for feeding to the Higgins continuous anion exchange unit.¹² The bed in this unit has an effective cross-sectional area of 1 ft², with a geometrically safe annular configuration, and a maximum capacity of at least three times the normal throughput of about 50 kg of plutonium per day. Plutonium product is evaporated continuously to 200 g of plutonium per liter and is held in geometrically safe annular storage tanks. It is assumed that the plutonium would be shipped as nitrate solution; however, for safety reasons it might be desirable to ship it as PuO₂. Addition of a small fluid bed or screw calciner¹³ to produce PuO₂ would affect the cost only slightly, and sufficient overage has been allowed in the cost estimate for this option.

In the present NFS flowsheet, all the anion exchange wastes are backcycled to the 2AF feed conditioner to recover any plutonium losses. When large amounts of plutonium are processed, the volume of the 2AF stream becomes very large (7.0 M HNO₃ anion exchange wastes are diluted to about 1 to 3 M HNO₃ for 2A column feed). The capacity of both the 2A and 2B extraction columns and the existing NFS

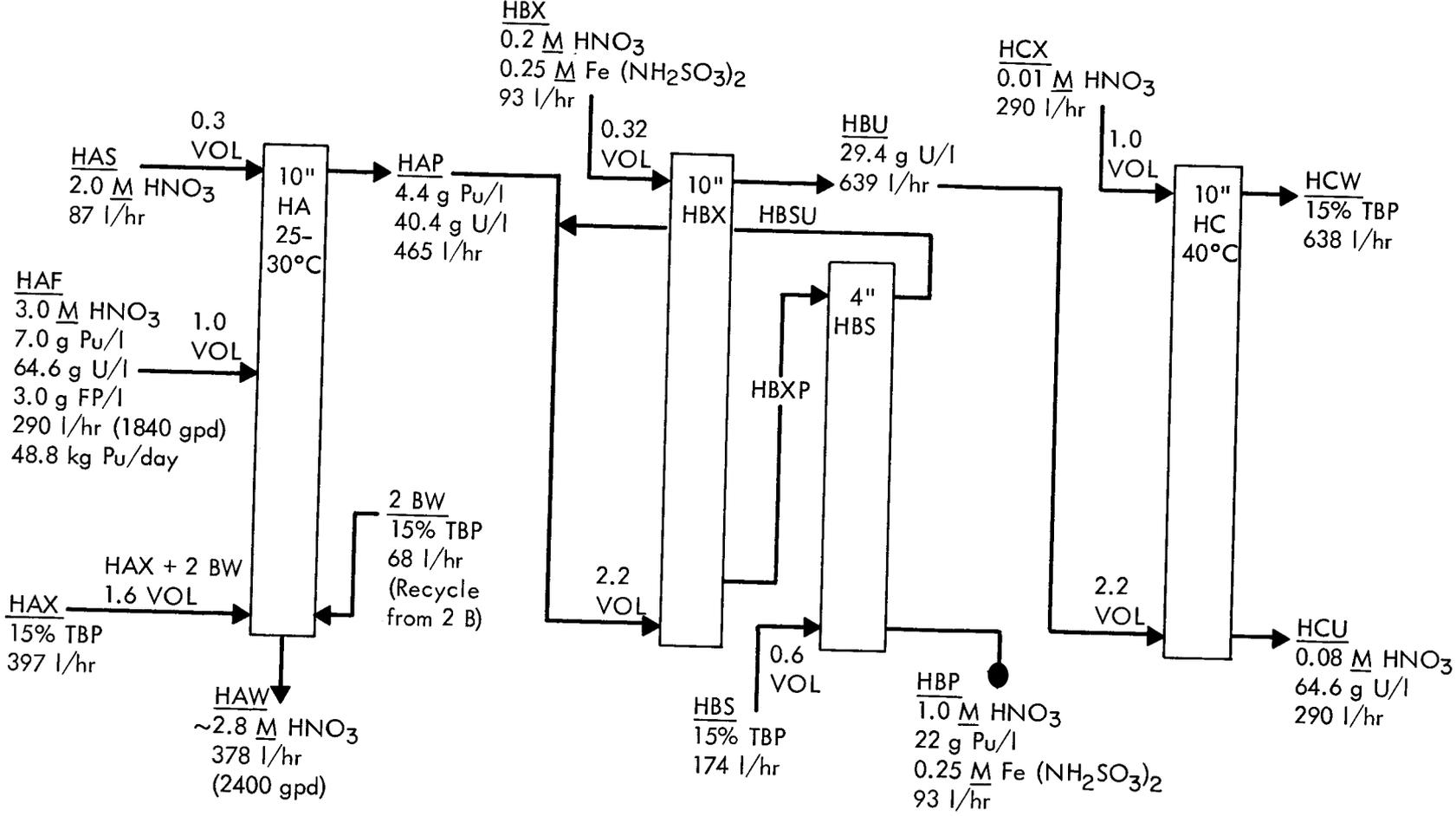


Fig. 1. Nuclear Fuel Services H Cycle Solvent Extraction Flowsheet for Processing Fast-Breeder-Reactor Oxide Fuel.

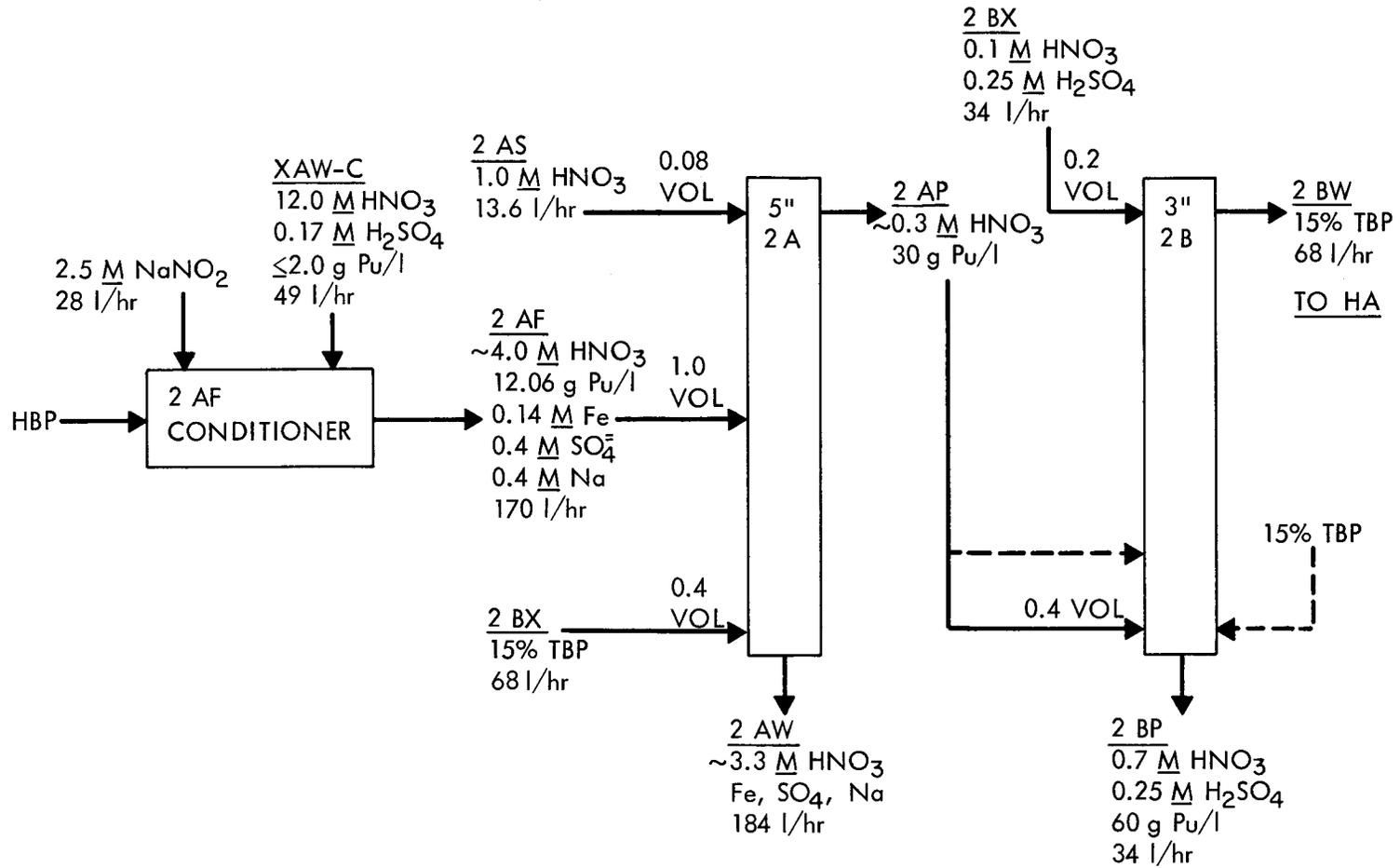


Fig. 2. Nuclear Fuel Services Plutonium Cycle Flowsheet for Processing Fast-Breeder-Reactor Oxide Fuel.

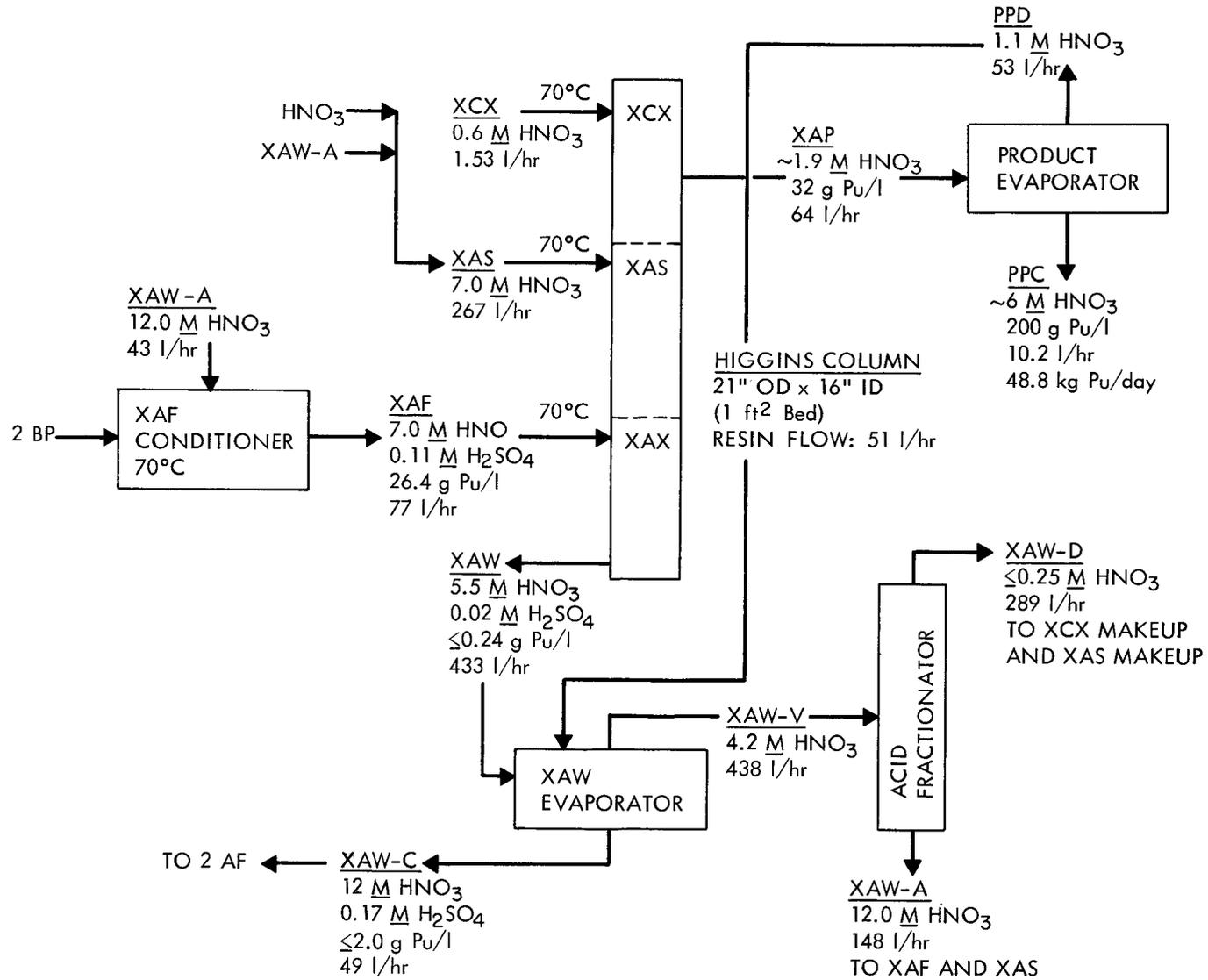


Fig. 3. Nuclear Fuel Services Plutonium Ion Exchange and Concentrator Systems for Processing Fast-Breeder-Reactor Oxide Fuel.

acid fractionation system would be limiting. To avoid overloading the existing equipment, it is assumed that the anion exchange system acidic wastes are continuously evaporated in a new geometrically safe titanium continuous evaporator in the tail-end facility and that the resulting 12 M acid and the slightly acidic condensate from the acid fractionating tower are recycled entirely within the tail-end system. The low-volume acid evaporator bottoms, containing any plutonium losses and fission products, are recycled to the 2AF tank as adjustment acid, thus permitting a high plutonium concentration in the 2AF and anion exchange system feed. The tail-end acid evaporator also permits considerable process flexibility, since more resin washing to improve fission product decontamination can thereby be permitted without overloading the main plant acid recovery system.

With this flowsheet, the acid contribution from the 2A column and the plutonium purification to the existing acid fractionation system is reduced to a reasonable level; thus a maximum capacity of 1.4 tons of fast-reactor fuel per day should be possible.

8. IRRADIATION DOSE RECEIVED BY THE SOLVENT

The problem most frequently cited as the major drawback for aqueous processing of fast-reactor fuels is probably irradiation damage of the solvent. This problem has been examined here in considerable detail so that the preferred operating conditions can be defined to minimize solvent irradiation. These calculations are not sophisticated in that only rough estimates of irradiation in terms of watt-hours per liter of solvent were made. No attempt was made to use G values to calculate dibutyl phosphate production to estimate plutonium stripping losses and decrease in the decontamination factor because the less-sophisticated results indicated that the irradiation levels encountered would be sufficiently low to preclude damage¹⁴ and because of the brevity of this preliminary study. Further investigation is being directed toward correlating calculations and observed effects in operating plants.

The calculations are presented in detail in Appendix A. Briefly, the procedure was as follows: The active volume of the extraction section of the NFS H cycle column was determined.⁵ Flooding data, flooding rate correlations, and equations that permit calculation of the solvent holdup in pulsed columns are included.⁹ Since data are not available for the 15% TBP system, the flooding data for 20% TBP, and in one case for 30% TBP, were used to calculate solvent holdup in the pulsed columns. In general, the flooding rates should be higher for 15% TBP than with 20 or 30% TBP; this tends to make the calculated irradiation results conservative. From flooding data, the characteristic droplet velocity for the dispersed phase was calculated and used to find the fractional holdup of the dispersed phase at realistic operating rates. (This velocity does not change as the total flow varies, provided that the flow ratio of the two phases is constant and the amplitude-frequency product setting of the pulse generator is unchanged from the setting used in determining flooding.) The dose of irradiation received by the solvent was calculated, using the fractional holdup of the

dispersed phase to determine solvent residence time and the relative aqueous-organic volumetric ratio in the column. Rough estimates were also made of the irradiation dose received by the solvent in the disengaging section during organic-continuous operation. It was assumed that all the beta energy and 40% of the gamma energy were absorbed by the solvent, regardless of the column size. Alpha irradiation was neglected since it is usually of minor importance compared with the fission products. It will be the major contributor to the additional irradiation the solvent receives in the scrub section and in the partitioning equipment. Table 1 shows results for the 10-in.-diam NFS column operating under a variety of conditions and for small-diameter columns operating at a high throughput.

From these results, it appears that organic-continuous operation of the pulsed column is preferred to reduce solvent irradiation to the lowest level. In any event, this type of operation is normally preferred for best decontamination since it minimizes the chance of carry-over of contaminated interfacial solids with the product.

It is difficult to compare the irradiation dose obtained during aqueous-continuous operation with previous estimates (for example, the widely quoted Table 2 in ref. 14) for the dose obtained during processing of thermal and fast-reactor fuels since the assumptions are probably different. A cursory examination of some of the 10-year-old Thorex Pilot Plant data¹⁵ indicated that solvent irradiation estimates (both past and present) should be put on a more rational basis. Davis¹⁶ has also pointed out uncertainties in the irradiation estimates originally made for the runs with short-decayed fuel in the Thorex Pilot Plant. My results, in general, agree with those of Davis when the differences in levels of radioactivity are considered. In any case, a correlation between calculated solvent irradiation effects and plant results is needed.

My results indicate that minimum solvent irradiation can be obtained by either grossly oversizing the pulse column and operating it at a low fraction of flooding or by designing it so that it is much smaller and operates much closer to flooding. The latter is preferred for minimizing equipment size but is restrictive if a substantially larger throughput is desired later.

When 30-day-cooled fuel is processed, the irradiation dose received by the solvent is only 0.043 whr/liter. Previously published data¹⁴ indicate that no effect is observed at 0.01 whr/liter, while a twofold decrease in ruthenium decontamination occurs at 0.5 whr/liter. Increasing the irradiation level to a burnup of 110,000 Mwd/ton but maintaining the 30-day cooling period increases the dose to only 0.12 whr/liter.

Solvent in the new Windscale separation plant is reported to receive an irradiation dose of 0.04 whr per liter per pass¹⁷ through the mixer-settlers, while that in the Dounreay fast-reactor fuel processing plant has been reported to receive up to 1.5 whr per liter.¹⁸ If this is correct, then solvent irradiation in pulsed columns for processing fast-reactor fuel that has been irradiated to burnups of $\geq 100,000$ Mwd/ton and cooled for 30 days or less does not seem to be a problem. If it is a problem,

Table 1. Effect of Pulsed-Column Operating Conditions on Irradiation Received by the Solvent
(15% TBP extraction of fast-breeder-reactor oxide fuel irradiated to a burnup of 38,500 Mwd/ton)

HA Column Diameter (in.)	Rate (tons/day)	Flooding (%)	Continuous Phase	Cooling Time (days)	Residence Time in Cartridge (hr)	Solvent Irradiation Received (whr/liter)		
						Cartridge	Disengaging Section	Total
10 (NFS)	0.52	13	Organic	150	0.66	0.0154	0.00152	0.017
10 (NFS)	0.52	13	Organic	30	0.66	0.0389	0.004	0.043
10 (NFS)	1.04	26	Organic	150	0.32	0.0156	0.0015	0.0172
10 (NFS)	1.40	35	Organic	150	0.24	0.0165	0.0017	0.0182
10 (NFS)	0.52	27	Organic	150	0.64	0.0324	0.0033	0.036
10 (NFS)	0.52	17	Aqueous	150	0.025	0.02 to 0.03	None	0.02 to 0.03
4.3	0.52	75	Organic	150	0.099	0.0182	0.006	0.024
4.9	0.52	75	Aqueous	150	0.0091	0.027	None	0.027

Note: The irradiation calculations assumed absorption of 40% of the gamma and 100% of the beta energy by the mixed phases, although a smaller amount of the gamma energy would be absorbed in the small-diameter columns. The irradiation results are generally "less than" values, since column flooding data for 20% and 30% TBP had to be used. Flooding rates for 15% TBP could be higher; hence, both solvent holdup and irradiation dose should be slightly lower than estimated above.

or if it is desired to reduce solvent irradiation to the lowest possible level, then short-residence-time contactors could be used. The heights of the transfer units (HTU's) for 30% TBP extraction with the organic phase continuous¹⁹ are typically about 1 ft. If we assume an HTU of about 2.5 ft for the 15% TBP flowsheet, the NFS HA column would have about eight extraction stages. This is equivalent to organic residence times of about 5 min and 44 sec per stage for the 10- and 4.3-in.-diam columns respectively. The stacked-column contactor²⁰⁻²² has the potential of reducing the contact time to about 6 sec or less per theoretical stage, thus offering at least a fivefold decrease in irradiation compared with the small-pulsed-column method. If the scale-up principle proposed for the stacked-column contactor is valid, then very high capacity could be attained in a geometrically safe unit.

Finally, the daily throughput of the 10-in.-diam NFS column would be about 3 tons of fast-reactor fuel at 75% of flooding. This would provide processing services, at 75% on-stream time, for 43,000 Mw (electrical) of installed fast-reactor power stations. Of course, the NFS plant could not operate at this rate because of various limitations, especially those associated with the head-end process. However, the figure indicates the very high aqueous processing capacity that can be obtained, even using dilute flowsheets that minimize solvent irradiation and criticality problems. Design of other equipment to conform to such high throughputs is possible.

9. IMPROVEMENTS IN AQUEOUS PROCESSING OF FAST-REACTOR FUELS

The following section is mostly speculation about topics that may deserve consideration in developing the optimized plant for processing fast-reactor fuel by aqueous methods.

9.1 Shear-Leach Head-End Process

The shear-leach head-end process could be improved considerably by adoption of a continuous shear-leach operation. This would eliminate a considerable amount of equipment associated with basket handling, as well as the liquid waste contribution from the iron basket liners presently used at NFS. Criticality and heat generation problems would be simplified. Off-gas evolution from a continuous dissolution system would proceed at a very low uniform rate provided that air leakage is minimized. Thus, the off-gas treatment problem would be simplified. Insoluble fission products may be present in the leach solutions, depending on the concentration and type of dissolvent used. Preferably, they would report to the extraction waste rather than being removed by a difficult head-end feed clarification. Pulsed columns have the capability of processing feeds containing large amounts of solids.

9.2 Sodium Disposal for Sodium-Bonded Fuels

Reaction with water may be the most practical method for disposal of sodium in an aqueous process. It is a safe procedure, provided that large amounts of sodium are not permitted to accumulate and the reaction is done in an inert atmosphere. It could be incorporated in the first step in a continuous shear-leach process in which the fuel fragments would be first steamed, then sprayed with water, and finally "soaked" to ensure removal of traces of sodium. Sheared pieces would be conveyed to the leacher, while the fines would be transferred as a slurry. Excess transfer water introduced into the dissolver could be evaporated from the dissolver, or during feed preparation, or compensated for by using a more-concentrated dissolvent. Steam would be a suitable inert atmosphere to use in the sodium hydrolyzer. An inert gas purge would probably be advisable in the dissolver to ensure that the small amount of hydrogen formed (if any sodium enters the dissolver) will be diluted to prevent reaction with the nitrogen oxides in the vapor space.

9.3 Carbide Fuels

The carbide fuel processing problem probably goes hand-in-hand with the sodium bonding problem. (Sodium bonding probably will not be characteristic of oxide fuels.) High-temperature oxidation, pyrohydrolysis, and hydrolysis with aqueous sodium hydroxide are the principal methods of interest for converting the carbides to a form that will yield a solvent extraction feed free of soluble organic materials. However, each of these requires considerably more equipment and more operations than the method suggested below. On the other hand, if the carbides are dissolved directly in acids, the resulting soluble organic compounds are detrimental to the conventional Thorex and low-acid Purex processes. Chemical destruction of these organics is possible but somewhat difficult. Laboratory experiments with feeds prepared by acid dissolution of mixed uranium-thorium carbides (HTGR fuel) indicated that the detrimental effects of soluble organics could be avoided by using high-acid feeds.²³ The extraction of the organics was thereby repressed, and satisfactory performance was obtained. If a similar system could be used with plutonium-uranium carbide fuels, the carbide problem would be simplified. Chemical development is needed to confirm that this procedure is applicable to plutonium carbide and that radiolysis does not make the flowsheet inoperable.

9.4 Iodine and Rare Gases

The processing of short-decayed fuel would require improved iodine containment. Containment would be much easier if a continuous dissolver were used, since only a very small gas volume would require treatment. A system might consist of a scrubber (caustic or other chemical) to remove most of the iodine (90 to 95% efficiency), one or more heated silver-nitrate-coated, berl-saddle packed towers ($\geq 99.9\%$ efficiency), and finally deep beds of impregnated charcoal to sorb any methyl iodide

type of compound²⁴ (99.99% efficiency) or elemental iodine that has passed through the preceding cleanup system. If short half-life xenon were present, it could be held up for decay in a charcoal trap, as is done at the MSRE. If retention of krypton ever becomes necessary, a number of processes such as low-temperature sorption or Freon scrubbing are available to trap it. Other vessels in the head-end and high-level waste system can be expected to be iodine emitters and, as such, should be connected to the iodine cleanup system. One could consider a head-end chemical treatment to volatilize all the iodine from the feed solution. However, this treatment for short-decayed fuel may not remove ^{131}Te and ^{132}Te , which are the precursors of ^{131}I and ^{132}I respectively.

Fission product iodine could also present a decontamination problem in solvent extraction. However, chemical treatment of the feeds in the short-decay Thorex runs yielded iodine DF's of $\geq 10^7$, and very little iodine reported to the solvent.²³ These treatment methods would have to be verified for plutonium-containing fuels. Iodine and rare gas release can be controlled with existing engineering safeguards.

9.5 Product Activities

The plutonium product will require shielding when it is refabricated. Therefore, a fission product decontamination factor smaller than that currently considered acceptable for power reactor fuels should be permissible.

It is definitely cheaper to "manufacture" the bulk component of the fuel, uranium, in unshielded facilities. However, the ^{237}U activity in the uranium might make the processing of short-decayed fuel disadvantageous. Previous studies²⁵ of thorium fuel showed that it was economical to store thorium for decay of ^{228}Th to permit fabrication in unshielded facilities. The same philosophy will be even more applicable to uranium, considering the very short half-life of ^{237}U . Longer-term holdup of uranium product to permit decay of residual ^{95}Zr , ^{103}Ru , and ^{106}Ru may even be indicated if a low-decontamination solvent extraction flowsheet is also specified for uranium.

9.6 Plutonium Partitioning

Development is needed to confirm that the valence of plutonium can be controlled in high-radiation fields and that high concentrations of the usual reductants are satisfactory in the partitioning column. Anion exchange partitioning after one or more co-decontamination cycles would serve to eliminate most of these problems, as well as to decrease the salt content of the high-level wastes.

10. UNIT PROCESSING COST ESTIMATES FOR FAST-REACTOR FUELS AT NFS

10.1 Cost Estimate for a Fast-Reactor Plutonium Facility at NFS

It was assumed that a new plutonium facility would be built adjacent to the main plant to accommodate the equipment for the tail-end flowsheet (Fig. 3) and to allow remote packaging of plutonium for shipment. The design throughput was assumed to be 0.52 ton of fast fuel per day, while the maximum throughput was set at a processing capability of 1.4 tons/day to equal that of the existing NFS plant. The facility was assumed to be shielded and was designed for remote operation and maintenance. The estimated cost was $\$4.9 \times 10^6$, including a 25% contingency factor (see Appendix B). Sufficient overage in the estimate was allowed so that plutonium could be shipped either as a solution or calcined and shipped as a dry solid.

10.2 Estimates of Unit Processing Costs for Fast-Reactor Fuels

Several processing cases are considered. The first, and most expensive, considers the processing of the fuel from one 1000-Mw (electrical) fast reactor at NFS with the addition of the plutonium tail-end facility. The other cases consider the processing of fuel loads for 3000-, 10,000-, and 16,700-Mw (electrical) fast reactors. (The latter represents the maximum capability of the NFS plant for fast fuel.)

The present NFS capital, operating, and waste disposal charges total \$7,050,000 per year,²⁶ or \$31,300 per process day. Annual fixed charges on a new facility could range from 15 to 22% of the capital investment, while operating charges could range from 8 to 10% of the capital investment. In each case, the lower number represents the present NFS cost basis and the higher number represents less favorable financing than NFS obtained on the original facility. The total operating plus capital charges could thus range between 23 and 32% of the capital investment; an average value of 28% will be assumed to apply for any new facilities. Thus the annual cost of the plutonium facility for fast-reactor fuel would be \$1,370,000.

One 1000-Mw (electrical) fast reactor operating at an 80% load factor would discharge 18.9 tons of fuel annually. At the design processing rate of 0.52 ton of fast fuel per day, NFS would require 36.4 process days, resulting in annual charges of \$1,138,000 for the main plant and \$1,372,000 for the plutonium purification facility, or a total of \$2,510,000. These costs, which are equivalent to 0.358 mill/kwhr (electrical) for processing and waste disposal, obviously represent an upper limit because the annual operating charges for the tail-end facility would not be used in a 36-day campaign and, if only one fast reactor existed, a much cheaper alternative to building this special and very expensive facility could be found. In any case, 0.36 mill/kwhr (electrical) is not a prohibitive price for a special fuel in NFS.²⁵

The corresponding annual costs for processing the fuel from three 1000-Mw (electrical) fast reactors at NFS at the design rate of 0.52 ton/day are \$3,413,000 for the main plant and \$1,372,000 for the plutonium facility, or a total of \$4,785,000. This is equivalent to 0.228 mill/kwhr (electrical). In this case, a reasonable use factor is obtained on the special facility (i.e., 108 days/year), which might make the investment worthwhile.

Ten 1000-Mw (electrical) fast reactors would discharge 189 tons of fuel per year. Since this is obviously too much for NFS to process at the design throughput of 0.52 ton/day, it would be processed at the maximum plant throughput of 1.4 tons/day. The tail-end facility is designed for this maximum capacity. However, some plant modifications might be necessary to increase the capacity of the shear-leach head-end process. Note that such a throughput would be theoretically possible and would still not exceed accepted standards (Sect. 4). A liberal assumption would be that the capital and operating costs for head-end improvements are equal to the charges for the plutonium tail-end facility. (This allows 4.9×10^6 for head-end improvements.) If NFS processed fast-reactor fuel at the maximum rate for 135 process days per year, the daily charge for the main plant would have to be increased by 10% to allow for increased chemical and waste disposal charges at the higher throughput. Then the annual costs would be: head-end improvements, \$1,372,000; main plant, \$4,648,000; tail-end facility, \$1,372,000; total, \$7,392,000, or 0.105 mill/kwhr (electrical).

A final case would be to consider NFS fully loaded with fast-reactor fuel at a processing rate of 1.4 tons/day for 225 process days per year. The annual load is 315 tons of spent fuel, which is equivalent to 16,700 Mw (electrical) output from fast reactors. The annual reprocessing costs would total \$10,491,000, or 0.09 mill/kwhr (electrical).

In each case, these costs represent a proportional share of all plant expenses and are based on an actual operating plant. They could be contested on the basis that the NFS charges are unrealistically low. However, public announcements by the General Electric Company about the proposed Midwest Fuel Reprocessing Plant, which is an advanced aqueous-volatility plant designed for fuel irradiated to levels comparable to this study, indicate a capital investment roughly one-half that of the NFS investment. Also, a substantially lower operating cost may be possible. In a competitive economy, it is clear that downward adjustment of processing prices from NFS levels would be possible, particularly in a plant specifically designed for fast-reactor fuel processing and incorporating future aqueous improvements.

The results of this study are compared with a study²⁷ of aqueous processing by Argonne National Laboratory in Table 2. The prime purpose of the ANL study, as stated, was to elucidate cost differences, but not absolute costs, between aqueous, pyrometallurgical, and volatility processing methods for three types of fast reactor fuels. Minor differences exist between the ANL study and this study on amortization, form of the processing plant products, etc. However, in general, these do not

Table 2. Comparison of Estimated NFS Processing Costs for Fast-Breeder-Reactor Oxide Fuel and an ANL Aqueous Processing Cost Study²⁷

Installed Fast-Reactor Power [Mw (electrical)]	Unit Costs [mills/kwhr (electrical)]					
	Processing and Waste Disposal		Spent Fuel Shipping		Total	
	This Study	ANL	This Study ^a	ANL ^b	This Study	ANL
1,000 ^c	≤ 0.36	0.68	0.03	On-site	≤ 0.39	0.68
3,000 ^c	0.23	0.41	0.03	On-site	0.26	0.41
10,000	0.11	0.26	0.03	<0.05 ^d	0.14	<0.31
16,700	0.09	Not estimated	0.03	Not estimated	0.12	Not estimated

^a Calculated by assuming 90-day cooling prior to shipment.⁴ It is technically and economically feasible to ship at cooling times less than 90 days.

^b Assumes negligible on-site shipment costs for fuel, even in a multireactor station.

^c Assumed to be close-coupled plants in ANL study.

^d Includes shipment of refabricated fuel back to the reactor in a special refabricated fuel shipping cask.

significantly affect the numbers in the table and could not possibly account for the observed differences in processing costs. Comparisons of inventory charges will not be discussed, since our study indicates that a truly advanced aqueous process should have the capability to process fuel after a considerably shorter time than the 125-day cooling period assumed by ANL.

Considering the probable growth of fast reactors, it is quite probable that the fuel from the first few will be processed as incremental loads in existing processing plants rather than in small special plants. Even when a substantial increase in inventory charges is allowed for central-plant processing, the small close-coupled plants cannot compete with a central aqueous plant, even including shipping. The most appropriate comparison can be made by examining the processing costs in the case of a 10,000-Mw (electrical) central plant. The costs developed by this study for an actual operating plant with extensive head- and tail-end additions are only 45% of the ANL cost figure of 0.31 mill/kwhr (electrical). If we assume that NFS processes only the fuel from the ten 1000-Mw (electrical) fast reactors, the unit cost, including charges for shipping and for head-end and tail-end additions, increases to only 0.18 mill/kwhr (electrical). ANL concludes from their study that volatility and pyrochemical processing are cheaper than aqueous processing. On the basis of our study, using data from an actual, but somewhat expensive, plant and with additions to adapt to fast-reactor fuel, the ANL conclusion appears questionable, particularly since the alternative methods are undeveloped.

11. ACKNOWLEDGMENTS

The assistance of and the very helpful discussions with E. D. Arnold, C. A. Blake, B. C. Finney, W. S. Groenier, J. P. Nichols, R. H. Rainey, J. T. Roberts, R. Salmon, and W. G. Stockdale are gratefully acknowledged.

APPENDIX A

CALCULATION OF SOLVENT RESIDENCE TIME IN THE HA COLUMN
EXTRACTION SECTION AND ESTIMATES OF IRRADIATION
DOSE RECEIVED BY THE SOLVENT

A-1. VOLUMETRIC DATA FOR PULSED COLUMNS

The NFS HA column operates with a bottom interface. The dimensions of the extraction section and the lower disengaging section of this column are shown in Fig. A-1.⁵ The details of the pulse plates are not specified. Nozzle plates would be the best cartridge choice for organic-continuous operation.^{12,14,19,28} If we assume that 1/16-in.-thick nozzle plates with 23% free area are used and if the volume of the cartridge stringer rods is neglected, then the active volume in the cartridge is:

$$V_{\text{extn.}} = \left[19.67 \text{ ft} - 115 \frac{0.0625 \text{ in.}}{12 \text{ in./ft}} (1 - 0.23) \right] 0.573 \text{ ft}^2 = 11.0 \text{ ft}^3.$$

For the lower disengaging section, the volume occupied by the solvent is estimated to be:

$$V_{\text{Sol.}} = \left[\frac{15 \text{ in.}}{12 \text{ in./ft}} \frac{(1.67 \text{ ft}^2 + 0.573 \text{ ft}^2)}{2} + \frac{7.5 \text{ in.}}{12 \text{ in./ft}} (1.67 \text{ ft}^2) \right] \frac{1.375 \text{ in.}}{1.500 \text{ in.}}$$

$$= 2.2 \text{ ft}^3.$$

A-2. SOLVENT HOLDUP IN THE CARTRIDGE

Groenier, McAllister, and Ryon⁹ have correlated pulse column flooding data and have presented equations that permit calculation of the solvent holdup. The following treatment was suggested by Groenier and is consistent with published flooding data for pulse columns by Gier and others.

For the tentative flowsheet:

$$\text{organic phase: } V_c = \frac{(397 \text{ liters/hr} + 68 \text{ liters/hr})}{28.3 \text{ liters/ft}^3 \times 0.573 \text{ ft}^2} = 28.68 \text{ ft/hr,}$$

$$\text{aqueous phase: } V_d = \frac{378 \text{ liters/hr}}{28.3 \text{ liters/ft}^3 \times 0.573 \text{ ft}^2} = 23.31 \text{ ft/hr,}$$

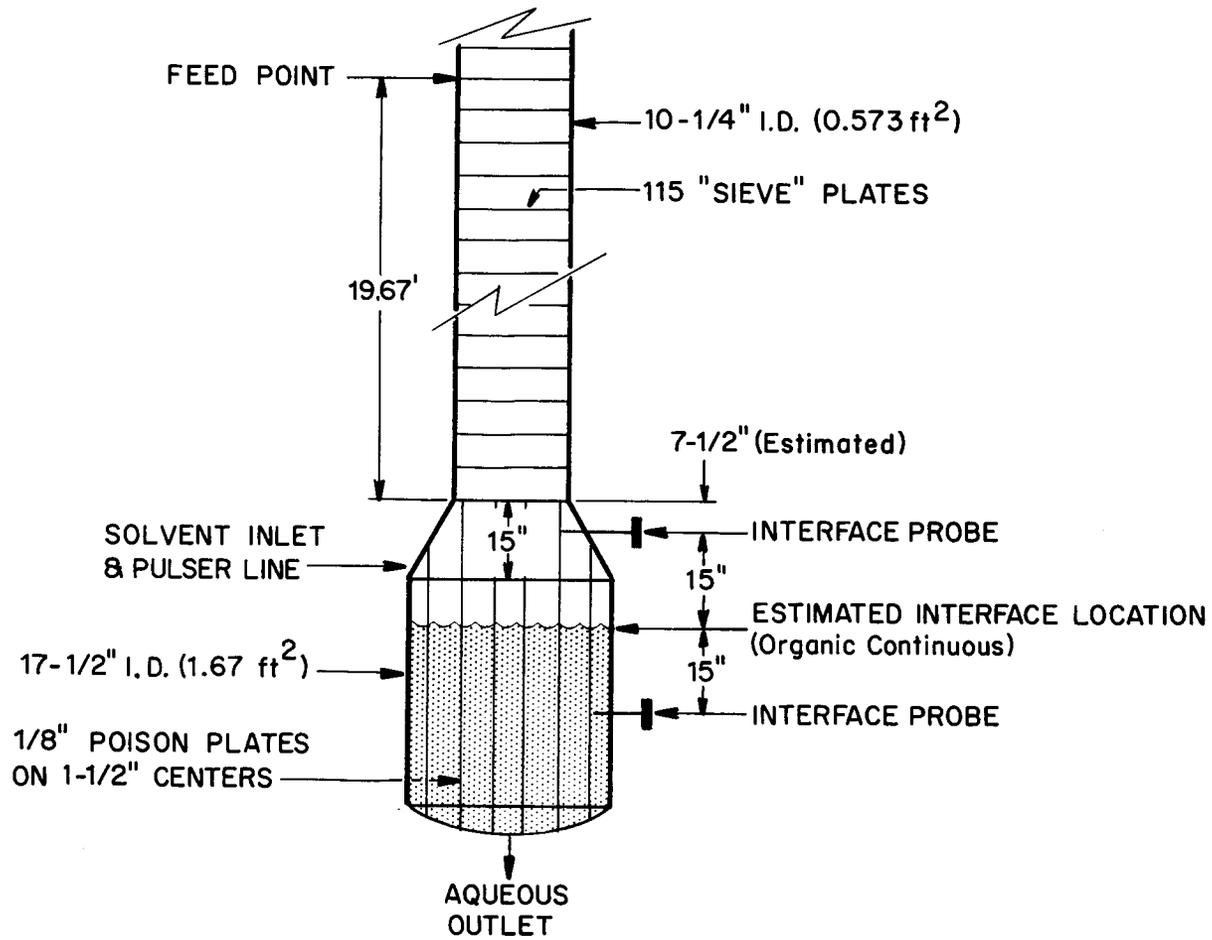


Fig. A-1: Extraction Section and Lower Disengaging Section of the Nuclear Fuel Services HA Column.

where V_c and V_d represent the velocities of the continuous and discontinuous phases respectively. Neither the flooding data for 15% TBP nor the necessary physical property data for calculating flooding for the tentative flowsheet are available. Also, the flooding correlations for the flooding data groups of interest, in which mass transfer is occurring, give a rather large deviation from experimental results. Therefore, even if the physical property data were available, the estimation of flooding rates from them would be somewhat questionable.

Assume that 20% TBP flooding data apply, although flooding rates should be somewhat higher for a 15% TBP system. From data points 1669-1678 and 1690-1703 (ref. 9), $V_c + V_d \cong 400$ ft/hr at flooding at maximum column throughput (amplitude-frequency product = 54.5 in./min) and $V_c \cong V_d$ as for the assumed flowsheet.

$$x_f = \frac{2}{3 + \sqrt{1 + 8 \frac{V_c}{V_d}}} \quad (\text{see ref. 9, p. 150}),$$

where x_f = fractional holdup of the dispersed phase (aqueous) at flooding. Substituting the values presented above for V_c and V_d , we obtain:

$$x_f = \frac{2}{3 + \sqrt{1 + 8 \left(\frac{28.68}{23.31} \right)}} = 0.318.$$

$$\bar{V}_o = \frac{V_c}{(1 - x_f)^2} + \frac{V_d}{x_f (1 - x_f)}, \quad (\text{ft/hr}) \quad (\text{see ref. 9, p. 150}),$$

where \bar{V}_o is the characteristic drop velocity for the system and has been derived from pulsed column experiments. In this case, flooding data are used to calculate \bar{V}_o , but \bar{V}_o is the same for less-than-flooding rates at the same amplitude-frequency product for the pulse generator and for the same phase flow ratio.

$$\frac{V_c}{V_d} = \frac{28.68 \text{ ft/hr}}{23.31 \text{ ft/hr}} = 1.23.$$

At flooding:

$$V_c + V_d \cong 400 \text{ ft/hr},$$

$$V_d \cong 179 \text{ ft/hr},$$

$V_c \cong 221$ ft/hr, and

$$\bar{V}_o = \frac{221}{(1 - 0.318)^2} + \frac{179}{0.318 (1 - 0.318)} = 1300 \text{ ft/hr.}$$

For the actual operating conditions:

$$V_c = 28.68,$$

$$V_d = 23.31,$$

$$\bar{V}_o = 1300 \text{ ft/hr,}$$

and $x_f = 0.0187$ by trial-and-error solution of the equation for \bar{V}_o as follows:

x_f	$(1 - x_f)$	$x_f(1 - x_f)$	$(1 - x_f)^2$	$\frac{V_c}{(1 - x_f)^2}$	$\frac{V_d}{x_f(1 - x_f)}$	\bar{V}_o
0.018	0.982	0.01768	0.9643	29.7	1318	1348
0.0185	0.9815	0.018158	0.9633	29.8	1284	1314
<u>0.0187</u>	0.9813	0.018350	0.9629	29.8	1270	<u>1300</u>

The column is operating at $\frac{(28.68 + 23.31) \text{ ft/hr}}{400 \text{ ft/hr}} \times 100 = 13\%$ of the maximum flooding rate.

$$\text{Organic holdup in cartridge} = 11.0 \text{ ft}^3 (1 - 0.0187) = 10.8 \text{ ft}^3,$$

$$\text{organic holdup time in cartridge} = \frac{10.8 \text{ ft}^3}{(397 \text{ liters/hr} + 68 \text{ liters/hr}) / (28.3 \text{ liters/ft}^3)},$$

$$= \underline{0.66 \text{ hr}},$$

organic holdup time in disengaging section = 0.13 hr.

A-3. SOLVENT IRRADIATION

The following calculations are approximate and are presented as a preliminary estimate of solvent irradiation. They should be modified when more information is available concerning radiation damage calculational methods for operating systems and the observed process effects of radiation damage.

Let us assume that all the beta energy and 40% of the gamma energy are absorbed in the 10-in.-diam column.²⁹ No correction will be made for loss of rare gases or possible previous removal of iodine. The fission product reflux and the alpha irradiation from plutonium will be neglected. Radiation damage in the scrub section resulting from fission products will be negligible, since a feed plate DF of at least 100 should be obtained.

The effective fission product energy in the aqueous phase in the extraction can be determined by using Fig. 1 and data obtained in a previous study.³

For a 150-day cooling period:

$$0.0746 \text{ kg of fuel per liter of feed} \left(\frac{290 \text{ liters of feed per hr}}{378 \text{ liters of feed} + \text{scrub per hr}} \right) (17.4 \beta \text{ watts per kg of fuel} + 0.4 \times 11.3 \gamma \text{ watts per kg of fuel})$$

$$= 1.25 \text{ watts per liter of aqueous phase.}$$

For a 30-day cooling period:

$$0.0746 \text{ kg of fuel per liter of feed} \left(\frac{290 \text{ liters of feed per hr}}{378 \text{ liters of feed} + \text{scrub per hr}} \right) (38.2 \beta \text{ watts per kg of fuel} + 0.4 \times 42.4 \gamma \text{ watts per kg of fuel})$$

$$= 3.16 \text{ watts per liter of aqueous phase.}$$

Let us assume that the organic and aqueous phases are homogeneously dispersed. The average irradiation rate of the combined phases is the product of x_f and the unit activity of the aqueous phase. This product multiplied by solvent residence time gives the solvent exposure per pass.

For a 150-day cooling period:

$$\text{solvent irradiation} = 1.25 \text{ watts per liter of aqueous phase} \times 0.0187 \text{ fraction aqueous} \\ \times 0.66 \text{ hr} \\ = 0.0154 \text{ whr per liter of organic phase per pass through the cartridge.}$$

For a 30-day cooling period:

$$\text{solvent irradiation} = 0.0389 \text{ whr per liter of organic phase.}$$

Some additional solvent exposure will occur in the lower disengaging section. Since relatively little dispersed phase is present, compared with the pulse cartridge, the effective irradiation rate of the solvent is probably lower, even though the interface area is fully active and the gamma energy should be more completely absorbed.

Let us assume the exposure rate is one-half that in the cartridge.

For a 150-day cooling period:

$$\text{solvent irradiation} = \frac{1.25}{2} \text{ watts per liter of aqueous phase} \times 0.0187 \text{ fraction}$$

$$\text{aqueous phase} \times 0.13 \text{ hr} = 0.00152 \text{ whr per liter of organic phase;}$$

$$\text{estimated total dose per pass} = 0.0154 + 0.00152 = 0.017 \text{ whr per liter of organic}$$

phase per pass through the column.

For a 30-day cooling period:

$$\text{solvent irradiation} = 0.043 \text{ whr per liter of organic phase.}$$

The calculated doses, even after a cooling period of only 30 days, are quite modest and should present no operational problems. If the 15% TBP system has a higher flooding rate than the 20% TBP data used for these calculations, the dosages will be even lower. Substantially higher total column throughputs do not change the dosage very much, provided that the same phase ratios and pulsing conditions are maintained. For example, doubling the flow rate, which is equivalent to processing 1.04 tons of fast-reactor fuel per day, increases the irradiation dose to the solvent to only 0.0172 whr per liter of organic phase; increasing the throughput to 1.4 tons/day increases it to 0.018 whr/liter. The dose increases at higher throughputs because the increase in aqueous holdup overrides the decrease in organic residence time. The net result is only an 18% increase in solvent irradiation in the pulse cartridge when the throughput is increased from 13% to 75% of flooding.

A-4. EFFECT ON SOLVENT IRRADIATION OF INCREASING THE AMPLITUDE-FREQUENCY PRODUCT OF THE PULSER ON THE HA COLUMN

The amplitude-frequency product of the pulser on the NFS column could be increased so that the column would operate nearer flooding at flowsheet rates. This would increase the aqueous holdup, decrease the solvent holdup, and increase the irradiation dose to the solvent. If it was desirable to keep solvent irradiation at the lowest possible value, this operational alternative (of increasing the amplitude-frequency product) would be considered only if losses were excessive with the existing column and it was desirable to decrease the HTU's to reduce losses at the expense of

increased solvent damage. This is probably not the case with the present column since it is tall enough to ensure low losses. The following values indicate the effect of increasing the amplitude-frequency product:

$$V_c = 28.68 \text{ ft/hr}, V_d = 23.31 \text{ ft/hr.}$$

From data point 1674 (ref. 9), $V_c = V_d = 97$ at an amplitude-frequency product of 90.9 in./min. (Even lower flooding rates could be obtained at higher amplitude-frequency products, but additional data are not available.)

The column will be operating at 27% of flooding at flowsheet rates.

Let us assume that $V_c + V_d = 194$ at flooding can also be achieved with the desired flow ratio of $\frac{V_c}{V_d} = \frac{28.68}{23.31}$; consequently, $x_f = 0.318$. Then at flooding with flowsheet flow ratios, $V_c = 107 \text{ ft/hr}$, $V_d = 87 \text{ ft/hr}$, and $\bar{V}_o = 631 \text{ ft/hr}$ for the amplitude-frequency product of 90.9 in./hr.

For the actual operating conditions of 27% of flooding, $x_f = 0.0405$,
 organic holdup in cartridge = 10.55 ft^3 ,
 organic holdup time = 0.64 hr,
 solvent irradiation in the cartridge (150-day-cooling period) = 0.032 whr per liter
 of organic phase,
 solvent irradiation in the disengaging section = 0.0033 whr per liter of organic phase.

Thus, an oversized column that is operated with a high amplitude-frequency product, so that the column approaches flooding at low liquid throughputs, increases the irradiation dose to the solvent as would be expected from the increased aqueous-phase holdup in the column.

A-5. EFFECT OF A SMALLER COLUMN ON SOLVENT DAMAGE

It is apparent that the NFS HA column extraction section is oversized ($\leq 13\%$ of flooding) for the proposed flowsheet. If a smaller column were used, x_f would increase while the organic holdup time would decrease. Consider the more normal case where the column is designed so that $V_c + V_d = 300 = 75\%$ of flooding for the amplitude-frequency product of 54.5 in./min (see Sects. 2 and 3).

Then the column cross-sectional area = 0.0995 ft^2 (4.3 in. ID),

active volume in the cartridge = 1.91 ft^3 (assuming the same column height as in the original NFS column),

$$x_f = 0.318,$$

$$\bar{V}_o = 1300 \text{ ft/hr},$$

$$V_c = 165.5 \text{ ft/hr},$$

$$V_d = 134.5 \text{ ft/hr at 75\% of flooding},$$

$$x_f = 0.147 \text{ at 75\% of flooding},$$

$$\text{organic holdup in cartridge} = 1.63 \text{ ft}^3,$$

$$\text{organic holdup time} = 0.099 \text{ hr},$$

$$\text{solvent irradiation in the cartridge (150-day cooling period)} = 0.018 \text{ whr/liter}.$$

Let us assume that the lower disengaging section is redesigned so that the solvent volume in the settler is reduced to one-half of the original inventory (2.2 ft^3). Based on the assumptions used in Sect. 3, the solvent irradiation (after 150 days of cooling) in the disengaging section would be 0.006 whr/liter . The solvent irradiation in the disengaging section in the small column was a much larger fraction of the total irradiation than it was in the large column (Sect. 3). This indicates the need for a better method for estimating irradiation in the disengager and more attention to disengager design. The overall conclusion is that a column designed closely for actual operating conditions will result in about an 18% increase in the irradiation in the cartridge; however, this level is still low. If the gamma absorption were corrected for the smaller-diameter column, the result would be an approximately 16% increase in the irradiation level.

HTU's might be lower in the small column, since they generally decrease as flooding is approached. For the small column, the extraction height probably could be decreased somewhat, assuming that the large-diameter column was adequate in this respect. This, in turn, would reduce the solvent exposure so that the solvent irradiation might actually show very little net increase calculated for a grossly oversized column operating at a small fraction of flooding.

A-6. EFFECT OF AQUEOUS CONTINUOUS OPERATION OF THE NFS COLUMN ON SOLVENT IRRADIATION

Let us assume that standard sieve plates are used, since they would be the usual choice for this operational mode. The operating conditions are: $V_c = 23.31$ ft/hr (aqueous), $V_d = 28.68$ ft/hr (organic).

No data are available⁹ for this case with 20% TBP, and the few data given for 30% TBP seem to be quite low compared with HAPO data for this system.^{19,28} From the latter two references for 30% TBP, $V_c + V_d = 2250$ gal hr⁻¹ ft⁻² = 301 ft/hr at flooding for an amplitude-frequency product = 44 in./min. The calculated results are:

$$x_f = 0.348, \bar{V}_o = 1049 \text{ from flooding data,}$$

$$x_f = 0.0373 \text{ at 17\% of flooding (operating conditions),}$$

$$\text{organic holdup} = 0.41 \text{ ft}^3,$$

$$\text{organic holdup time} = 0.025 \text{ hr,}$$

$$\text{solvent irradiation (after 150 days of cooling)} = 0.03 \text{ whr per liter per pass (total).}$$

This is about twice the calculated irradiation dose for organic-continuous operation (see Sect. 3 at about 13% of flooding). However, the flooding rate for 15% TBP is probably higher than that for 30% TBP. If we assume that it is twice as high (which is unlikely), then the above figure would reduce to around 0.02 whr/liter. Thus the irradiation level will be between 0.02 and 0.03 whr/liter.

All the preceding calculations indicate that only a slight difference exists between organic-continuous and aqueous-continuous operation from the standpoint of solvent irradiation, which is low in any case. Organic-continuous operation may be a little better in this respect and is definitely preferred for highest decontamination, higher capacity, and lower HTU's. Better flooding data and a better method for estimating the contribution from the disengaging section are needed.

APPENDIX B

COST ESTIMATE FOR ADDITION OF PLUTONIUM PURIFICATION
EQUIPMENT AND BUILDING TO EXISTING NFS PLANT FOR
PROCESSING FAST-REACTOR FUEL³⁰

Assumptions: All equipment is designed and built for remote installation and removal. Titanium is used for heater tube bundles, evaporator shells, and acid rectification towers.

Equipment, piping, instrumentation, etc.	\$1,545,000
Hot cell - with 2-ft concrete walls, six viewing windows, remote crane (5-ton capacity), and miscellaneous mechanical equipment (i.e., retrieval of remote crane if inoperable)	
Net processing volume of cell, 15,000 ft ³ ; volume of crane and equipment decontamination cell, 3750 ft ³	1,195,000
Building volume ~300,000 ft ³ (this allows ~20 ft on all sides of cell face + 30 ft over cell top + 25-ton crane)	680,000
	<hr/>
Subtotal	\$3,420,000
Architect-engineer fee (about 15% of subtotal)	\$ 510,000
	<hr/>
Total without contingency	\$3,930,000
Contingency (25% of total)	\$ 983,000
	<hr/>
Total cost	\$4,913,000

The above cost contains a sufficient allowance for packaging plutonium in powder form; this cost includes space requirements. The cost of land preparation is included. No allowance is made for carriers, shipping containers, etc. No cost is included for relocating any existing services at NFS.

REFERENCES

1. M. J. McNelley, Liquid Metal Fast Breeder Reactor Design Study [1000 Mw (electrical)] UO₂-PuO₂ Fueled Plant, GEAP-4418.
2. K. P. Cohen and G. L. O'Neill, "Safety and Economic Characteristics of a 1000 Mw (electrical) Fast Sodium-Cooled Reactor Design," pp. 185-204 in Proc. Conf. on Safety, Fuels, and Core Design in Large Fast Power Reactors, Oct. 11-14, 1965, ANL-7120.
3. E. L. Nicholson, Reference Fast Oxide Reactor Fuel for Processing in an Existing Plant, ORNL CF-66-12-25 (Dec. 15, 1966).
4. R. Salmon, Shipping Cost Calculations for Reference GE Fast Oxide Breeder, ORNL CF-67-1-20 (Jan. 12, 1967).
5. Nuclear Fuel Services, Spent Fuel Processing Plant - Safety Analysis, AEC Docket No. 50-201 (Oct. 12, 1962).
6. Nuclear Fuel Services, Inc., and New York State Development Authority Docket No. 50-201, Provisional Operating License No. CSF-1.
7. B. C. Finney, ORNL, personal communication.
8. Nuclear Safety Guide, TID-6016, Rev. 1, Table 1 (1961).
9. W. S. Groenier, R. A. McAllister, and A. D. Ryon, Flooding in Perforated-Plate Pulsed Extraction Columns: A Survey of Reported Experimental Data and Correlations, and the Presentation of New Correlations with Physical Properties, Operating Variables, and Column Geometry, ORNL-3890 (March 1966).
10. R. H. Rainey, ORNL, unpublished data.
11. J. L. Ryan and E. J. Wheelwright, "Application of Anion Exchange to the Reprocessing of Plutonium," Proc. Intern. Conf. Peaceful Uses At. Energy, 2nd, Geneva, 1958, vol. 17, P/1915 (1958).
12. E. R. Irish, Description of Purex Plant Process, HW-60116 (May 19, 1959).
13. R. E. Felt (GE, Richland, Wash.), Equipment for the Direct Calcination of Plutonium Nitrate, RL-SEP-674 (December 1965).
14. V. R. Cooper and M. T. Walling, Jr., "Aqueous Processes for Separation and Decontamination of Irradiated Fuels," Proc. Intern. Conf. Peaceful Uses At. Energy, 2nd, Geneva, 1958, vol. 17, P/2409 (1958).

15. W. T. McDuffee and O. O. Yarbrow, Thorex Pilot Plant: Short-Decay Processing Run Summary, Runs SD-1, 2, and 3, ORNL CF-58-6-1 (June 30, 1958) (classified).
16. W. Davis, Jr., Radiation Densities and TBP Radiolysis During Thorex Short-Decay Runs, ORNL-2764 (Aug. 6, 1959).
17. A. J. Huggard and B. F. Warner, "Investigations to Determine the Extent of Degradation of TBP/Odorless Kerosene Solvent in the New Separation Plant, Windscale," Nucl. Sci, Eng. 17(4), 638-50 (December 1963).
18. W. Davis, Jr., ORNL, personal communication, Nov. 1, 1966.
19. R. G. Gier, "Improved Pulsed Extraction Column Cartridges," Proc. Intern. Conf. Peaceful Uses At. Energy, 2nd, Geneva, 1958, vol. 17, P/515 (1958).
20. M. E. Whatley and W. M. Woods, The Performance of an Advanced Experimental Stacked-Clone Contactor: A High-Performance Solvent Extraction Machine with Potential for Application to Very Highly Radioactive Solutions, ORNL-3533 (April 1964).
21. M. E. Whatley et al., Unit Operations Section Quarterly Progress Report, October-December 1965, ORNL-3958 (July 1966).
22. W. S. Groenier, ORNL, personal communication.
23. R. H. Rainey, ORNL, personal communication.
24. R. E. Adams et al., Removal of Radioactive Methyl Iodide from Steam-Air Systems, ORNL-4040 (January 1967).
25. E. L. Nicholson, L. M. Ferris, and J. T. Roberts, Burn-Leach Processes for Graphite-Base Reactor Fuels Containing Carbon-Coated Carbide or Oxide Particles, ORNL-TM-1096 (Apr. 2, 1965).
26. Chemical Processing Plant, Hearing Before the Joint Committee on Atomic Energy, Congress of the United States, May 14, 1963, U.S. Govt. Printing Office, Washington, D. C., 1964.
27. M. Levenson, V. G. Trice, Jr., and W. J. Mechem, Comparative Cost Study of the Processing of Oxide, Carbide, and Metal Fast-Breeder-Reactor Fuels by Aqueous, Volatility, and Pyrochemical Methods, ANL-7137 (May 1966).
28. Purex Technical Manual, HW-31000 (classified).
29. E. D. Arnold, ORNL, personal communication.
30. W. G. Stockdale, ORNL, personal communication.

DISTRIBUTION

1. C. A. Blake
2. R. E. Blanco
3. R. E. Brooksbank
4. K. B. Brown
5. Leslie Burris, Jr. (ANL)
6. D. J. Crouse
7. F. L. Culler, Jr.
8. W. Davis, Jr.
9. D. E. Ferguson
10. L. M. Ferris
11. B. C. Finney
12. H. E. Goeller
13. W. S. Groenier
14. F. J. Hurst
15. W. H. McVey (AEC, Wash.)
16. E. L. Nicholson
17. R. H. Rainey
18. J. T. Roberts
19. Orville Roth (AEC, Wash.)
20. A. D. Ryon
21. R. Salmon
22. Harry Schneider (AEC, Wash.)
23. W. G. Stockdale
24. W. E. Unger
25. V. C. A. Vaughen
26. R. C. Vogel (ANL)
27. C. D. Watson
28. M. E. Whatley
29. R. G. Wymer
- 30-31. Central Research Library
32. Document Reference Section
- 33-37. Laboratory Records
38. Laboratory Records, RC
39. ORNL Patent Office
- 40-54. DTIE
55. Research and Development Div., ORO