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REPROCESSING METHODS AND COSTS FOR SELECTED THORIUM-BEARING REACTOR FUEL TYPES*

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ABSTRACT

Head-end and solvent extraction processes are described for two classes of thorium-bearing power reactor fuels: (1) massive graphite elements containing pyrolytic-carbon-coated thorium-uranium oxide or carbide particles, and (2) metal-clad elements containing thorium-uranium metal, oxide or carbide cores. Burn-leach, declad-dissolve and shear-leach head-end methods are described in detail. Solvent extraction flowsheets for recovering both the thorium and uranium or the uranium only are presented. The technical and economic aspects of these processes, including the interim and ultimate waste disposal problems, are discussed, by comparison with processes for standard uranium fuels.

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1. INTRODUCTION AND SUMMARY

This paper reviews U. S. developments in the field of chemical reprocessing of thorium-bearing spent fuels from nuclear reactors. In order to limit the length of the paper, consideration is restricted to fuels of the most promising reactor types in the U. S. advanced converter reactor program. Emphasis is placed on the differences in reprocessing methods and costs between these thorium fuels and standard low-enrichment uranium metal or oxide fuels clad in aluminum, zirconium, or stainless-steel alloys. The relationship between reprocessing and the rest of the fuel cycle for the most important proposed thorium-uranium recycle schemes is discussed. Head-end processes, starting with irradiated fuel elements and ending with a nitrate solution suitable for feed to solvent extraction, are described for the various fuel types. Solvent extraction flowsheets for separating the uranium and thorium from each other and from the fission products are evaluated. The interim and ultimate waste disposal problems associated with the various recycle schemes, head-end processes and solvent extraction flowsheets are presented. Finally, comments are made on the costs of reprocessing thorium fuels, by comparison with standard uranium fuels and with respect to the economic problems associated with sizing and starting a reprocessing plant when the fuel load is small initially but promises to increase substantially with time.

Primary consideration is given to (1) pyrolytic-carbon-coated thorium-uranium oxide or carbide microspheres contained in massive graphite fuel elements, i.e., fuels for "Target" type high-temperature gas-cooled reactors (HTGR); and to (2) thorium-uranium metal, oxide or carbide clad with zirconium or aluminum alloys, which includes fuels for heavy-water-cooled-and-moderated reactors (HWR), heavy-water-moderated organic-cooled reactors (HWOCR), and also spectral-shift-control reactors (SSCR) and light-water-cooled-and-moderated seed-blanket reactors (SBR or LPR). For comparison purposes, standard fuel reprocessing methods and costs are assumed to be those typical of Hanford, Savannah River, and the Nuclear Fuel Services plant. The thorium fuels all are non-standard in at least some of the following aspects:

(1) Graphite, carbon and carbide type fuels may require pre-treatment steps, such as burning or pyrohydrolysis or grinding, not presently provided in industrial-scale reprocessing plants.

(2) Thorium fuels in general require more powerful dissolvents, e.g., more concentrated nitric acid and the presence of fluoride as a catalyst, than uranium fuels to get acceptable dissolution rates, and even so may dissolve at significantly lower rates than similar uranium fuels. The use of these dissolvents may complicate the succeeding feed adjustment, solvent extraction and waste disposal steps as a result of enhanced corrosion and dissolution of part or all or the cladding material along with the thorium.

(3) Thorium fuel dissolver solutions usually require a feed adjustment step to remove excess acid, perhaps all the way to an acid-deficient condition, (and perhaps also to decompose organic compounds in the case of carbide fuels after low-temperature hydrolysis or leaching) prior to the solvent extraction step.

(4) In solvent extraction, the phase equilibria and the contacting problems associated with third-phase formation are such that thorium fuels typically must be processed through a given size of equipment at a lower rate than uranium fuels.

(5) Thorium fuels as a class tend to have higher economic-optimum fissile enrichments and burnups than uranium fuels, and this may lead to throughput-rate limitations or other processing restrictions, because of criticality control or permissible fission product off-gas release, etc.

(6) The fact that ^{233}U production involves the relatively slow decay of 27-day ^{233}Pa and the growth of the gamma-active daughters of ^{232}U and ^{228}Th into the recovered products complicates the pre- and post-processing handling of thorium fuels by comparison with uranium fuels.

(7) If thorium is recovered, the ^{228}Th and ^{234}Th in it will cause it to have a lower market value than virgin thorium, because of the extra fabrication costs involved, and it may even have a negative value in the sense that the cheapest thorium recycle scheme might include 10- to 15-year storage of the recovered thorium to provide for decay of the ^{228}Th.

(8) If thorium is not recovered and/or if aluminum nitrate is used as a salting agent in solvent extraction, the high-level fission product waste will have to be stored in larger volume in the acid condition in stainless-steel tanks, which is more expensive than the highly concentrated, neutralized storage in mild-steel tanks of wastes from standard uranium fuels.

Some of these differences can involve an increased cost of a factor of two or more on a weight basis, or may even prevent the processing of certain fuels, in a plant designed for standard uranium fuels. Because of their higher burnup and/or thermal efficiency, some thorium fuels can stand the increased cost-per-unit-weight without incurring much of a cost-per-kilowatt-hour penalty; but for other fuels, especially those which cannot be processed in existing plants, the disadvantage is quite serious.

On the other hand, design and cost studies reviewed in this paper show that reprocessing plants built specifically to handle thorium-bearing

fuels, using processes described in this paper, could process these fuels at only slightly higher cost per unit weight than standard uranium fuels, hence permitting the thorium-fueled advanced converter reactors to take almost full advantage of their higher burnup and/or thermal efficiency in achieving lower power costs. Thus, the fuel processing problems of thorium-fueled reactors are viewed as temporary "start-up" problems, perhaps involving some initial economic disadvantages for the early thorium-fueled advanced converters, but which will fade away as the industry reaches a reasonable size.

2. HEAD-END PROCESSES

2.1 Burn-Leach Processes for HTGR Fuels

This discussion will be limited to processing methods for HTGR fuels that contain pyrolytic carbon-coated thorium-and/or-uranium carbide or oxide fuel particles in massive graphite elements. A promising processing method consists of burning the fuel in a fluidized bed of inert alumina and then leaching with fluoride-catalyzed nitric acid (Thorex reagent) to recover the uranium and thorium.[1] Laboratory- and engineering-scale studies of this process have been made with unirradiated prototype fuel specimens, and a few hot-cell experiments have been carried out with irradiated material.

The burn-leach process for graphite-base fuels is shown schematically in Fig. 1. Initially, the fuel is chopped or crushed to a convenient size for handling and fed to a fluidized-bed burner where it is burned at 700 to 750°C in a fluidized bed of granular alumina. Burning is started by injecting preheated oxygen into the fluidized bed and simultaneously heating the bed by external heaters. When the fuel starts to burn, the heaters are turned off, and the heat of reaction is removed by air-cooling the bed. For efficiency, continuous operation, with feeding of fuel, fresh alumina, and oxygen to the burner, and withdrawal of ash, all at the proper rates, is preferred. Under normal operation, nearly quantitative consumption of the oxygen is achieved, resulting in an off-gas composed mainly of CO_2 with less than 5% of carbon monoxide in the off-gas. Particles in the off-gas are removed mostly by filters, and a gas-cleanup system prevents the release of all radioactivity except the noble gases. After burning, the product bed is transferred to a leacher where the uranium and thorium are dissolved. The alumina may be recycled or discharged to waste. Uranium and thorium recoveries should be greater than 99.5%.

Design of the burner and the leaching system may be dependent on the type of fuel being burned and whether it is desirable to prevent isotopic mixing of the high ^{236}U content material remaining in the fueled particle and the freshly bred ^{233}U in the thorium particle. Burning of fuels containing carbon-coated Th-U dicarbide particles converts the carbides to finely powdered oxides, dispersed homogeneously throughout the bed. Consequently, to recover the uranium and thorium, the entire bed must be leached. However, oxide fuel particles of high ThO_2 content may not be affected during combustion in a fluidized bed and probably could be separated from the alumina if desired before the leaching operation. This leads to the possibility of preventing isotopic mixing by a physical separation or by a selective chemical dissolution to separate the ThO_2 particles containing the bred ^{233}U from the U_3O_8 derived from either uranium carbide or uranium oxide fuel particles.

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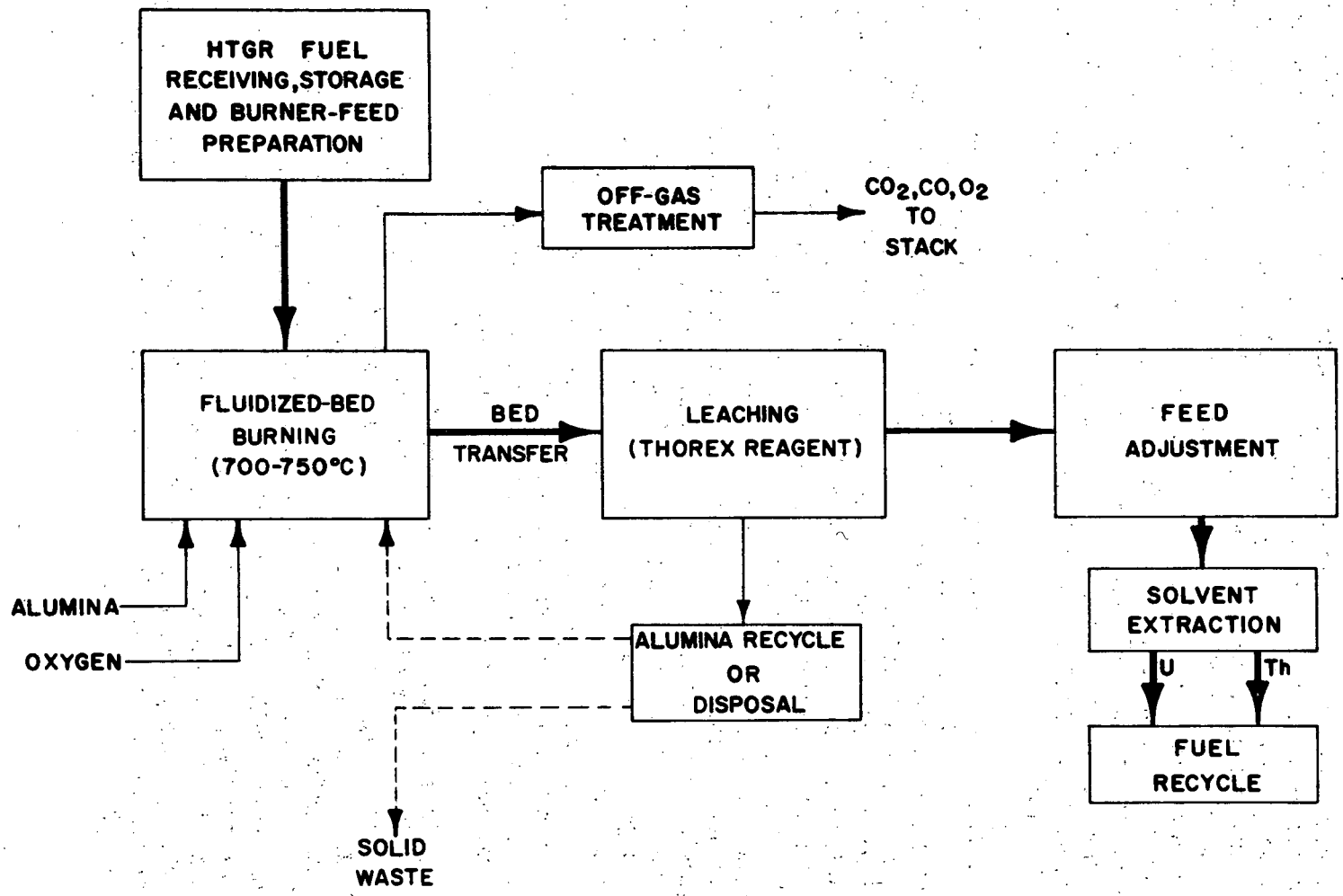

FIG. 1. POTENTIAL BURN-LEACH PROCESS FOR GRAPHITE BASE REACTOR FUEL.

2.1.1 Fluidized-Bed Combustion of Graphite-Base Fuel

2.1.1.1 Fuels Containing Carbon-Coated Carbide Particles. Tests of the combustion of fuel containing carbon-coated carbide fuel particles were conducted in 2-in.-diam and 4-in.-diam fluidized beds.[2,3] The 4-in.-diam fluidized bed (Fig. 2) used at Oak Ridge National Laboratory for pilot plant studies had the essential features of a plant-size burner. The unit was made of nickel 201, although type 310 stainless steel would probably be preferred for a large-scale burner because of the better high-temperature mechanical properties of this material. The fluidized bed of alumina was an efficient heat-transfer medium and it diluted and suspended the fuel oxides formed during combustion so that the product was a free-flowing powder. The combustion was easily controlled and good operation, with loading of the alumina up to 30 wt % U-Th oxides, was achieved when the starting bed was composed of equal parts of 60- and 90-mesh fused alumina. Attrition of the alumina was negligible in a one-week test.[3]

In a typical experiment, chopped or crushed fuel was added to approximately 20 kg of alumina, and the bed was heated to the ignition temperature of 650°C. Bed centerline temperatures and wall temperatures were held at about 750 and 700°C, respectively, by air-cooling the finned exterior of the fluidized bed. The CO_2 and CO contents of the off-gas were continuously monitored and were relatively constant when there was an excess of carbon in the burner. A decrease in the CO_2 and CO contents showed that the carbon inventory in the bed was being depleted and more fuel was added as needed to maintain the desired oxidation rate. Alumina was added periodically when product was continuously withdrawn. Any small particles of carbon entrained in the alumina below the grid were rapidly burned in the hot oxygen, and it was possible to continuously withdraw a product stream containing less than 0.1% carbon and 30 wt % U-Th oxides from the bottom of the bed. Toward the end of a combustion run, when the carbon concentration in the bed was low, it was necessary to supply heat to the burner to ensure combustion of the last traces of carbon. The superficial gas velocity in the bed was about 0.76 ft/sec at the bed mid-point pressure of 17.6 psia and average temperature of 725°C.

Continuous oxidation rates varying from 1.1 to 1.4 kg of carbon per hour were obtained in pilot plant tests with a 4-in.-diam fluidized bed by varying the oxygen flow rate over the range of 1.3 to 1.6 scfm. Oxygen utilization decreased from 97 to 90% as the flow rate was increased. The heat transfer coefficient from bed to wall was estimated at 85 Btu hr^{-1} ft^{-2} °F and temperatures could be easily controlled by air cooling the finned outer wall. Plugging of the filters was not a problem, and the filter blowback system was not used during routine operations. Micropore filtration[3,4] of the off-gas showed that practically no particles escaped through the primary sintered-metal filters. The typical off-gas consisted of about 90% CO_2, 5% CO, and 5% O_2. Corrosion of the burner was negligible.[2]

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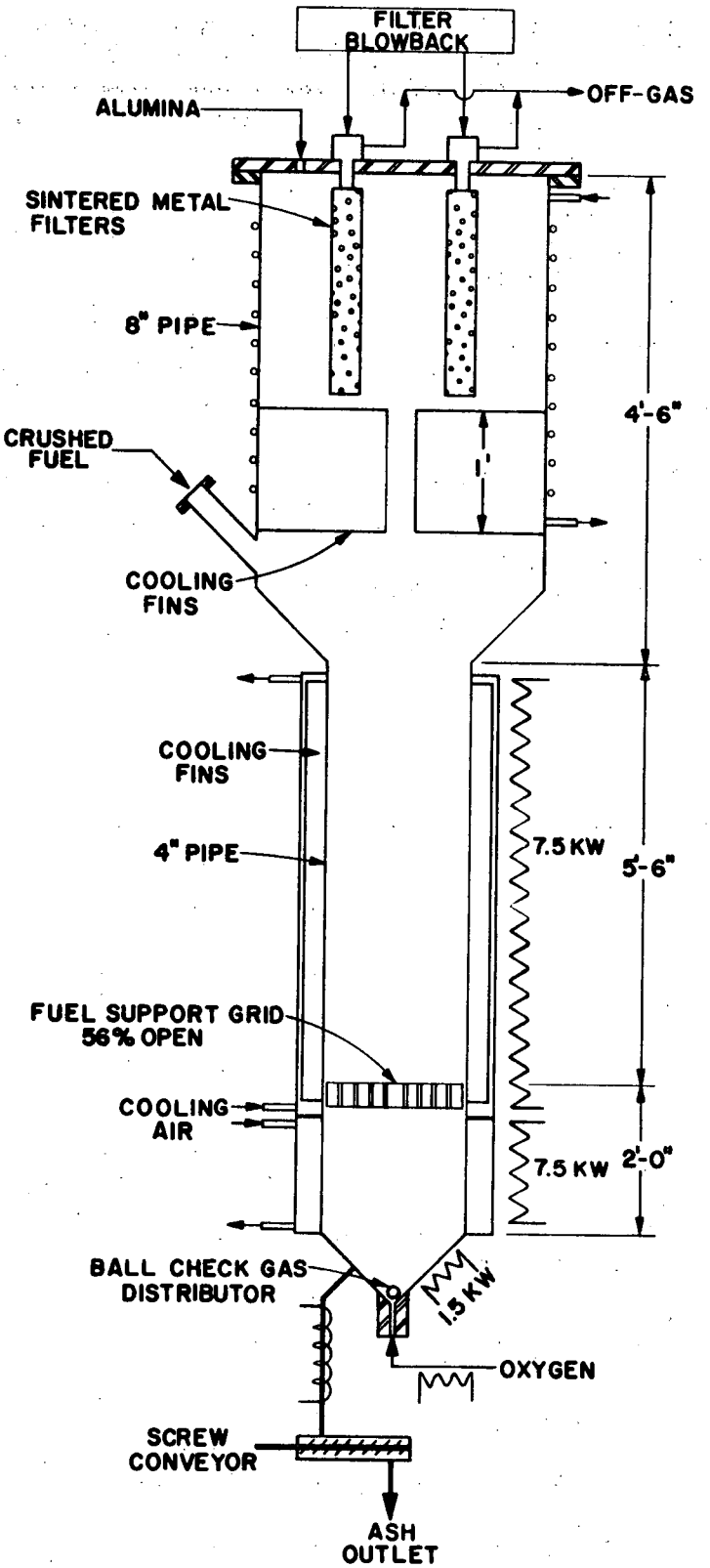


FIG. 2. FOUR-INCH-DIAMETER FLUIDIZED-BED BURNER FOR PILOT PLANT STUDIES.

2.1.1.2 Fuels Containing Carbon-Coated Oxide Particles. To date, no fuel containing carbon-coated ThO_2 and UO_2 or mixed ThO_2-UO_2 microspheres has been burned in a fluidized bed. A potential problem with this fuel arises from the uncertainties concerning the integrity of the ThO_2 or mixed ThO_2-UO_2 microspheres after irradiation to projected burnups of 50,000 to 80,000 Mwd/MT. If the ThO_2 or mixed ThO_2-UO_2 microspheres retain their shape, it might be possible to separate them from most of the alumina after burning the carbon. This might result in a simpler leaching system in that only the fuel particles would be fed to the leacher. For example, preliminary tests have shown that ThO_2-UO_2 particles containing up to 75% ThO_2 are unaffected by oxygen at 800°C while pure UO_2 is readily oxidized to U_3O_8. However, if the particles are broken during irradiation or combustion, they will be dispersed throughout the alumina with the U_3O_8 derived from oxidation of the UO_2 seed particles; thus all the alumina must be leached.

2.1.1.3 Fuel and Reprocessing Systems to Permit Segregation of ^{236}U In the TARGET concept[5] for the HTGR, the uranium fuel particles after irradiation contain a relatively large amount of ^{236}U, the recycle of which is undesirable if high conversion ratios are to be achieved. Some possibilities for the design of the fuel and the reprocessing system to permit the ^{236}U to be withdrawn from the fuel cycle are given in the following table, which assumes fluidized-bed oxidation of the fuel as the first step:

Fuel System		Segregation of the U^{236} might be achieved by:
Fuel Particle	Fertile Particle	
UO_2	ThO_2	(1) Leaching the ash with dilute nitric acid to remove the $^{235,6}U_3O_8$ before dissolving the ThO_2-$^{233}UO_2$ particle in Thorex reagent.
UC_2	ThO_2	
		or (2) Physical separation of the ThO_2-$^{233}UO_2$ from the alumina containing the $^{235,6}U_3O_8$.

This table oversimplifies the problem, since there are many possibilities for including refractory materials (such as ZrO_2, ZrC, SiC, BeO, etc.) in the fuel or fertile particle or as a coating for the same. Obviously, this would complicate the reprocessing of these fuels.

2.1.1.4 Fission Product Behavior During Combustion. The behavior of the fission products was not studied during actual fluidized-bed combustion but was examined cursorily in laboratory-scale tube-furnace experiments. In one series of experiments,[6] in which a large excess of oxygen was used to burn prototype Peach Bottom fuel irradiated to about 10,000 Mwd/MT, up to 35% of the cesium and 96% of the ruthenium were volatilized from the high-temperature zone during 6-hr combustions at 800°C. Experiments[7] in the same equipment with a slightly irradiated

fuel at 700°C showed that up to 1.1% of the cesium and 65% of the ruthenium were volatilized in 6 hr, thus illustrating the desirability of low combustion temperatures. In each case, practically all the fission products were trapped in the cool end of the reaction tube and nearly all remaining activity was removed by filtering the off-gas through a sintered metal filter. The overall decontamination factor was greater than 10^4 in all experiments. In other studies[8] only a small amount of cesium and ruthenium were volatilized from the hot zone when the fuel was burned in a deficiency of oxygen at 800°C.

Supplemental treatment of the off-gas may be required. An attractive method might be to mix steam with the off-gas, condense the vapor, and then filter through absolute filters. Waste-calcination work[8,10] indicated that a system combining sintered-metal filters, condensation of vapor, and finally absolute filtration can yield decontamination factors $\geq 10^8$ for the off-gas. If rare gas retention ever becomes necessary for the reprocessing plant, the large amount of CO_2 in the off-gas may be a serious problem for the rare gas retention system.

2.1.2 Leaching of Fluidized-Bed Products

2.1.2.1 Products from Fuels that Contain Carbon-Coated Carbide Particles. An efficient bench-scale batch leacher was devised[3] in which the leaching acid was recirculated upflow at a low rate through the bed, fluidizing the bed for better contact of the solids and the acid. After leaching, the product solution was drained from the bed, and the bed was washed with water. The bed material for these studies was produced by burning unirradiated Peach Bottom fuel compacts (carbon-coated Th-U dicarbide particles dispersed in a graphite matrix) in a fluidized bed of Norton RR alumina at 700 to 750°C. In laboratory-scale 5-hr leaches, more than 99.5% of the uranium and thorium were recovered when the HNO_3 concentration was 4 M or higher, and when the HF concentration was 0.02 to 0.05 M. Uranium and thorium recoveries were inadequate with 13 M HNO_3 and with 2 M HNO_3 - 0.05 M HF. Less than 2% of the alumina was dissolved.

2.1.2.2 Products from Fuels Containing Carbon-Coated Oxide Particles. We have not yet done experiments with fuels containing carbon-coated mixed ThO_2-UO_2 particles. Leaching of uranium and thorium may simply involve dissolution of ThO_2-UO_2 microspheres in the presence of a small amount of alumina if the microspheres are physically separated from the alumina after burning. Laboratory tests showed that unirradiated ThO_2-UO_2 microspheres probably cannot be dissolved readily in a dilute Thorex solution; however, dense 300- to 600-μ-diam ThO_2 microspheres were dissolved in 3 to 6 hr in boiling 13 M HNO_3-0.05 M HF, even in the presence of a large excess of alumina. In other studies,[11-13] irradiated ThO_2-UO_2 pellets appeared to dissolve faster than unirradiated oxide, and a 6-hr dissolution period is estimated to be adequate.

2.1.3 Conceptual Design for a Large HTGR Fluidized-Bed Burner and Leacher System

A conceptual design was prepared[1] for a head-end reprocessing facility to permit reprocessing HTGR fuel at a multipurpose reprocessing plant (e.g., the Nuclear Fuel Services, Inc. plant). The conceptual design of a large burner is shown in Fig. 3; it is a scaleup of the pilot plant burner and would be operated similarly. Provisions for removing heat from central portions of large fluidized beds must be made to avoid excessive centerline temperatures. It was assumed that the fuel contained only mixed ThO_2-UO_2 microspheres, that the microspheres would not be broken, and that a classifying operation after burning would permit leaching the microspheres in the presence of very little alumina. The conceptual design of the leaching and feed adjustment system is shown in Fig. 4. For criticality control, use of two geometrically safe slab leachers in series is envisaged for dissolving practically all the fuel particles before the solution and alumina slurry flow into large-diameter feed adjustment vessels. The leachers would be equipped with thermosiphon heating and solids pumping loops and would operate continuously in series. Leachant would be pumped into the first slab-shaped tank and maintained at its boiling point throughout the leaching system. Solids and solution from the first leacher would overflow continuously into the second one. Alumina would be transported through the system without being attacked appreciably by the dissolvent. Solution from the second leacher would be transferred to a feed adjustment system where any small fuel particles still remaining in the leacher overflow would be dissolved rapidly.

A conceptual design of a complete HTGR head-end facility is shown in Fig. 5. It provides for fuel element receipt and storage, crushing, burning, leaching, and feed adjustment for up to 40 elements per day and up to 225 days per year in two parallel processing lines. The elements were assumed to be 4.5-in.-diam., 20-ft long graphite logs, each containing 107 kg of carbon plus 10.9 kg of thorium plus uranium plus fission products. A total capital investment of about $9 million was estimated for construction and start-up of this facility:

Building, Cells and Services		$2,787,000
Process Equipment		1,098,000
Process Piping		906,000
Process and Radiation Instrumentation		350,000
Site Improvements and Utilities		481,000
	Subtotal	$5,622,000
Design and Contingency		2,811,000
Interest, Working Capital		607,000
	TOTAL	$9,040,000

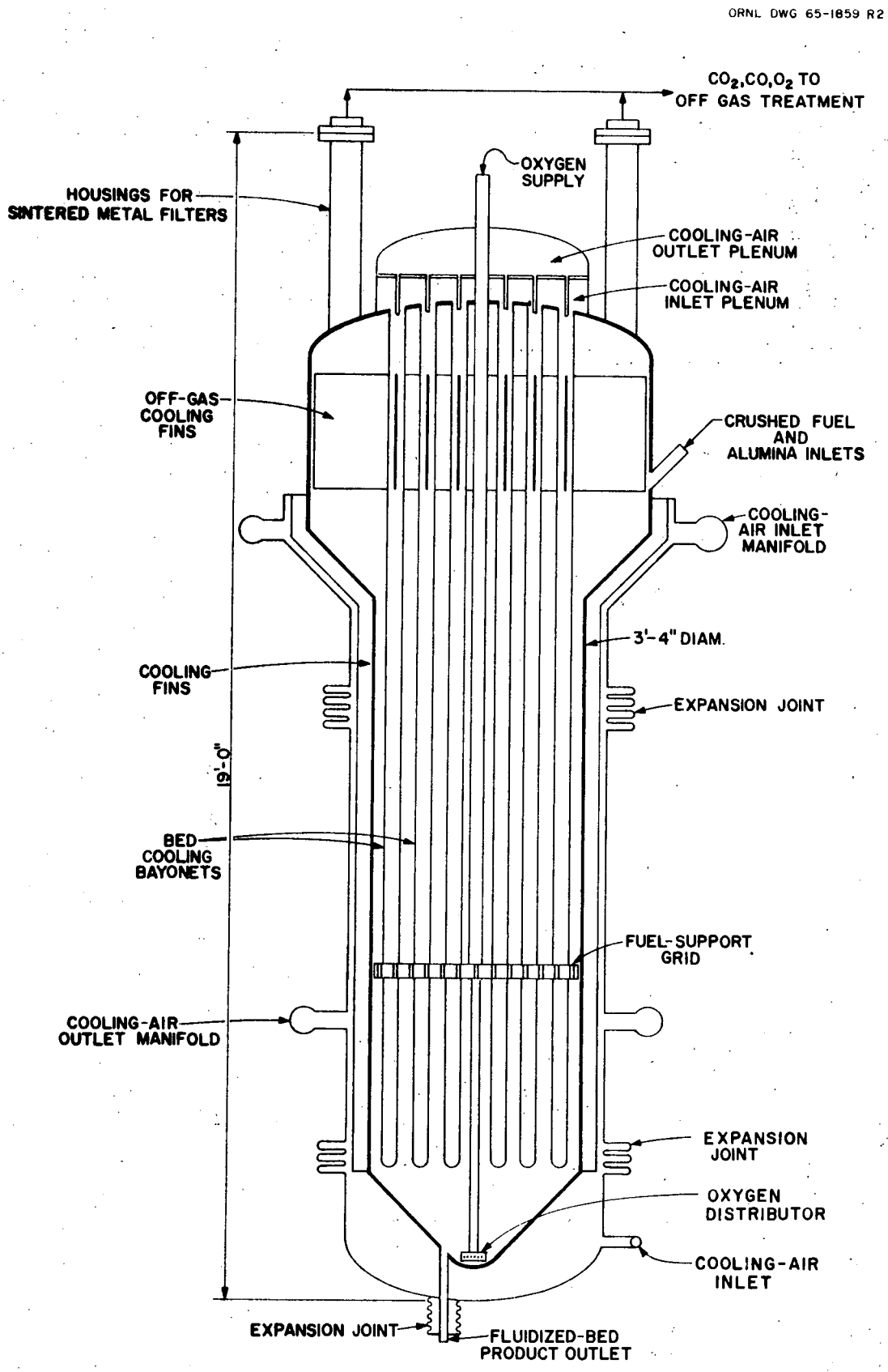


FIG. 3. CONCEPTUAL DESIGN FOR A LARGE, FLUIDIZED-BED BURNER FOR GRAPHITE-BASE FUELS.

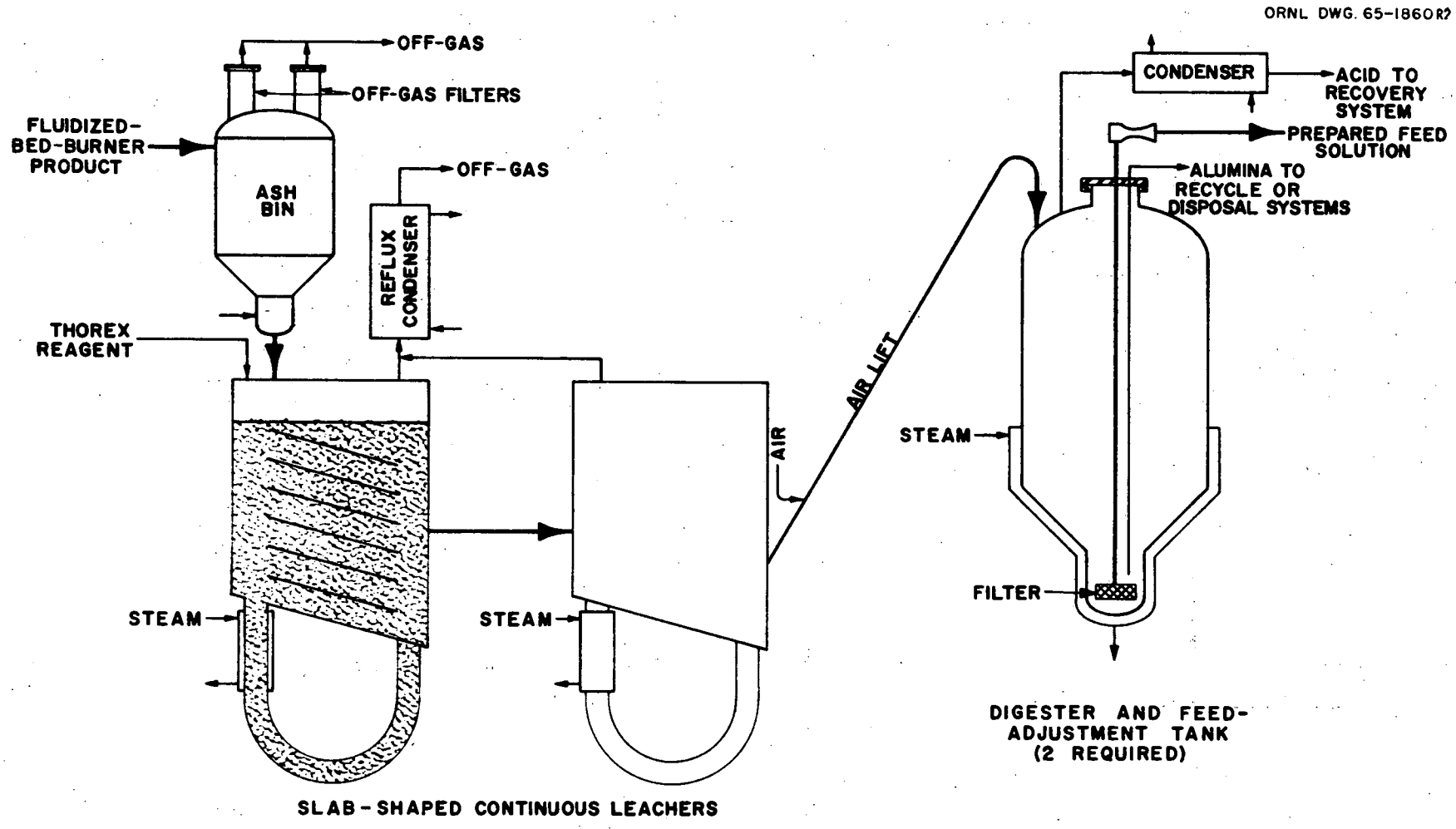


FIG. 4. CONCEPTUAL LEACHING AND FEED-ADJUSTMENT SYSTEM FOR GRAPHITE-BASE FUELS THAT CONTAIN ThO_2-UO_2 MICROSPHERES.

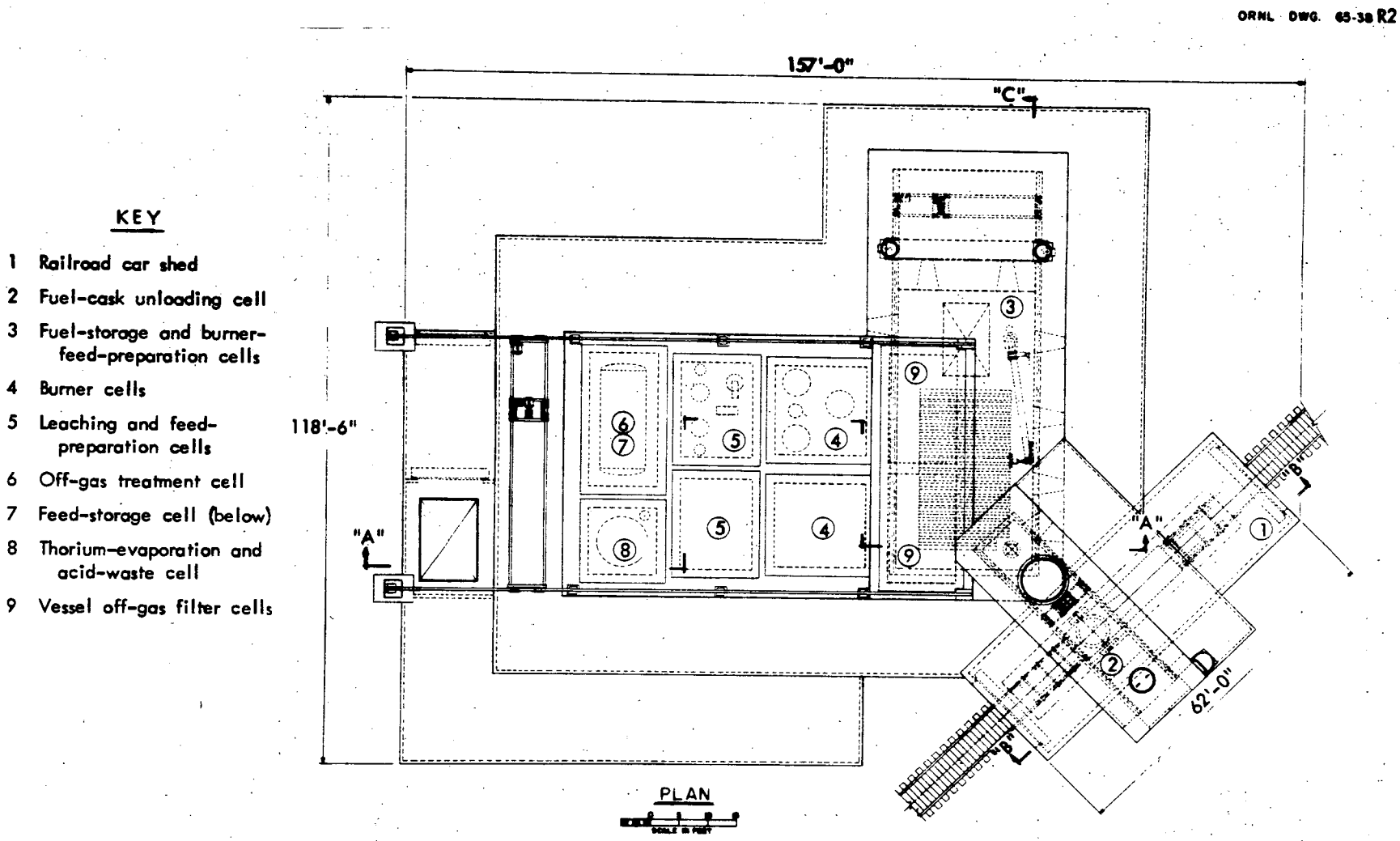


FIG. 5. FUEL-REPROCESSING HEAD-END BUILDING FOR HTGR FUEL.

2.2 Chemical Decladding and Dissolution Processes

In this section we will consider primarily thorium-uranium oxide, metal and carbide fuels clad in zirconium or aluminum alloys, with only limited discussion of other fuel types such as those clad in stainless steel, since they are of less interest in advanced converter reactors.

2.2.1 Zirconium- or Zircaloy-Clad Fuels

Zirconium claddings can be separated chemically from core materials by either aqueous or non-aqueous methods. The aqueous method (Zirflex process) involves dissolution of the cladding in ammonium fluoride-ammonium nitrate solutions.[14-17] The non-aqueous techniques are oxidative disintegration in $HF-O_2$ mixtures,[18-20] the Thermox process,[21] and hydrochlorination (Zircex process).[22] After decladding, the core materials are dissolved in appropriate reagents in preparation for solvent extraction recovery of the uranium and thorium. Each of these methods will be discussed briefly below.

2.2.1.1 Zirflex Decladding.[14-17] Zirconium or Zircaloy claddings are readily dissolved in boiling 4 to 6 M NH_4F--0.5 to 1 M NH_4NO_3. The overall reaction is approximated by the equation:

$$Zr + 6NH_4F + 0.5\ NH_4NO_3 \rightarrow (NH_4)_2ZrF_6 + 5NH_3 + 1.5H_2O.$$

Actually, about 0.1 mole of hydrogen is also evolved per mole of zirconium dissolved. In the absence of ammonium nitrate a gas having the composition 33% H_2--67% NH_3 is liberated. The average dissolution rate of Zircaloy-2 that has been exposed to high-temperature air or water is about 5 mg min^{-1} cm^{-2}; thus, a 30-mil-thick cladding is penetrated in 2 to 3 hr. Since the solubility of $(NH_4)_2ZrF_6$ at 25°C decreases markedly with increasing excess NH_4F concentration, the most concentrated decladding waste solutions are obtained by consuming as much of the ammonium fluoride as possible. To optimize both dissolution rate and volume of waste solution, the decladding is conducted with an overall F/Zr atom ratio of about 7, making the maximum attainable zirconium concentration in the waste solution about 0.6 M. Pilot plant studies with unirradiated fuel[17] showed that the ammonia must be removed from the dissolver continuously to prevent an increase in pH of the solution and subsequent precipitation of zirconium oxide, which causes a marked reduction in the rate of reaction. Ammonia removal was effected by using a high boilup rate and a high temperature in the downdraft condenser or by steam sparging at a rate that maintained a constant liquid level in the dissolver. Suitable materials of construction for the dissolver were stainless steel or Nionel. The decladding solution probably will require centrifugation to recover core material fines prior to discharge to the waste system.

Preliminary experiments[23] with unirradiated $ThO_2-UO_{2.7}$ fuel pellets (4.2% $UO_{2.7}$, 83% of theoretical density) indicated that zirconium claddings could be dissolved in 6 M NH_4F--1 M NH_4NO_3 with attendant

uranium and thorium losses of less than 0.4%. Even lower losses are expected with higher density ThO_2-UO_2 fuel. No studies of the decladding of irradiated Zr-clad ThO_2-UO_2 have yet been made; however, soluble uranium and plutonium losses in the decladding of Zircaloy-clad UO_2 irradiated up to 17,700 Mwd/MT averaged only about 0.05%.[24] Thus, Zirflex decladding of thorium oxide core fuels appears feasible.

No data appear to exist on the behavior of Th-U alloy in ammonium fluoride solutions. Uranium metal is attacked[15] at a rate of about 0.05 g hr^{-1} cm^{-2} in boiling 6 M NH_4F with the rate increasing about a factor of 2 when the NH_4NO_3 concentration of the solution is 0.5 to 1 M. If Th-U alloy were attacked at a comparable rate, a significant fraction of the alloy would be converted to insoluble ThF_4 and UF_4. Consequently, the practicability of Zirflex decladding for Th-U alloy fuel is questionable at this time.

Reaction of ThC-UC with ammonium fluoride solutions has not been investigated. If the irradiated carbide were not passive (irradiated UC is passive in boiling water and 6 M NaOH[25]) hydrolysis with NH_4F would be expected to yield solid ThF_4-UF_4 and a gas composed mainly of methane and hydrogen.[26,27] This formation of an insoluble fluoride would be highly undesirable.

2.2.1.2 Non-Aqueous Decladding.[18-20] Treatment of Zr-clad fuels with HF-O_2, with steam, or with HCl in fluidized beds of inert alumina, such as Norton RR and Alcoa T-61 grades, results in a product from which uranium and thorium can be recovered by acid leaching. Reaction of zirconium or Zircaloy with a gaseous mixture of HF and oxygen results in conversion of zirconium to ZrO_2 and simultaneous disintegration of the cladding. Optimum conditions appear to be about 625°C and a gas mixture containing 20 to 40% HF.[18] The penetration rate is at a maximum of about 40 mils/hr with 40% HF--60% O_2 at 625°C. The product bed can contain a total of up to about 40 wt% ZrO_2 plus core oxides such as U_3O_8 and ThO_2, and always contains 3 to 8% fluorine. This amount of fluorine is very high relative to the amount of uranium (F/U atom ratio in the bed usually is 1 to 3). Thus, removal of the fluorine by pyrohydrolysis prior to leaching is recommended. In laboratory-scale experiments, greater than 90% of the fluorine was removed from typical product beds in 4-hr reactions with steam (1 atm pressure) at 600°C. During HF-O_2 decladding, UO_2 cores are converted primarily to U_3O_8 powder which is dispersed throughout the bed. The behavior of ThO_2-UO_2, Th-U, and ThC-UC in this system has not yet been tested. It is expected, however, that both the alloy and carbide would react, although perhaps slowly. Mixed oxide of high ThO_2 content would be expected to be relatively inert.

A similar technique for converting zirconium claddings to ZrO_2 powder involves reaction of the cladding with oxygen-water vapor mixtures at about 825°C using nitrogen as a catalyst.[21] This "Thermox" decladding of Zr-clad UO_2, followed by oxidation of the UO_2 to U_3O_8, and leaching of the product with nitric acid gave uranium recoveries of greater than 99%. The Thermox method has not been tested with Th-U alloy or ThO_2-UO_2

fuels. Of the core materials being considered, only the carbide would be expected to react rapidly with the decladding reagent.

An alternative non-aqueous decladding route is removal of the zirconium cladding as volatile $ZrCl_4$ by reaction with HCl at about 500°C.[22] The $ZrCl_4$ would be converted to ZrO_2 in a separate fluidized bed by reaction with steam allowing disposal of the cladding as a solid waste. The application of this "Zircex" process to thorium-bearing fuels has not yet been studied. Again, only the carbide core materials would be expected to react with the HCl during decladding. The oxide fuels are practically inert.

2.2.1.3 Core Dissolution. After decladding, the core materials would be dissolved to produce solutions suitable as feeds for a solvent extraction recovery system. Thorium metal, thorium oxide, and ThO_2-UO_2 mixtures dissolve in nitric acid containing small amounts of hydrofluoric acid as catalyst (fluoride-catalyzed nitric acid).[25,28] Dissolution of unirradiated ThO_2-UO_2 in the optimum reagent, 13 M HNO_3--0.05 M HF, is slow, with up to 40 hr being required for a batch dissolution yielding a 1 M $Th(NO_3)_4$--9M HNO_3 solution. Irradiated ThO_2-UO_2 dissolves much more rapidly, regardless of method of preparation. Pelletized, arc-fused, and sol-gel-derived oxides (about 5% UO_2) were greater than 99.9% dissolved in about 7 hr after irradiation to 3000 to 98,000 Mwd/MT(U + Th).[11-13,29] Irradiated thorium metal fuel has been processed on a pilot-plant scale.[30,31] Metal slugs were dissolved in boiling 13 M HNO_3--0.04 M HF using an 8-hr dissolution period and leaving a 100% heel. The dissolution product was 6.5 M in HNO_3 and 1 M in Th. Thus, a feed adjustment step is probably required after dissolution of both ThO_2-UO_2 and Th-U alloy to provide suitable feed solutions for solvent extraction.

After non-aqueous conversion of the cladding to ZrO_2, or its removal as $ZrCl_4$, the product bed would be transferred to another vessel and leached with fluoride-catalyzed nitric acid. Tests with simulated Zr-clad sol-gel ThO_2-UO_2 fuel that had been treated with HF-O_2 showed that when the fluorine present in the product bed is removed by pyrohydrolysis, greater than 99% of the uranium and thorium, but only up to 15% of the ZrO_2, are leached in 5 hr with boiling 13 M HNO_3--0.05 M HF. The product solution was about 0.2 M in Th, 0.02 M in U, 0.02 M in Zr, 0.1 M in F, and 0.03 M in Al. If the fluorine had not been removed from the bed prior to leaching, up to 70% of the ZrO_2 and 90% of the fluorine would have been leached and the product solution would have been about 0.2 M in Th, 0.1 M in Zr, and 0.4 M in F. In all tests with fluidized-bed products, less than 2% of the alumina was dissolved. The leached alumina can, therefore, either be recycled or discharged to waste.

Dissolution of ThC has received only slight attention. Preliminary laboratory studies[32] indicate that arc-melted ThC dissolves readily in boiling fluoride-catalyzed nitric acid but only slowly in nitric acid itself. Dissolution in fluoride-catalyzed nitric acid results in a large fraction of the carbide carbon being converted to soluble organic species such as oxalic acid and mellitic acid. In this respect, the behavior

of ThC is identical to that of the uranium carbides in nitric acid.[33] Preparation of a suitable solvent extraction feed would, therefore, probably require dissolution of the ThC-UC in 13 M HNO_3--0.05 M HF followed by digestion of the resultant solution in acid permanganate to oxidize most of the soluble organic species. Such a process was tested on a laboratory scale with irradiated UC specimens.[34] An alternative to direct dissolution in nitric acid solutions is pyrohydrolysis of ThC followed by dissolution of the resulting ThO_2 in fluoride-catalyzed nitric acid. Although this method has not yet been studied, it is expected that ThC will behave like UC. Uranium monocarbide reacts rapidly with steam at 700 to 750°C, to give UO_2, CO_2, CO, and hydrogen.[34] Pyrohydrolysis of irradiated UC resulted in practically no volatilization of fission products. Combustion, in oxygen, is an alternative to pyrohydrolysis for carbide fuels, although the rate of oxidation of arc-melted carbides at 700 to 750°C is lower than the rate of reaction with steam. This fact, and the possibility of volatilizing ruthenium and cesium during combustion, makes the pyrohydrolysis method an attractive non-aqueous approach.

Other core materials such as UO_2-ZrO_2 and Th-Zr alloy have been considered for various reactors. No satisfactory aqueous dissolution process is evident for UO_2-ZrO_2, but it might be possible to dissolve the alloy in fluoride-catalyzed nitric acid.

2.2.2 Aluminum-Base Clad Fuels

Dispersions of Al_2O_3 in aluminum (designated as SAP or AMP) are being considered as fuel claddings, especially for organic-cooled reactors. Very little work has been done on the processing of SAP-clad fuels. Preliminary laboratory-scale experiments show that SAP can be dissolved either in NaOH-$NaNO_3$ solutions or in Hg-catalyzed nitric acid solutions. In experiments using boiling 2 M NaOH--1.78 M $NaNO_3$ (Na/Al atom ratios of 2 and 4), the initial dissolution rate of a SAP sample containing about 6% Al_2O_3 was about 20 mg min^{-1} cm^{-2}. This rate is high enough to allow penetration of a 30-mil-thick clad in 2 to 3 hr, and is about the same as that obtained for pure aluminum under the same conditions. Reaction with the NaOH-$NaNO_3$ solution left an Al_2O_3 residue corresponding to about 80% of the alumina in the original sample. On the other hand, dissolution of the SAP in boiling 4 M HNO_3--0.005 M $Hg(NO_3)_2$ (HNO_3/Al mole ratios of 4 to 8) resulted in practically complete solubilization of the sample, although the rate of reaction was much lower than that obtained in NaOH-$NaNO_3$ solution. Complete dissolution required about 20 hr; the initial rate of dissolution was only about 0.5 mg min^{-1} cm^{-2}. In contrast, type 2S aluminum and extruded 15% U--85% Al alloy dissolved[35] under the same conditions at rates of about 140 mg min^{-1} cm^{-2}.

Soluble losses of uranium and thorium in caustic decladding should be negligible with each of the core materials being considered. Although unirradiated ThC reacts readily with NaOH solutions,[36] it is highly probable that after irradiation the carbide will be inert, as is the case with uranium carbide.[25] If this is true, caustic decladding of carbide

(and the other types of) fuels probably would be a practicable approach.

2.2.3 Stainless Steel-Clad Fuels

Stainless steel claddings can be chemically separated from core materials either by aqueous or non-aqueous methods. The aqueous method (Sulfex process[14,37-40]) involves dissolution of the cladding in boiling 4 to 6 M H_2SO_4. The non-aqueous method is oxidative disintegration in HF-O_2 mixtures.[18-20]

2.2.3.1 Sulfex Decladding. With a 200% stoichiometric excess of boiling acid, the initial dissolution rate increases from about 2 to 30 mg min^{-1} cm^{-2} as the sulfuric acid concentration increases from 2 to 8 M. In cold pilot plant studies, penetration rates of 3 to 4 mils/hr were obtained in a recirculating dissolver.[40] Stainless steel that has been in contact with high-temperature water may be passive to sulfuric acid; in this event, dissolution is initiated by contacting the fuel with a piece of soft iron. The solubility of stainless steel sulfates at 25°C decreases from about 80 to 20 g of stainless steel per liter as the sulfuric acid concentration increases from 2 to 8 M.[38] Nionel appears to be suitable as a material of construction for the dissolver.

Irradiated UO_2 and ThO_2-UO_2 are practically inert to boiling 4 to 6 M H_2SO_4. Decladding of stainless steel-clad UO_2 fuel specimens that had been irradiated up to 28,200 Mwd/MT resulted in soluble uranium and plutonium losses of only about 0.05%.[24] Similar experiments with stainless steel-clad ThO_2-UO_2 fuel[11-13] irradiated up to about 25,000 Mwd/MT showed that uranium and thorium losses were 0.5% or less. No tests have been made of the reactivity of irradiated Th-U alloy in sulfuric acid; however, experiments with unirradiated uranium metal[22] showed it to be relatively inert to boiling 2 to 8 M H_2SO_4. The reaction of ThC with sulfuric acid has not been studied; however, both unirradiated UC and UC irradiated up to 6000 Mwd/MT reacted with 6 M H_2SO_4 at 80°C yielding a gas composed mainly of methane and hydrogen and a solid which was probably $U(SO_4)_2 \cdot 4H_2O$.[25] Conversion of the uranium to an insoluble sulfate is highly undesirable; thus, the use of Sulfex decladding for carbide fuels will probably not be practical.

After Sulfex decladding, the core materials would be washed and dissolved by the techniques described in Section 2.2.1.3.

2.2.3.2 HF-O_2 Disintegration. Stainless steel-clad fuels react with gaseous HF-O_2 mixtures in a fluidized bed in a manner similar to that of zirconium.[18-20] The stainless steel is converted to its respective oxides at the optimum temperature of about 650°C at a rate of about 60 mils/hr. The optimum gas composition appears to be about 40% HF--60% O_2. Bed products containing up to 30% U_3O_8 and 15% stainless steel oxides have been produced from stainless steel-clad UO_2 fuels. After removal of the fluorine by pyrohydrolysis, leaching with 1 to 15 M HNO_3 resulted in the recovery of greater than 99.9% of the uranium. The product solution was

about 0.2 M in U. The amount of iron oxide dissolved increased from 16 to 85% as the nitric acid concentration increased from 1 to 15 M. As discussed in Section 2.2.1.2, the effect of HF-O_2 on carbide, metal, or ThO_2-UO_2 cores has not yet been investigated.

2.3 Mechanical Head-End Processing

The mechanical approach to head-end processing of spent reactor fuels has been investigated during the past eight years in the U. S. The investigation has included disassembly, mechanical decladding, grind-leach, and shear-leach methods. Some of these pre-chemical mechanical treatment methods may offer attractive alternatives to the heretofore discussed chemical processes for thorium-bearing advanced converter fuels, e.g., crushing, grinding and leaching of graphite fuels, and the shearing and leaching of metal-clad fuels.

2.3.1 Crushing, Grinding and Leaching of Graphite-Type Fuels

In the burn-leach process described in Section 2.1, crushing of the graphite fuel is required as the first step unless the coated particles can be separated from the massive graphite log. Following crushing, an alternative to burning the graphite in a fluidized bed would be to further size-reduce the rough crushed fuel by additional crushing or grinding. All of the coated particles must be broken to permit recovery of fissile and fertile values by leaching. At ORNL, investigation of this alternate route of grind-leach has begun with HTGR fuel. Some very preliminary early work[41] has indicated wear problems in hammer-mill crushing equipment and difficulties in attaining complete leaching and washing of the graphite fines. Crushing, either with or without burning to destroy the bulk graphite, followed by grinding and leaching may be the only way to recover fuels with refractory coatings such as SiC on the fuel particles.

2.3.2 Shear-Leach Processing

The shear-leach process has been intensively investigated[42-44] and is to be used commercially by Nuclear Fuel Services, Inc., to process power reactor fuels clad with stainless-steel or zirconium alloys and containing cores of UO_2, ThO_2-UO_2, U-Mo metal, or Th-U metal. The process, developed at Oak Ridge National Laboratory, involves the shearing of the fuel bundle into short lengths to expose the fuel oxide or alloy, after which the exposed fuel is leached from the cladding.

The method is capable of processing any aluminum-, stainless-steel-, or Zircaloy-2-clad oxide or metal fuel, using the same equipment. Currently, there is no apparent advantage in processing graphite fuels by shear-leach, unless a shearing or breaking operation is used to subdivide graphite fuel logs. There are distinct advantages for the metal-clad fuels with core materials which can be leached without dissolving the clad. One considerable advantage is the lower cost of storing the

cladding material as metal waste rather than as Zirflex or Sulfex type liquid waste. It has been estimated that the waste storage cost for the leached metal clad is about 1/20th the cost of storing the corresponding chemical decladding waste.[43-45] Existing processing plants could be adapted by the addition of a mechanical head end to process a greater variety of fuel in conventional stainless steel equipment.

2.3.2.1 Shear-Leach Flowsheet. A typical shear-leach process flowsheet for stainless-steel-clad or Zircaloy-2-clad uranium oxide and/or thorium oxide fuel is presented in Figure 6. Fuel assemblies that have been manufactured by high temperature brazing are disassembled by sawing off the inert end fittings, sheath, and tube sheets. The resulting fuel tube bundles are then sheared. The sheared pieces are collected in a perforated basket containing a consumable carbon steel liner, and leached in a batch leacher. The liner is required to contain the fuel fines during transport of the basket to the leacher. The leached cladding is sent to underground waste storage along with the other metallic scrap from the fuel element.

The newer type of fuel element that has been assembled by retaining the fuel rod with wire grids or spring clips can be sheared intact; however, the grids and clips tend to remain as large pieces, complicating the shear operation and basket loading somewhat. An alternative method is the withdrawal of tubes from the parent assembly. In demonstration tests with Consolidated Edison Core B fuel, mechanical equipment, consisting of a hydraulic cylinder, bumper, support rack, elevating jacks, and ejector withdrew 14 fuel tubes simultaneously using a force of only 300 pounds. Fuel tubes removed from the fuel assembly in this manner can be satisfactorily sheared as a loose bundle.

The 250-ton prototype shear has been used successfully to shear unirradiated stainless-steel-clad uranium oxide and thorium-uranium-oxide fuel assemblies up to about 6 in. square, and containing up to 144 fuel tubes, into lengths of from 1/2 to 2 in. Zircaloy-2-clad oxide type fuel has also been sheared satisfactorily, with some sparking but without encountering any significant safety problems caused by the presence of Zircaloy-2 metal fines. Zircaloy-2-clad uranium metal fuel assemblies of the NPR type were successfully sheared into 1/2- to 2-in. lengths. In this case also, some sparking occurred during shearing, but there appeared to be no real fire hazard involved.

2.3.2.2 Shear-Leach Design Concept. A conceptual mechanical head-end and leaching equipment layout for shear-leach processing of about one metric ton of fuel per day is illustrated in Figure 7. This layout is based largely on the results of the developments carried out over the past several years. The basic uninstalled equipment cost, exclusive of the shielded cell, manipulators and supporting facilities is very roughly estimated to be $600,000, broken down as follows:

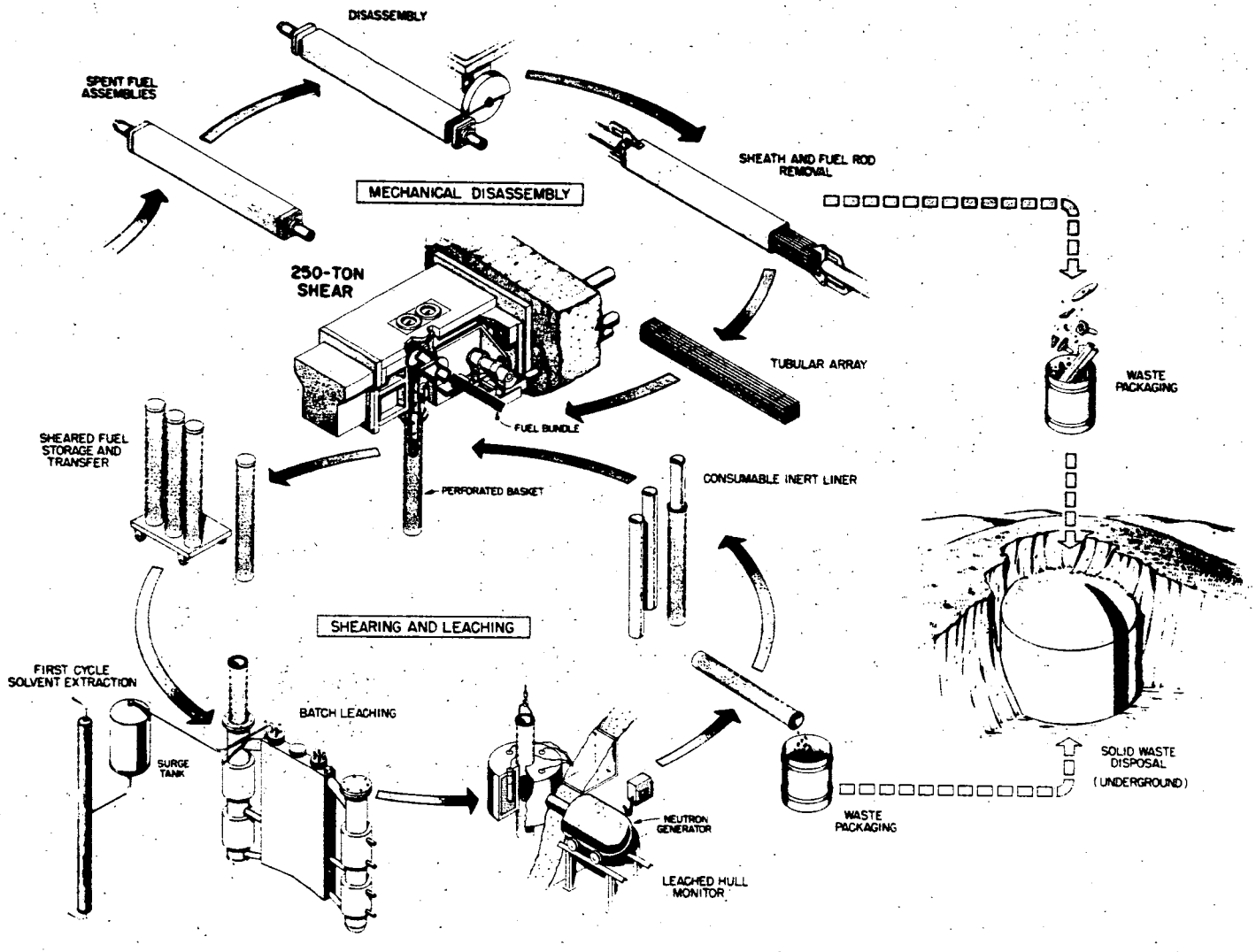
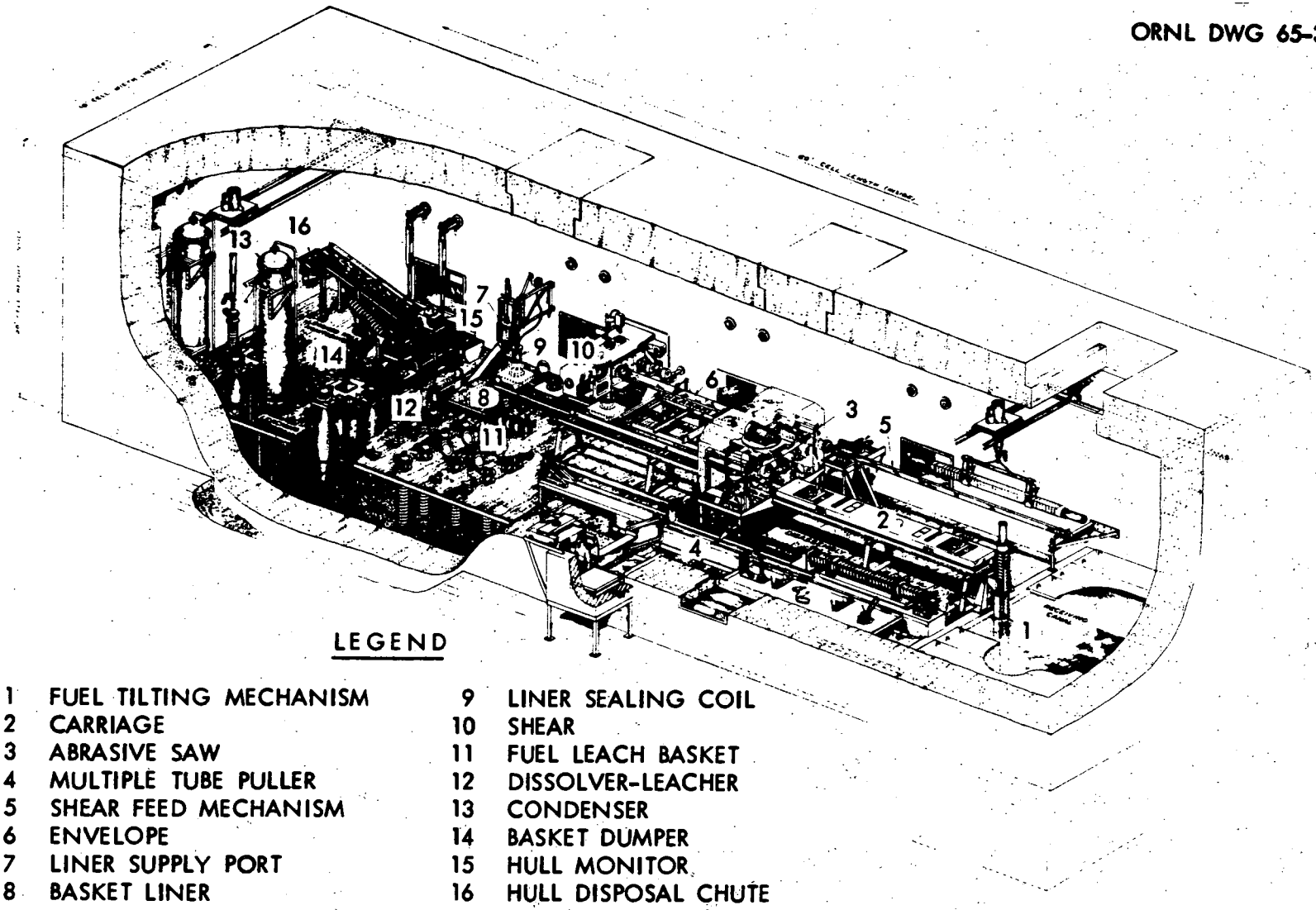


FIG. 6. SHEAR-LEACH PROCESS FOR SPENT REACTOR FUELS.

ORNL DWG 65-3075



LEGEND

1	FUEL TILTING MECHANISM	9	LINER SEALING COIL
2	CARRIAGE	10	SHEAR
3	ABRASIVE SAW	11	FUEL LEACH BASKET
4	MULTIPLE TUBE PULLER	12	DISSOLVER-LEACHER
5	SHEAR FEED MECHANISM	13	CONDENSER
6	ENVELOPE	14	BASKET DUMPER
7	LINER SUPPLY PORT	15	HULL MONITOR
8	BASKET LINER	16	HULL DISPOSAL CHUTE

FIG. 7. CONCEPTUAL DESIGN CONCEPT FOR A SHEAR-LEACH HEAD END FACILITY.

Shear	$225,000	Multiple Tube Puller	$10,000
Handling Table (carriage)	20,000	Fuel Leach Baskets (36)	36,000
Disassembly Saw	150,000	Magniforming	8,000
Accessory Equipment	50,000	Dissolver Leachers (2)	80,000
		Hull Monitor	25,000

With the exception of remote maintenance, all operations have been investigated sufficiently to show that each is feasible in a hot cell.

2.3.2.3 Leaching. Some typical bench-scale data are presented in Table 1 for shearing and leaching of unirradiated UO_2-ThO_2 and UO_2. The leacher used was similar to that of Figure 8, which was used for engineering-scale studies.

In the case of Zircaloy-2-clad urania-thoria fuels, some of the Zircaloy cladding and fines dissolved in the fluoride-catalyzed nitric acid required for core dissolution.[47] In tests conducted with pelletized, sol-gel and arc-fused thoria containing 4 to 5% urania in boiling 13 M HNO_3-0.04 M HF-0.04 M $Al(NO_3)_3$ dissolvent in the presence of Zircaloy-2 cladding and fines, from 1-5% of the massive cladding dissolved along with 60-80% of the minus-10 mesh fines. Recent shearing tests with Zircaloy-clad UO_2 indicate less than 1% of minus-10 mesh fines are formed. The rate of dissolution of ThO_2-UO_2 in fluoride-catalyzed nitric acid was diminished in the presence of zirconium but not enough to render shear-leach unfeasible for this type fuel.

Hot cell tests on the batch dissolution and leaching of irradiated sol-gel-derived, pelletized, or arc-fused ThO_2-UO_2 in boiling 13 M HNO_3-0.04 M NaF-0.04 M $Al(NO_3)_3$ indicated that irradiation increased the dissolution rate over unirradiated oxides, with up to 95% in solution at 8 hr and 99.8% in 24 hr.[11] Greater than 99.8% of the thorium and uranium was recovered in leaching tests with sheared fuel pieces. Uranium and thorium losses were less than 0.05%. In other tests, sheared stainless-clad UO_2 irradiated to about 8000 mwd/MT was easily leached in 4 M HNO_3.[48] Only about 0.6% of sheared stainless-steel cladding dissolves in fluoride-catalyzed nitric acid in 20 hours.

The batch leaching of stainless steel clad unirradiated UO_2-ThO_2 pellets sheared into 1/2- or 1-inch lengths has been investigated in an engineering-scale Pyrex glass and stainless steel leacher (Fig. 8). Dissolvent is circulated by convection. Variables affecting dissolution rates were studied with boiling (120°C) 12.7 M HNO_3-0.1 M $Al(NO_3)_3$-0.04 M NaF as the dissolvent. Typical UO_2-ThO_2 leaching data are presented in Fig. 9. The consumable carbon steel liner dissolves almost immediately. During the dissolution of the liner (about 2 min) 14 to 38% of the core is discharged from the basket and settles to the bottom of the leacher. A dissolvable 10-mil thick carbon steel liner adds about 11 grams Fe per kilogram of uranium or thorium to the solvent extraction feed.

Table 1. Typical Shear-Leach Data for Prototype Power-Reactor Fuels[46]

Fuel	Cladding	Shearing Force (tons)	Recommended Sheared Length (in.)	Core Dislodged (%)	Clad Dislodged (%)	Packing Density (g/cm^3)	Void Fraction (%)	Time to Batch Leach 99.9% (hr)	Notes
UO_2 (pellets)	Stainless steel or Zircaloy-2	50-90	1	36	2	4.8	50	1-1/2	
			1-1/2	28	2-1/2		55	2	
UO_2-ThO_2 (pellets)	Stainless steel	50-75	1/2	85	8	4.4	48	8	Sparged
			1	36	2		50	12	Not sparged
UO_2-ThO_2 (sol-gel)	Stainless steel	50-75	1/2	85	8	4.4	48	20	25% heel
			1	36	2		50	65	No heel

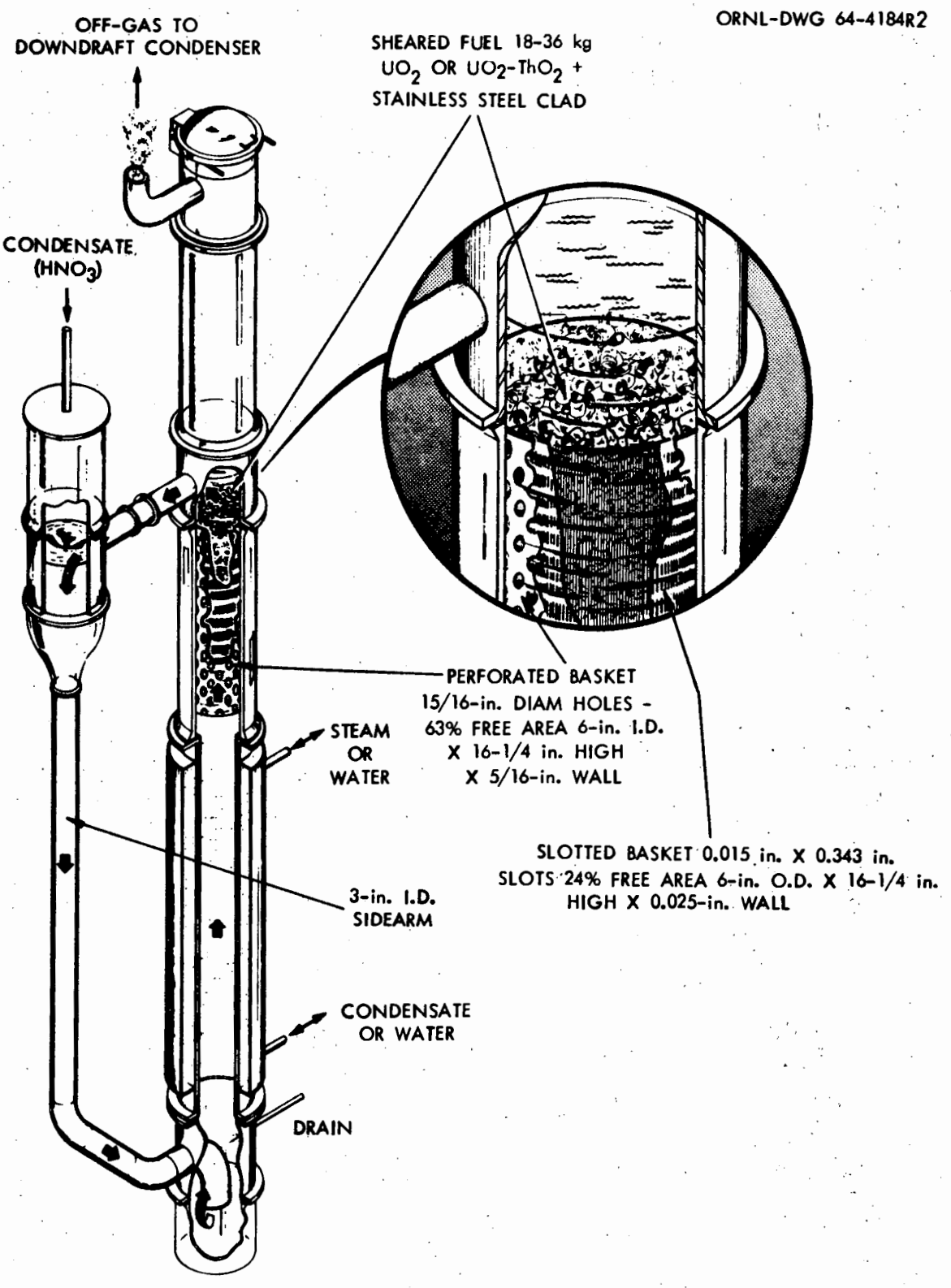


FIG. 8. ENGINEERING SCALE BATCH LEACHER (COMBINATION PYREX GLASS AND STAINLESS STEEL - 9.0 in. I.D. x 10 ft HIGH - CONVECTIVE CIRCULATION OF SOLUTION).

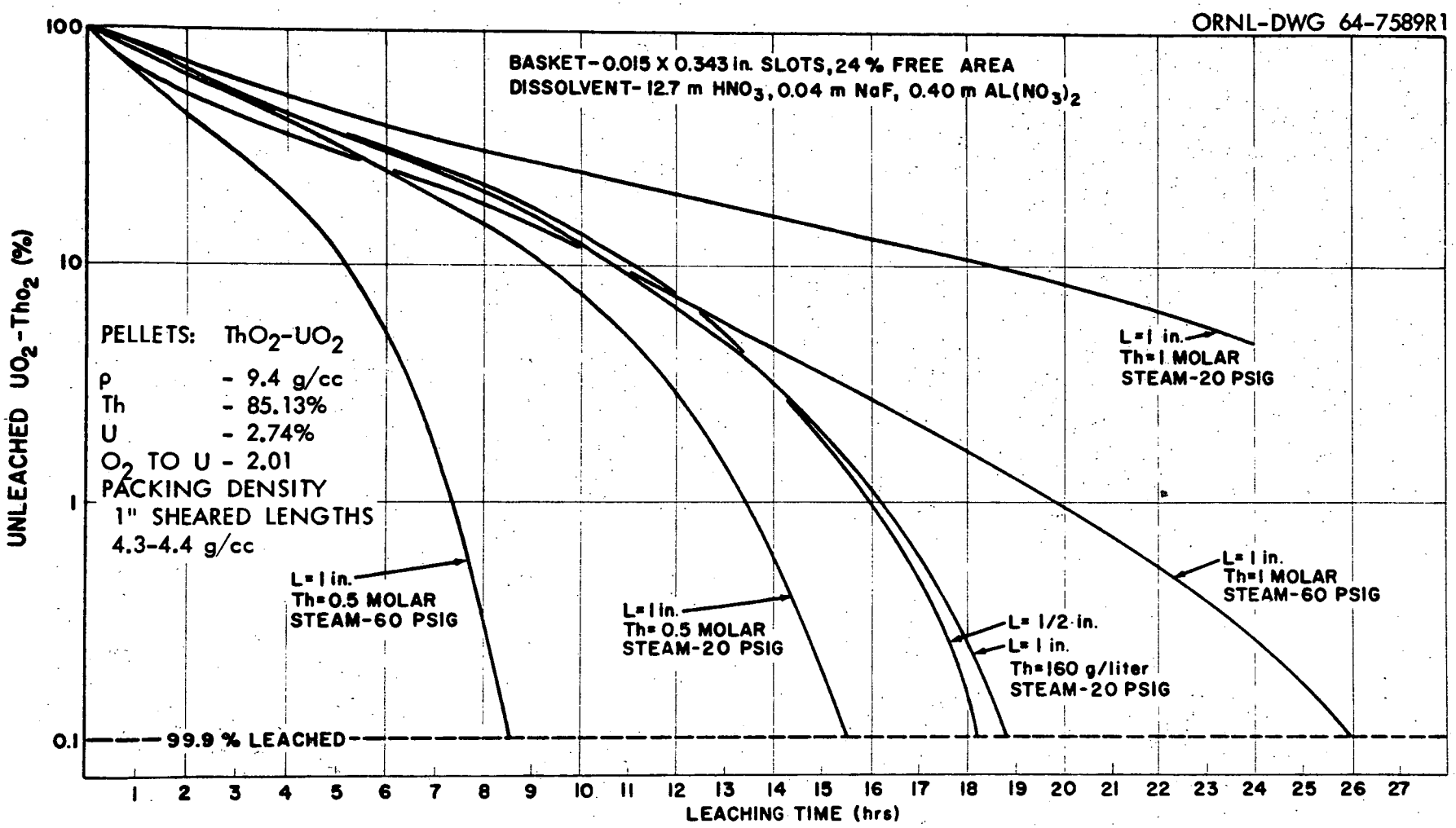


FIG. 9. ENGINEERING SCALE BATCH LEACHING OF UO_2-ThO_2 FROM SHEARED STAINLESS-STEEL-CLAD FUEL ELEMENTS.

The data of Figure 9 show the effect on dissolution time of terminal thorium loading, boilup rate as represented by steam pressure, and a comparison of 1/2 and 1 in. sheared lengths. The basis of the comparison is the time to dissolve 99.9% of the fuel. It required 8-1/2 hrs to produce a solution 0.5 molar in thorium at 60 psig steam as compared to 26 hrs to produce a 1 molar thorium solution. Fifteen hours was required to attain 99.9% dissolution when producing a solution 0.5 molar thorium at 20 psig steam as compared to the 8-1/2 hrs at 60 psig steam. About 18 to 19 hrs was required to leach both 1/2 and 1 in. sections when operating at the same conditions of terminal thorium loading and steam pressure. While producing a 1 molar thorium solution at 20 psig steam, only 95.5% was leached in 24 hrs as compared to 99.9% leached in 26 hrs at 60 psig steam. It was concluded that a rapid boil-up rate enhances dissolution and 1 in. sections are leached as efficiently as 1/2 in. sections. The amount of core remaining as unleached UO_2-ThO_2 was negligible in all runs. The empty hulls or leached shells were washed free of product solution by four separate water washes. The volume of wash water used each time was about one-fifth of the volume of empty hulls.

In sharp contrast to thoria fuels, unirradiated stainless clad UO_2 sheared into 1 inch lengths are easily completely leached in the same leacher with boiling 7 M HNO_3 in about 2 hrs.

Although shear-leach studies have not yet been performed using the advanced converter fuels under discussion, ample information has been developed with stainless-clad and Zircaloy-2-clad urania-thoria fuels to indicate that the projected fuels can be processed by shear-leach techniques. There are, however, two principal areas of doubt: (1) SAP cladding may tend to dissolve in the fluoride-catalyzed nitric acid dissolvent, and (2) carbide fuels may cause an unusually high wear rate of shear blades. There are also the chemical problems associated with the hydrolysis of carbides.

2.3.2.4 Recommended Practice. In applying the shear-leach process to a given fuel, the following practices are recommended:

a) Use sheared lengths of 1 inch.

b) Use perforated basket(s) in single or multi-legged recirculating type leacher. The free or open area of basket can range from 5% to 25% with the sheared fuel retained by a dissolvable liner of carbon steel or aluminum.

c) Leach UO_2-ThO_2 to a 25% heel in a period of 20-25 hrs.

d) Use hull wash water (or acid) to make up acid for next leaching step.

3. SOLVENT EXTRACTION PROCESSES

3.1 General Flowsheet Considerations

The emphasis of this discussion is on solvent extraction flowsheets suitable for reprocessing plants designed specifically to support a thorium-fueled power reactor industry, with secondary consideration given to the problems of processing thorium fuels in plants designed to handle standard uranium fuels. Recovery and purification of both the uranium and the thorium is emphasized, since discarding the thorium with the high-level fission product waste is not a desirable long-term solution for a large thorium-fueled power reactor industry from either the fuel-utilization or the waste-disposal points of view. The implications of the thorium recycle scheme, whether immediate recycle or delayed recycle after decay of ^{228}Th, on the choice of solvent extraction processes are considered.

3.1.1 Standard Uranium Fuels

The standard uranium fuel reprocessing method is the Purex Process,[49,50] based on extraction of both the low-enrichment uranium and the plutonium with 30% tri-n-butyl-phosphate (TBP) in a suitable diluent. The uranium and plutonium may be partitioned in the first cycle of extraction, or they may be co-stripped in the first cycle and partitioned in a second cycle. After partitioning, additional decontamination of the separated uranium and plutonium is obtained by one or more additional cycles of solvent extraction or, in the case of plutonium, by anion exchange. For high-enrichment ^{235}U fuels, the 25-TBP Process[51] recovers the uranium by extraction with 1.5-to-6% TBP if aluminum nitrate is used as salting agent or with 10-to-30% TBP if only nitric acid salting is desired. The corresponding processes for thorium fuels are described below in some detail. The differences in the flowsheets for uranium fuels and thorium fuels are: (1) uranium is the major constituent in one case and the minor constituent in the other; (2) thorium is extracted less strongly than uranium; and (3) the buildup of the gamma-active daughters of ^{232}U, ^{228}Th and ^{234}Th in the recovered products makes high-degree decontamination from fission products a relatively less important consideration than in uranium-plutonium recovery. Standard uranium fuels usually can be dissolved to give acceptably high uranium concentration and acceptably low excess acid concentration for feeding directly to solvent extraction. On the other hand, thorium fuels usually require a feed adjustment step after dissolution to increase the thorium concentration and remove excess acid.

3.1.2 Protactinium

Protactinium recovery is not assumed to be of interest in power reactor fuel processing for the purposes of this discussion. Since the 27-day half life ^{233}Pa is not normally extracted with the uranium,[52]

its primary effect on processing is the pre-processing time delay required for its decay to negligible levels. For thorium power reactor fuels this probably means a minimum of 120 days between reactor discharge and fuel processing and a typical delay of 180-to-210 days. This compares with a minimum delay of 90 days and a typical delay of 120-to-150 days for uranium power reactor fuels, for which the controlling factor is the decay of 8-day ^{131}I.

3.1.3 Extraction of Uranium Only

When only the uranium is to be recovered from the thorium fuel, dilute TBP or di-sec-butyl-phenylphosphonate (DBPP) may be used for the extraction. The Acid Interim-23 Process[53] uses 2.5-to-10% TBP (Fig. 10). The higher concentration permits higher processing rates but lower decontamination from fission products and a smaller separation factor from thorium would result. The lower concentration might be useful for criticality control, at the price of reduced throughput. The Kilorod Interim-23 Process[54,55,56] used 2.5% DBPP, which has a higher uranium-thorium separation factor than TBP by about a factor of 4 and also provides excellent decontamination from fission products. The thorium which remains in the aqueous phase may be recovered by a second extraction, either immediately or at a later date, or may be permanently discarded with the fission products. As discussed below, however, if the thorium is to be recovered a simultaneous co-extraction with the uranium probably is preferable.

These Interim-23 flowsheets, using thorium nitrate as the primary salting agent, are, of course very similar to the 25-TBP flowsheets using aluminum nitrate as salting agent.

3.1.4 Co-Extraction of Uranium and Thorium

Several systems have been developed for the co-extraction of uranium and thorium with TBP. In the original Thorex flowsheet[57] aluminum nitrate resulting from the dissolution of the aluminum cladding of the thorium metal slugs acted as the salting agent for the extraction of thorium and uranium from an acid-deficient solution into 42.5% TBP. A modification of this process involves a similar co-extraction from a solution containing both aluminum nitrate and nitric acid.[58] This modification gives good, though not equal, decontamination and has the advantage of not requiring a feed adjustment step; however, the acid in the feed decreases the processing capacity of a given size plant by decreasing the solubility of the thorium-TBP complex in the diluent, requiring operation with lower thorium concentration in the solvent phase in order to avoid formation of a third phase (i.e., a second organic phase).[59] Another variation of the Thorex flowsheet is the Acid Thorex Process[60] shown in Figure 11, for fuel solutions which do not contain aluminum. It involves the extraction of thorium and uranium from an acid deficient solution into 30% TBP, with nitric acid added at a lower stage in the contactor to provide salting when the thorium nitrate concentration has been reduced. This process results in maximum processing capacity and excellent decontamination, and

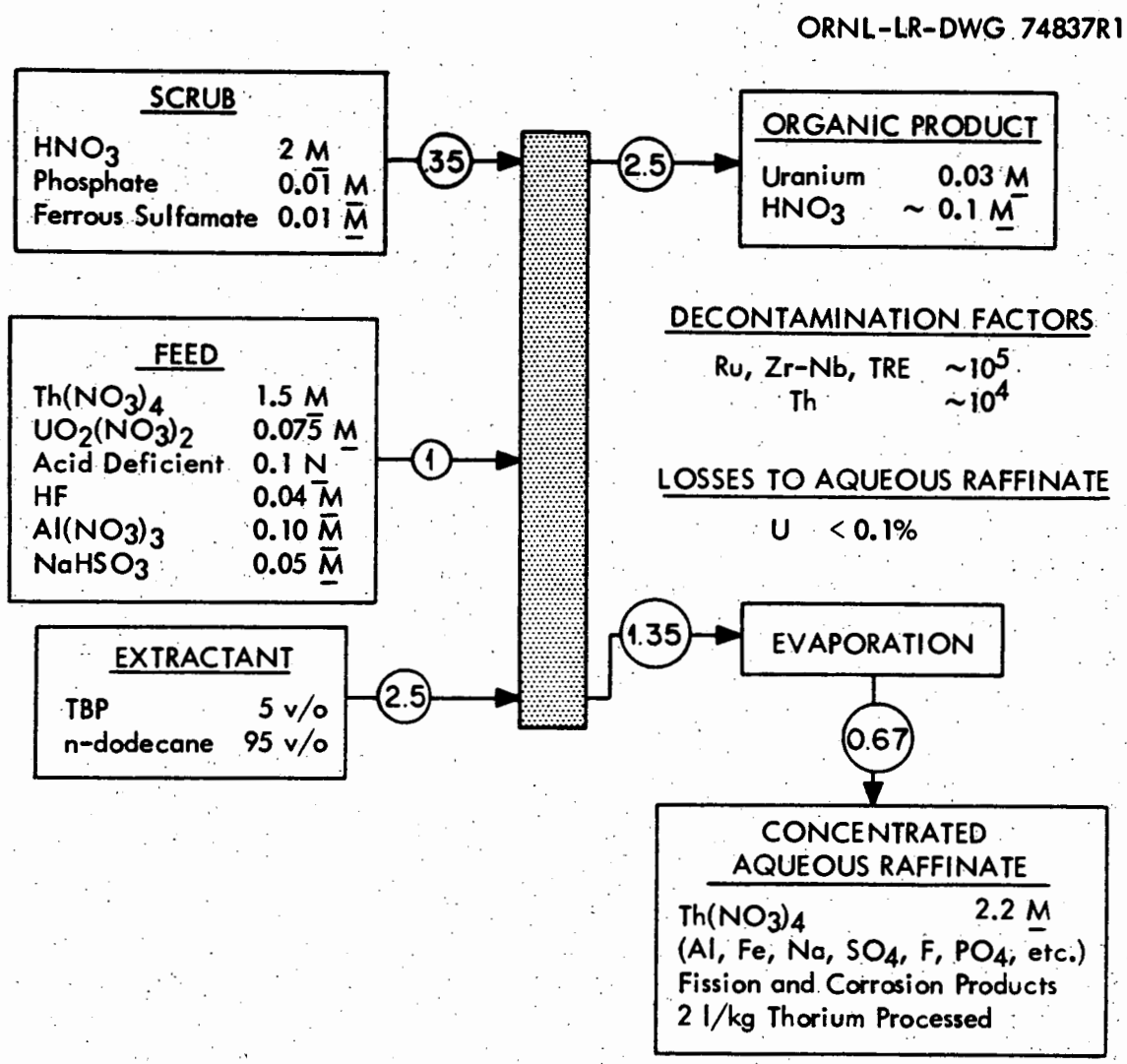


FIG. 10. ACID INTERIM-23 PROCESS.

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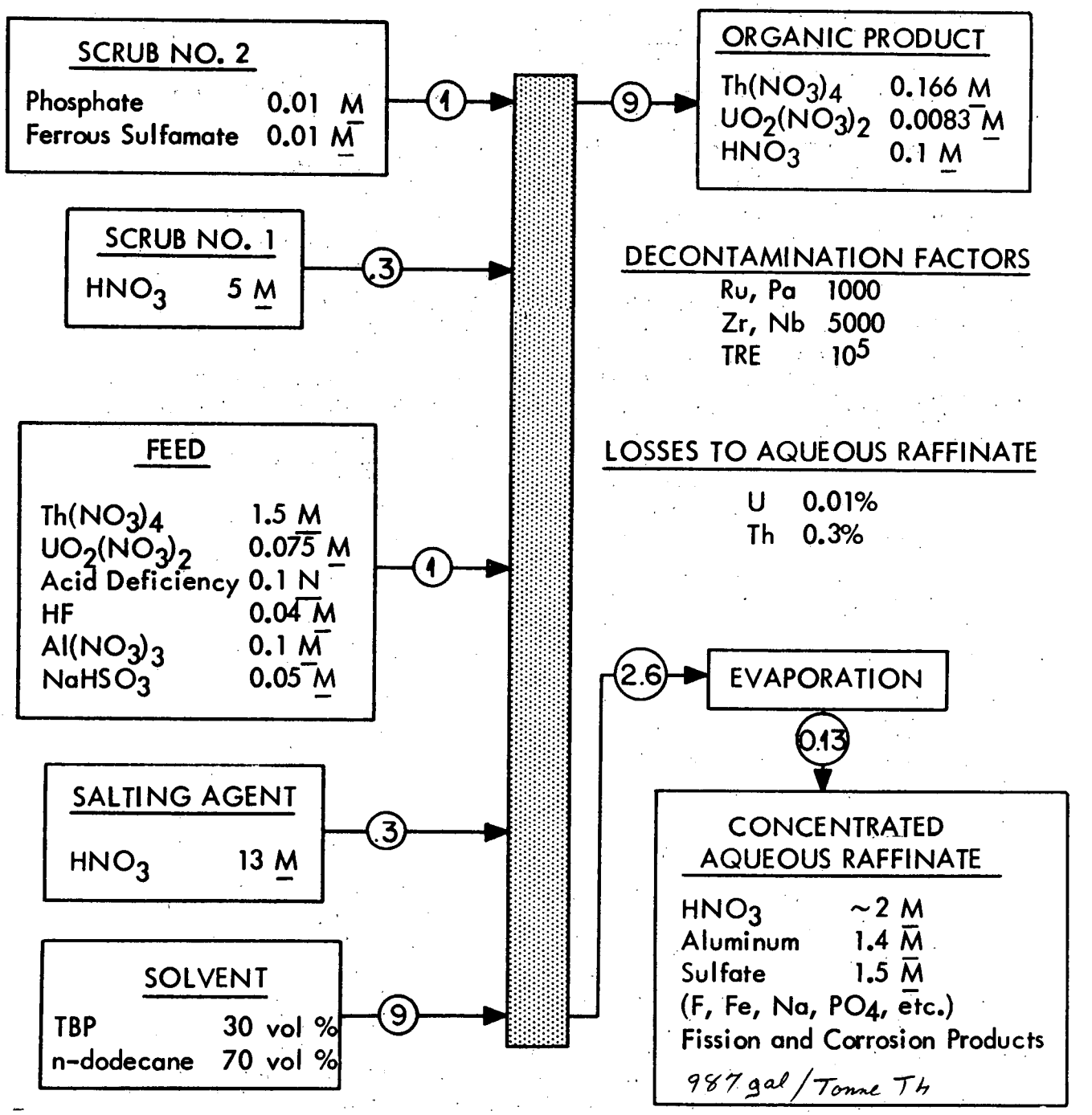

FIG. 11. ACID THOREX PROCESS FOR CO-EXTRACTION OF URANIUM AND THORIUM

$\frac{2.5 \times 10^5}{103} = 250$

reduces the volume of waste to be stored by eliminating the non-volatile aluminum from the aqueous waste. It does require a feed adjustment to produce the acid-deficient condition.

3.1.5 Effects of Fuel Type and Cladding

Any of the aforementioned flowsheets can be adapted for the processing of either oxide or metal fuels. Carbide fuels may also be processed, but if the thorium is dissolved by a low-temperature hydrolysis in nitric acid a feed adjustment step would be required to destroy the organic materials in the solution before feeding it to solvent extraction.[34]

The type of cladding must also be considered in the choice of a fuel recovery process. If the clad is dissolved with the fuel the added metal salts will act as salting agents for the solvent extraction, but they also will restrict the flowsheet to be chosen and affect the waste disposal operations. Large quantities of stainless-steel or zirconium salts in the solution would eliminate consideration of acid-deficient flowsheets since large amounts of precipitates would be formed in these cases. Even small amounts of solids in the feed, resulting from partial dissolution of the cladding or even from high concentrations of fission products in high-burnup fuels, may cause trouble. Batch contactors or pulse columns can handle solutions containing up to several percent solids but mixer-settlers are usually designed for solids-free solutions.

3.1.6 Non-Nitrate Systems

All of the fuel recovery systems being considered at present are based on nitrate solutions. Small amounts of other anions, such as fluoride, may be tolerated though adjustments may have to be made to compensate for their presence. If other dissolvents, such as hydrofluoric, hydrochloric or sulfuric acids, were required to dissolve the fuel, new solvent extraction flowsheets would have to be developed for these systems, perhaps using other organophosphorus compounds or amines.

3.1.7 Equipment Consideration

A few equipment items should receive special consideration in a thorium processing facility. A feed adjustment tank, in which the dissolved fuel can be heated to ~160°C, adds to flexibility by allowing a choice between acid and acid-deficient feed solutions for the solvent extraction system, and would also provide a means of destroying the organic materials which may be formed during the dissolution of carbide fuels. Equipment designers also should consider the possibility of third-phase formation in the thorium extraction systems. In a mixer-settler, the second organic phase accumulates between the organic and aqueous exit parts of the settler so that it cannot move out of the equipment. Since there is low turbulence in the settler unit, redissolution of this TBP heavy phase in the organic system is quite difficult. As a result, the operating conditions must be maintained conservatively

away from the region of third-phase formation. On the other hand no major difficulties occur if there is a small amount of third-phase formation in a pulse column and recovery is practically instantaneous when operating conditions are corrected. Also, as mentioned already, pulse columns have a much greater tolerance for solids in the aqueous feed to the solvent extraction system.

3.2 Power Reactor Fuel Processing

Figure 12 shows a general purpose solvent extraction flowsheet for thorium-bearing power reactor fuels. Insofar as practicable, cladding materials and any other unnecessary cationic or anionic constituents should be kept out of the dissolver solution, to permit a choice of the best possible feed adjustment and solvent extraction conditions and to avoid complicating waste treatment and disposal problems. Feed adjustment to maximum thorium concentration, preferably 1.5M, and to minimum acidity, preferably slightly acid-deficient, is desirable to permit maximum processing rate and maximum decontamination from fission products. Co-extraction of the thorium and uranium with 30% TBP, using the Acid Thorex flowsheet, decontamination by scrubbing with nitric acid, and selective stripping of first thorium and then uranium, will provide good recovery, separation and decontamination in a single cycle of solvent extraction. With an acid-deficient feed the decontamination factors from rare earths and ruthenium will be about 10^4 and 10^3, respectively, for both uranium and thorium. If it is necessary to use an acid feed the decontamination factors will be somewhat lower though not by more than a factor of 10. In either case, this degree of decontamination is more than is required from the reactor physics point of view, so that any additional decontamination requirement must be justified by fuel refabrication and other handling requirements before it is put back into the reactor. No additional fission product decontamination of the thorium is justified since the gamma activity of the daughters of ^{228}Th and ^{234}Th will require either remote fuel refabrication anyway or else storage for a sufficiently long time that the activity from extractable fission products will reach direct handling levels before the ^{228}Th does. Additional fission product decontamination of the uranium may be justified if it is to be recycled with virgin or long-decayed thorium, and an optional second uranium cycle is indicated to take care of this case.

3.2.1 Extraction Equipment Capacity

In general, the maximum thorium processing rate through solvent extraction equipment of a given size will be less than the maximum uranium processing rate for a similar flowsheet. For example, the total volumetric capacity (combined aqueous and organic flow rates) of pulsed extraction-scrub columns is about 900 gal/hr·ft^2 for either the Purex or the Acid Thorex processes;[61] but since the maximum capacity of the solvent for thorium is only about half that for uranium, the effective capacity of the equipment is only about half as much for thorium as for uranium.

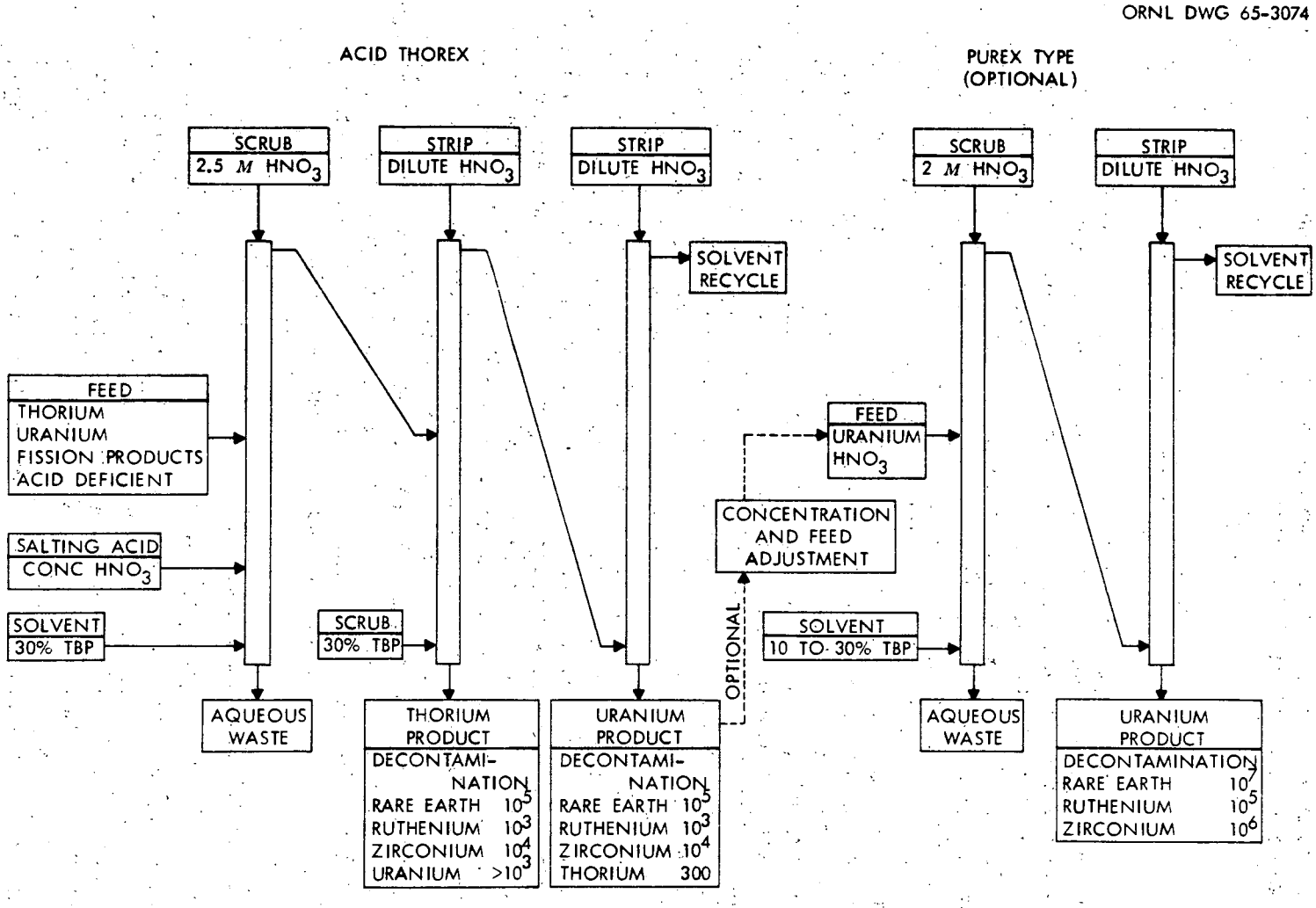

FIG. 12. SOLVENT EXTRACTION RECOVERY OF URANIUM AND THORIUM FROM POWER REACTOR FUELS.

A similar argument applies to the partitioning and strip columns. In a two- or three-column system designed for Purex the overall effect would be greater than a factor of two, since the capacity of a Purex plant for processing thorium is limited by the partitioning columns. In the Purex system the minor constituent, plutonium, is being selectively stripped from the major, uranium; whereas in the Thorex system, the major constituent, thorium, is being selectively stripped from the minor. For processing thorium fuels in an existing Purex plant, one might choose to use an Interim-23 first cycle, either discarding the thorium or recovering it from the waste in a second extraction, or one might choose to co-extract and co-strip the thorium in the first cycle and then separate them in an Interim-23 second cycle.

3.2.2 Waste Volumes

For low-burnup fuels, the volume of concentrated high-level fission product waste from either the Purex or the Acid Thorex flowsheets is about 50 gallons per metric ton of uranium or thorium processed. The waste concentrate is primarily a solution of nitric acid, fission products and miscellaneous other components such as aluminum, fluoride and corrosion products, and can be stored in the acid form in stainless-steel tanks, or neutralized and stored in mild-steel tanks, or else it can be calcined or converted to a glassy material for disposal as solids. For high-burnup fuels, above about 10,000 MWD/MT, the waste volumes may have to be larger to permit removal of the decay heat from the storage tanks, especially in the case of neutralized wastes since these will contain more solids from corrosion products and even from precipitation of the fission products themselves.

Waste solutions containing thorium, aluminum, or other cladding or salting agents will be much larger in volume and must be stored in the acid condition in stainless steel. If calcined or converted to glasses their volumes will also be much larger.

3.2.3 Close-Coupled Processing and Fabrication

If economic evaluation of thorium fuels cycles indicates that immediate recycle of the thorium is competitive despite the cost penalties of remote fuel refabrication, further attention should be given to the possible economic advantages of low-decontamination-factor processes closely coupled to the fabrication operations. A single cycle of Acid Thorex followed immediately by Sol-Gel oxide preparation and vibratory compaction into fuel tubes is an example of a promising close-coupled process. Even the solvent extraction step might be further simplified since decontamination factors as low as 10 might be acceptable in the overall fuel recycle scheme if this permitted sufficient cost savings to outweigh the disadvantages.

For recycle of uranium with virgin or long-decayed thorium, a study has been made of the Sol-Gel vibratory compaction route as regards the relationship between radiation dose to personnel, type of handling,

amount of ^{232}U in the uranium and scale of the uranium-thorium production operation.[62] For fabricating fuels containing 3% $^{233}UO_2$ in ThO_2, if the ^{233}U contains no more than 20 ppm ^{232}U a production rate of 100 kg/day can be achieved in an unshielded facility without overexposure of personnel. For the same conditions except for 250 ppm ^{232}U, shielding of two inches of lead would be required. For higher ^{232}U contents or larger production rates or for use of recycled thorium without $^{228,234}Th$ decay, remote fuel fabrication techniques will be required.

The direct maintenance of the equipment used in the Sol-Gel flowsheet is simplified by ease of decontamination. The Kilorod Facility,[56] in which the fission product contamination of the feed was negligible, was easily decontaminated with a vacuum cleaner to the degree that maintenance could be performed in "air suits" without excess worker exposure. The presence of fission products in the recycled uranium or thorium would complicate fabrication operations not only by increasing the shielding requirements but also by increasing the difficulty of equipment maintenance (and possibly even by affecting the Sol-Gel chemistry, though there is not enough data available at present to show what concentration of fission products can be tolerated). When the gel is fired to 1050°C, ruthenium, and perhaps other fission products also, would be volatilized and deposited in process equipment, especially the furnace. Previous experience has shown that decontamination would be difficult in such a case, and remote maintenance may be required.

The economic dependence of the overall fuel cycle cost upon the degree of decontamination, the fabrication and maintenance techniques, the amount of ^{232}U and $^{228,234}Th$ in the fuel being processed, and the scale of the production operation is sufficiently complex that further studies, both experimental and theoretical, are needed to indicate the best long-term route and timing for thorium fuel cycle development to follow, either in general or for a particular reactor and fuel type. In the short term, the equipment available and the particular reactor and fuel type considered will have a marked effect on the optimization of the fuel cycle.

4. POWER REACTOR FUEL REPROCESSING COSTS

4.1 USAEC Reference Fuel-Processing Plant

From 1957 to 1963 the standard basis for evaluating the spent-fuel processing contribution to nuclear power costs was the calculated cost for processing the fuel in question in the "AEC Reference Fuel-Processing Plant,"[63] a hypothetical plant capable of recovering purified uranium and plutonium from irradiated fuel at the rate of 1000 kg of uranium per day, up to 3% enrichment. At higher enrichments the capacity of this conceptual plant decreased, as a result of criticality considerations, to, for example, 930 kg/day at 4% enrichment, 537 kg/day at 10%, and 44 kg/day at 93%. The reference plant could process thorium fuels at the rate of 1000 kg/day if only the enriched uranium were to be recovered or 600 kg/day if thorium also had to be recovered, again subject to criticality limitations on the enriched-uranium processing rate. The USAEC announced in the Federal Register of March 12, 1957 that it would provide spent-fuel processing services at calculated charges based on the conceptual plant "on an interim basis ... until the time when processing is available commercially." Initially, the standard USAEC daily charge was $15,300, both for the calculated number of processing days required for a batch of fuel and for the calculated number of "turnaround" days (the time required between processing batches for shutdown, cleanout, and startup), but there was a provision for escalation that increased this figure to more than $17,000 in 1961[64] and to an estimated $19,800 by 1965.[65] The charges on a per-metric-ton basis were typically 25 to 100% or more higher than on the per-day basis, depending on batch size and daily processing rate.

4.2 NFS Commercial Processing Plant

In 1963 the USAEC accepted an offer by Nuclear Fuel Services, Inc., to provide fuel processing services on a commercial basis, beginning in 1965.[65] Nuclear Fuel Services (NFS) is now completing a plant with a nominal capacity of 1000 kg/day for uranium of up to 3% enrichment irradiated to burnups of up to 20,000 Mwd/MT and lower capacities for higher enrichments and burnups. The nominal capacity for thorium fuels is 500 kg/day for recovering only the enriched uranium. An extra charge will be assessed for the disposal of the thorium-bearing waste. The initial base charge of $23,500 per "revenue day" (processing plus turnaround time) is subject to future escalation. Since the minimum turnaround time under the NFS formula is one third the processing time, the minimum per-metric-ton price for processing is $31,300. For 3% enriched fuel irradiated to a burnup of 20,000 Mwd/MT at a thermal efficiency of 31%, this corresponds to a processing cost contribution to nuclear power of about 0.21 mills/kwhr(e). Typical NFS processing costs for first-generation power reactor cores will be considerably higher than this, for example:[65]

	mills/kwhr(e)
Southern California Edison	0.31
Indian Point UO_2	0.36
Indian Point ThO_2	0.75

These higher charges are the result of lower burnup, lower thermal efficiency, smaller processing batch size (which leads to a higher ratio of turnaround time to processing time) or higher enrichment than in the example calculation above. The large difference in the Indian Point UO_2 and ThO_2 figures is caused primarily by the 2-to-1 processing rate ratio between uranium and thorium, and secondarily by the extra charge for disposal of the high level waste containing the thorium. This extra charge results from larger volumes and the necessity of storing the wastes in the acid condition in relatively small stainless steel tanks, instead of neutralized storage in large mild-steel tanks.

4.2.1 Through-put Rate

Since the NFS price schedule is based on daily charges, the amount of fuel that can be processed per day determines the unit cost of processing, which is normally reported as $/kg (of uranium or thorium) for fuel cycle cost purposes. The NFS nominal throughput rate is 1000 kgU/day up to 3% enrichment, and falls to 880 kg/day at 4%, 465 kg/day at 10%, and 40 kg/day at 93%. The nominal processing rate for thorium is 500 kg/day to up 8.5% highly enriched uranium content, and is inversely proportional to the uranium content above this level. At present these enrichment penalties are based on pre-irradiation enrichment. There is some possibility that these criticality penalties may be relaxed in the future, by use of post-irradiation enrichment and/or by use of nuclear poisons in the processing solutions and materials of construction. The NFS-AEC contract[65] has an "isotopic limits per processing lot" clause which can result in the processing rate being inversely proportional to burnup above approximately 20,000 Mwd/MT. This limitation may be waived insofar as actual operating experience permits, and the actual limitation may be nearer 30,000 Mwd/MT.[66] The throughput rate penalties for enrichment, burnup, thorium, etc., are calculated separately and only the most restrictive applied, rather than all of the penalties being applied consecutively. For example, there would be no burnup penalty for thorium fuels up to 40,000 Mwd/MT (or possibly 60,000).

4.2.2 Fuel Type

The "standard" fuel for NFS is UO_2 or ThO_2-UO_2 sheathed in stainless steel, zirconium, or zircaloy, in an assembly up to 16 ft long and up to 6 in. diameter, weight up to one ton, with assembly casing and end fittings easily removable in the NFS mechanical cell, with individual fuel element diameters up to 0.75 inches and cladding thickness up to 50 mils, and with metallic hardware inside the assembly up to 1/8 in. thickness. Other fuel types may suffer processing rate penalties imposed by the

physical or chemical limitations of the NFS plant; for example, for U-Zr or U-Al alloy fuel clad in Zr or Al the processing rate is 400 kg/day gross weight of alloy plus cladding. An important consideration in advanced power reactor planning is that NFS is not now equipped to process certain fuel types, e.g., graphite-or-carbon-type fuels such as HTGR. In connection with proposals to build an HTGR, there has been some consideration of the possibility of adding a special head-end facility to crush and burn the fuel to permit more-or-less standard aqueous processing. A substantial extra charge would have to be assessed to an HTGR fuel to pay for the extra capital and operating charges involved.

4.2.3 Turnaround Time

The standard NFS turnaround time requirement is 8 days or one third of processing time, whichever is greater. To minimize turnaround charges relative to processing charges, the processing batch size should thus be equivalent to 24 or more processing days. For small fuel batches, requiring less than 8 processing days, the turnaround time can be reduced to equal to processing time (down to a minimum of 2 days turnaround) provided that these small batches can be combined with other similar small batches and also provided that processing can be delayed by NFS to permit convenient scheduling of combined small batches.

4.2.4 Waste Disposal Charges

For "standard" uranium fuels, as defined above, the $23,500/day base charge includes interim radioactive waste storage in mild-steel tanks by NFS and eventual perpetual maintenance by New York State. Fuel types which generate more high-level liquid processing waste than standard uranium fuels are subject to extra charges for interim and ultimate waste disposal. At present, thorium is not recovered and must be stored with its fission products in stainless steel tanks at an extra charge on the order of $9-16/kg. Alloy fuels such as U-Mo, U-Zr, and U-Al are also subject to substantial extra waste charges.

4.2.5 Escalation and Other Costs

The NFS price schedule is subject to escalation to cover increased labor pay rates and material prices. This is estimated to increase the base daily charge from $23,500 to $25,000 by mid-1970.

The processing charge does not include inventory or use charges on fuel prior to, during or after processing. For a typical standard fuel this might involve 120 days pre-shipping hold-up, 60 days shipping plus pre-processing hold-up at the plant, 30 days processing hold-up, plus another 30 days post-processing and shipping hold-up. Losses of nuclear material during processing, up to 1 - 1-1/2% at NFS, are not included in the processing charge. Shipping costs and costs of converting recovered material to forms other than concentrated nitrate solution are likewise not included.

4.3 Future Processing Costs

4.3.1 NFS

The base NFS price schedule discussed above is based on a nominal 300 revenue days per year for 15 years, 1965-1980. The actual amount of power reactor fuel to be processed during the first years of this period will be less than the nominal NFS capacity but will exceed it in the early 1970's and grow rapidly thereafter, according to estimates of increase in nuclear power generation. NFS probably will be able to handle up to 350 revenue days per year with minor increased costs, and the NFS-USAEC contract provides for a corresponding reduction in the daily charge, up to 10% at a load of 350 or more revenue days per year. Thus the Oyster Creek[67] cost analysis assumes $21,150/day after 12/31/74. Assuming that burnup and enrichment penalties can be relaxed to cover standard uranium fuel irradiated to 30,000 Mwd/MT at 31% thermal efficiency, the lower daily charge would correspond to only 0.13 mills/kwhr(e). On the other hand, escalation may override the base price reduction and optimum burnup for large PWR reactors may be only 20-25,000 Mwd/MT, so that 0.2 mills/kwhr(e) may still be a more normal cost of processing. At the expense of modest additional capital investment and operating costs, NFS may be able to significantly increase their processing rate capability. This should permit a substantial reduction in unit processing costs, but when and whether this actually occurs probably depends on future competitive conditions (see below). Modifications to NFS to permit thorium recovery (in addition to uranium) would eliminate the extra thorium waste disposal charge, but may substitute an interim thorium storage charge for the 7-15 year period required for ^{228}Th decay.

4.3.2 Other Near-Term Commercial Plants

For an industry predicted to grow as fast as is estimated for nuclear fuel processing after about 1973, it would be normal for other private companies to enter the field, in competition with NFS. The General Electric Co. has announced its interest in building a processing plant in the western U. S. Westinghouse has indicated its interest in offering a complete fuel cycle service, including processing. Other companies also have expressed an interest in processing; for example, a plant designed specifically for thorium fuels has been suggested. If this proliferation of processing plants occurs during the next decade, these new plants probably will be approximately the same size as NFS, and the economies of large-scale processing will be postponed.

4.3.3 Large Processing Plants

A design study[68] has shown that a 10 ton/day processing plant should cost less than twice as much to build and operate as a 1 ton/day plant, indicating a reduction in unit processing costs by a factor of approximately five. Depending on the burnup and thermal efficiency, a 10 ton/day

plant could service a very large nuclear power reactor economy, a size which may be many years away. On the other hand, a large nuclear desalination industry using natural uranium fuel might need such a large processing plant sooner than a power-only reactor industry.[69]

4.3.4 Ultimate Waste Disposal

The NFS base daily charge includes approximately $700 for perpetual maintenance of the liquid-waste tank farm, plus a similar amount for interim (15-year) waste disposal costs. For a typical power reactor this amounts to approximately 0.01-to-0.02 mills/kwhr(e). ORNL studies of the waste problem[70,71] indicate that this amount may be inadequate to cover perpetual tank storage costs, that perpetual tank storage of liquid wastes may not be adequately safe in any event, and that a safer ultimate disposal scheme (calcination to dryness and storage in a salt mine) may cost 0.02-0.03 mills/kwhr(e) on a large scale. These studies did not include the cost of disposal of the cladding waste. For disposal of these as leached metallic solids the cost is relatively small, but recent ORNL studies[72] of chemical decladding waste solution disposal indicate that this can easily cost as much as the high-level waste, because of the large volumes and their chemical composition, i.e., another 0.02 mills/kwhr(e). The large volume of Interim-23 waste, containing the thorium plus the aluminum nitrate salting agent, also would cost about 0.02 mills/kwhr(e) more than the Acid Thorex type waste.[72]

4.3.5 Advanced Converter Fuels

In support of the Advanced Converter Evaluation program,[73] fuel processing cost estimates were made for six types of advanced converter reactors: (1) uranium-fueled pressurized-water (PWR), assumed to include also boiling-water; (2) thorium-fueled spectral-shift-controlled (SSCR); (3) uranium-fueled, pressure-tube, heavy-water-cooled-and-moderated (HWR-U); (4) thorium-fueled, heavy-water (HWR-Th); (5) thorium-fueled high-temperature-gas-cooled (HTGR), of the "TARGET" type; and (6) uranium-fueled sodium-graphite (SGR).

The cost estimates, summarized in Table 2 , are based on 15% annual fixed charge rate (FCR) on total capital investment, which rate is approximately equivalent to that applicable to the first private commercial reactor fuel processing plant, Nuclear Fuel Services (NFS).[65] At a 22% FCR, which would be more typical of most private chemical companies, the unit costs would be about 25% higher.

These cost estimates for the evaluation were made for hypothetical future nuclear power economies of 15,000 Mw(e) of a given reactor type, with all of the fuel from that reactor type being processed in a single-purpose plant designed to exactly match its load. Thus, differences in annual throughput rate and nominal daily capacity in the various plants were caused by differences in burnup, thermal efficiency and discharge batch size for the various reactor types. Table 2 shows the estimated

Table 2

FUEL PROCESSING COSTS AS A FUNCTION OF NUMBER AND TYPE OF REACTORS

Reactor Type	PWR-U	HWR-U	SGR-U	SSCR-Th	HWR-Th	HTGR-Th
Burnup, Mwd/MT	21,000	11,100	22,000	28,800	27,800	58,000
Thermal Efficiency, %	31.1	26.8	43.6	31.2	26.1	44.4
Batch Size, MT	33.9	19.5	12.2	66.3	13.5	7.89
Number of 1000 Mw(e) Reactors	Processing Cost in mills/kwh(e), 15% F.C.R.					
5	0.332	0.396	0.321	0.357	0.381	0.306[1]
10	0.201	0.237	0.195	0.212	0.232	0.184[1]
15	0.154	0.177	0.144	0.159	0.173	0.138[1] 0.149[2] 0.135[3]
20	0.125	0.136	0.104	0.116	0.125	0.099[1]

Footnote: The superscripts 1, 2, 3 refer to three alternate HTGR processing schemes (see text).

processing contribution to nuclear power cost as a function of industry size, from 5,000 to 20,000 Mw(e). In this table the burnup values are held constant at approximately the economic optimum indicated by consideration of overall fuel cycle costs at 15,000 Mw(e), though the optimum burnup would be higher for smaller industry size, and vice versa. All other things being equal, the power cost of processing decreases with increasing burnup, thermal efficiency and batch size, is lower for uranium fuels than for thorium, and is lower for metal-clad oxides than for carbon-carbide-graphite or sodium-bonded fuels. The combined interplay of all these considerations in this study was to minimize differences in cost per kwhr(e) though the costs in dollars per kilogram of fuel varied widely.

For all of the fuels except HTGR all of the uranium and plutonium, or thorium and uranium, in one processing batch (assumed to be the same as one reactor discharge batch) were dissolved together and then partitioned and decontaminated by solvent extraction. For HTGR three alternate schemes proposed under the TARGET[5] concept were considered: (1) mixed thorium-uranium fuel, as with the other reactors, (2) thorium and uranium in separate particles before irradiation, with the thorium-plus-bred-uranium particles processed separately from the high-burnup-uranium particles; and (3) separate particles as in (2) but with the high-burnup-uranium particles discarded directly to waste disposal instead of being processed, on the basis that their high U-236 content makes recycle of this uranium to HTGR reactors undesirable from the overall economics and physics points of view. Scheme (2) costs more than scheme (3) and would have to be justified on the basis of a market value (for reactors other than HTGR) in excess of the additional cost of $1-to-$2 per fissile gram recovered.

These cost estimates were based on modifications of previous estimates of processing plant costs, by duPont,[68] and of ultimate waste disposal costs, by ORNL.[70] They are conservatively high in that they would predict higher costs than the actual NFS pricing formula for a plant of comparable fuel processing capability, and also in penalizing thorium fuels for their known processing disadvantages vis-a-vis uranium while not granting any cost credit for potential advantages such as the possibility that only one solvent extraction cycle will be sufficient since the thorium-uranium recycle scheme may require remote fabrication anyway. On the other hand, they may not be quite so conservative in assigning only a moderate head-end cost penalty to SGR and HTGR fuels on the assumption that present development programs will be successful. These estimates for the Advanced Converter Evaluation agree with those made earlier for large desalination reactors,[69] except that the earlier estimates made less allowance for turnaround time between batches and for ultimate waste disposal and used 7.7% fixed charge rate (for municipal or similar financing).

4.3.6 Costs in an Expanding Economy

The cost studies described above assumed an "equilibrium" economy with the fuel from 5,000 or 10,000 or 15,000 or 20,000 Mw(e) of a given

reactor type being processed in a plant designed to match its load and operating at constant full load. A dynamic economy starting small and growing large over a period of years cannot automatically expect to experience processing costs as low as those equivalent to a static economy of the same size at any given time. A processing plant must be built at a particular time and with a particular design capacity - it may be able to increase its actual capacity somewhat over a period of time as a result of technological improvements and inherent over-capacity in its basic design; but it cannot be built initially to match a small reactor economy and then expanded incrementally each year at marginal additional cost to keep up with the load as the reactor economy grows. Thus, in general a processing plant will be over-sized initially, enough so that it eventually can achieve unit costs low enough to give it an economic life long enough to permit it to recover its capital investment plus an acceptable rate of return on investment and still meet actual or potential competition from other plants. Since such a plant must start up on less than a full load, its average load over its life will be less than its equilibrium capacity and hence its average unit costs over its life will be higher than the calculated equilibrium costs. This "start-up penalty" can be appreciable for plants, such as spent fuel processing plants, which have a high ratio of capital cost to operating cost. In the case of the NFS plant, the USAEC is providing a "base load" during the first five years of operation, permitting a pricing policy based on a full load for an assumed 15-year plant life. The USAEC has indicated a willingness to provide a base load also for a second private processing plant, but this type of support cannot be assumed for all future plants in a private competitive economy.

A study of optimum processing plant size, timing and location in a growth economy has been started. A computer code will be developed to calculate the minimum cost strategy as a function of input assumptions regarding growth curve, cost scaling factors, financing conditions, regulatory and competitive conditions, etc. As a first step in this study, an economic evaluation of HTGR head-end processing costs was made for the design and cost estimate presented in Section 2.1.3.[1] This head-end facility could handle the fuel from up to 10,000 Mw(e) of HTGR reactors, but a reasonable estimate is that it might be 10 years after the first commercial-size HTGR begins to discharge fuel before the HTGR industry reaches 10,000 Mw(e). A present-worth economic analysis, similar to that of Vondy,[73] indicated head-end capital and operating unit charges varying from less than $30/kg for a plant with a full load for 15 years to more than $130/kg for the same plant with a growing load for only seven years. It was indicated that the plant size was not optimum for the growth curve assumed, but the optimum size has not yet been calculated.

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