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SURVEY OF HEAD-END PROCESSES FOR THE RECOVERY OF URANIUM AND THORIUM FROM GRAPHITE-BASE REACTOR FUELS

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ABSTRACT

The literature on development of head-end processes for graphite-base reactor fuels was surveyed. For the coated-particle fuels that are currently receiving the most attention, only two of the processing methods considered appear to be feasible on a technical scale. Work on these methods, the burn-leach and grind-leach processes, is summarized in detail.

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SURVEY OF HEAD-END PROCESSES FOR THE RECOVERY OF URANIUM AND THORIUM FROM GRAPHITE-BASE REACTOR FUELS

I. INTRODUCTION

Graphite fuel elements for gas-cooled high-temperature reactors consist of uranium and/or thorium carbide or oxide particles dispersed throughout a graphite matrix. Fuel elements proposed in the design of the AVR reactor in Juelich would contain pyrolytic carbon-coated carbide or oxide particles in a graphite sphere.[1] Processing methods, especially head-end methods, other than those used for metal-clad uranium metal or oxide fuels will be required for the graphite fuels. It is first necessary to separate the bulk of the graphite from the uranium and/or thorium. The shells of the coated particles, which are very inert chemically, have to be cracked and the uranium and/or thorium have to be transformed from the carbide or oxide into another compound prior to further processing. Especially with thorium-bearing elements, there are some points requiring special attention.[2] The concentration of the dissolvent is high, the fission product concentration is high because of the high optimum burnup, and the gamma activity of the daughter nuclides of ^{232}U and ^{228}Th decay chain is also high. If remote handling is to be avoided, storage of the ^{228}Th for 10 to 15 years to allow for its decay is necessary. Costs of reprocessing irradiated thorium are high, so that consideration of processes that recover only uranium and store thorium in the waste is worthwhile.

Methods for reprocessing graphite-bearing fuels have been tested recently,[2-4] especially in the U. S., but the only process which is feasible today on a plant scale and which separates thorium and uranium from each other and from their fission products is the Thorex process.[3,5,30] Thus, head-end processes which give a product suitable for the Thorex process are to be preferred. The burn-leach[6] and grind-leach[20] processes look today as the best possibilities, but others[2,4,9] have also been tested with only moderate success. Consequently, only the burn-leach and grind-leach processes will be reviewed in the following sections.

2. BURN-LEACH PROCESS

2.1 Flowsheets[6,7]

The process is shown schematically in Fig. 1. First, the fuel is crushed and fed into a fluidized-bed burner. The fluidized bed contains a suitable amount of alumina which provides for good heat transfer. The fuel is burned continuously in the temperature range of 700 to 750°C. The oxygen is consumed almost quantitatively, so that the off-gas is mainly CO_2 with less than 5% CO. In the case of carbide particles, the product is a free-flowing fine pulverized ash. The whole batch is transferred into a dissolver and leached with Thorex reagent. The alumina will not be attacked and can be recycled to the fluidized bed or stored as waste. The dissolver product goes to feed adjustment and then to solvent extraction.

2.2 Burning of Carbon-Coated Carbide Particles

In recent years there has been a large number of experiments on the burning of graphite fuel elements.[4,6,8-11] The graphite elements burn rapidly in the temperature range of 700 to 750°C. In pilot plant tests using a 10-cm-diam fluidized-bed burner,[8] burning rates of 1.1 to 1.4 kg of fuel per hr were achieved with oxygen flow rates of 37 to 45 liters per minute. The off-gas consisted mainly of CO_2; the CO concentration was less than 5%. The bed consisted of 60 to 100 mesh alumina. The oxygen utilization decreased with increasing oxygen flow from 97% to 90% as the flow rate increased from 37 to 45 liters per minute. Peach Bottom fuel has been burned in a 13-cm-diam reactor filled with equal parts of 90 and 120 mesh alumina.[12] The fuel was crushed to -10 mesh prior to burning. The fuel burned in less than 8 hr in the temperature range of 700 to 750°C with an oxygen utilization of more than 90%. The fluidized bed contained less than 0.2% unburned graphite. The product bed contained 30% $ThO_2 + U_3O_8$ which were homogeneously dispersed throughout the alumina.

In another typical experiment,[2,6] crushed Peach Bottom elements were burned in a 10-cm-diam reactor with 20 kg of alumina. The bed was preheated with

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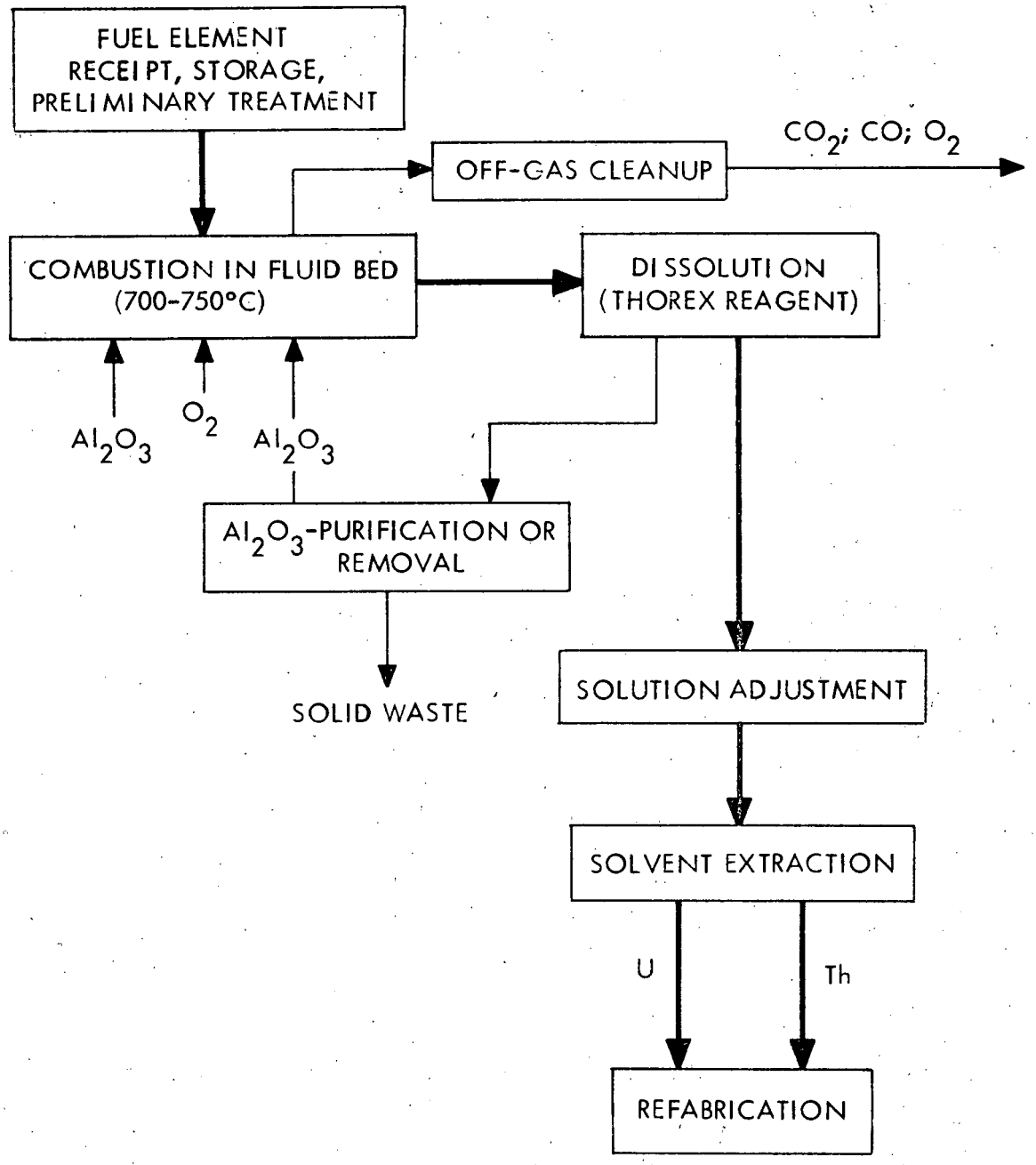

Fig. 1. Fluid Bed Burn-Leach Process.

hot nitrogen and by use of external heaters. The fuel ignited at 650°C. On reaching this temperature, the nitrogen was replaced by oxygen preheated to about 600°C.[13] The temperature inside of the reactor was held constant in the range of 700 to 750°C by air-cooling the finned outside of the reactor. The CO_2 and CO concentrations in the off-gas were monitored continuously by gas chromatography and infrared absorption. When the CO_2 and CO concentrations decreased, new fuel was fed to the reactor. Fresh alumina was also added periodically to compensate for the semicontinuous removal of product bed material. Up to 4 kg of fuel are burned in a single run. At the end of the experiment, when the graphite content of the bed is low, the fluidized bed was again heated externally to provide for complete burning of the graphite. The oxygen flow rate was about 110 liters/min, and the pressure in the bed was about 1.2 atm. The off-gases were cooled to about 550°C in the finned upper part of the fluidized-bed burner.[13] Previous experiments[4] showed that burning of HTGR elements produced a dust in which 99% of the particles had diameters of less than 0.3 micron. Micro-filtration of the gas that passed through the sintered metal filters removed practically all of the solid particles. The off-gas contained an average of 90% CO_2, 5% CO, and 5% O_2. Corrosion in the reactor and attrition of the Al_2O_3 were negligible. The product bed contained less than 0.1% unburned carbon and up to 30% ThO_2-U_3O_8.

2.3 Burning of Carbon-Coated Oxide Particles

Although no studies have yet been made on the burning of oxide particles coated with carbon, the method should be as feasible as for carbide particles.[13] It is not certain that oxide particles will retain their shape in the fluidized bed, especially after irradiation to burnups of 50,000 to 80,000 Mwd/t. UO_2 oxidizes rapidly in the temperature range of 500 to 800°C giving U_3O_8 powder. Therefore, UO_2 particles should behave in the fluidized bed similarly to carbide particles. Studies with unirradiated UO_2-ThO_2 showed[2,7] that solid solutions containing up to 75% UO_2 were not attacked by oxygen at 800°C. It is probable that such oxide particles, because of their high density and large diameter, will settle to the

bottom of the reactor. In this case it should be possible to have a relatively alumina free product.

2.4 Behavior of the Fission Products During the Burning Step

As yet, no irradiated fuel has been burned in a fluidized bed. Burning of irradiated Peach Bottom elements [10,000 Mwd/t(U + Th)] in a tube furnace at 800°C using a large excess of oxygen showed that in 6 hr 35% of the cesium and up to 96% of the ruthenium had been volatilized. The amounts volatilized increased with increasing temperature. The volatilized products condensed in the cold part of the tube furnace. The remaining activity was retained by a sintered metal filter with a porosity of 40 microns.[14] Using the quantitative amount of oxygen, it is possible that RuO_4 will be reduced to the less volatile RuO_2.[15,16] Colby[17] volatilized RuO_4 from irradiated UO_2. Below 600°C there was no volatilization of ruthenium; at 700°C, 50%; and, at 1300°C, 96%.

2.5 Off-Gas Cleanup

Some possibilities of off-gas cleanup have been discussed.[2] A decontamination factor of more than 10^8 should be possible by mixing the off-gas with steam, then condensing the steam, and, finally, filtering the gas through absolute filters. A severe problem, especially for reprocessing plants in a country with high population, is the contamination of the relatively high off-gas volume by xenon and krypton. Possibly oxide particles will remain intact in the reactor and burner and will retain part of the gaseous fission products; however, one has to work with the hypothesis that all of the rare gases will be in the off-gas stream. Absorption on charcoal beds, scrubbing, or freezing of CO_2 are very expensive methods because of the large volume of the off-gas. A method proposed in a patent by Dennis[19] or the use of permselective membranes, as proposed by Holmes,[18] give new possibilities.

2.6 Leaching of the Fluidized-Bed Product

Dissolution of ThO_2 and UO_2 goes by the reactions:

$$ThO_2 + 4HNO_3 \rightarrow Th(NO_3)_4 + 2H_2O$$

$$2UO_2 + 6HNO_3 \rightarrow 2UO_2(NO_3)_2 + NO + NO_2 + 3H_2O.$$

Bond[21] dissolved sintered pellets (96% ThO_2, 4% UO_2) in HNO_3-HF. Dependent on economic consumption of acid, concentration of the metal ions in the product, and the dissolution rate, there was an optimum at 200% excess of 13 M HNO_3, 0.05 M HF. No precipitation of ThF_4 was noted at fluoride concentrations up to 0.1 M. The product was 1 M $Th(NO_3)_4$ and 8.5 M HNO_3. Aluminum complexes fluoride ion, and so decreases the corrosion rate of the dissolver. Aluminum concentrations up to 0.2 M had no marked influence on the rate of dissolution.[22] Up to 10% UO_2, the rate of dissolution was not dependent on the UO_2 concentration in the pellet.[25] In some studies,[23-29] unirradiated and irradiated pellets of different densities, and pellets containing fissia, have been dissolved in 200% excess of 13 M HNO_3, 0.04 M NaF, 0.1 M $Al(NO_3)_3$. The rates of dissolution varied from 1 to 18 mg min^{-1} cm^{-2} depending on the density and particle size of the pellet, and the HNO_3, HF, and thorium concentrations in the solution.[28] Oxide ash of burned pyrolytic carbon-coated UC_2-ThC_2 particles dissolved completely in boiling 13 M HNO_3, 0.04 M HF, 0.1 M $Al(NO_3)_3$ in 6 to 8 hr (Ferris, et al., ORNL-3186 and ORNL-TM-193, cited in reference 28). HTGR fuel, irradiated to a burnup of 10,000 Mwd/tonne, has been burned in the temperature range of 800 to 1200°C to examine the influence of the temperature on the rate of dissolution.[14] In each case, the ash (ThO_2-U_3O_8) was a free-flowing powder which dissolved completely in 7 hr in boiling 13 M HNO_3, 0.04 M HF, 0.04 M $Al(NO_3)_3$. Most of this experimental work has been done with pelletized ThO_2-UO_2. Studies with 150- to 600-micron-diam sol-gel particles[7] showed again that the optimum nitric acid concentration is 13 M. In boiling 13 M HNO_3, 0.05 M HF, 250- to 300-micron-diam sol-gel particles dissolved completely in 3 hr. The product solution was 0.5 M Th. The

presence of a large excess of alumina (up to 80%) was not harmful at all. Particles with more than 50% UO_2 perhaps can be dissolved without adding HF. Solid-solution sol-gel particles with more than 50% UO_2 dissolve in 13 M HNO_3 only slightly more slowly than in 13 M HNO_3, 0.05 M HF.[7]

Demonstration of the burn-leach process on an engineering scale[6,13] showed that the process was especially suited as a head-end process for graphite fuels that contain pyrolytic carbon-coated carbide or oxide particles. Silicon carbide coatings and alumina are not attacked.[4,31] Experiments of the burning and leaching of high-burnup fuel elements have not yet been made. The problem of off-gas cleanup has not yet been solved.

3. GRIND-LEACH PROCESS

3.1 Flowsheet[4,20,32]

The fuel elements are crushed in a hammer mill to a few millimeters particle size. After crushing, the fuel is ground in a roll mill to less than 80 microns. The product is fed into a dissolver and then leached with boiling 13 M HNO_3, 0.04 M HF. Dependent on the design of the dissolver (sparging plus thermosyphon), there can be rapid circulation of the slurry. Foaming, caused by feeding of the product, and graphite dust in the off-gas will be problems on a plant scale. The off-gases, H_2O, O_2, NO, traces of N_2O, NO_2, and with carbide particles, also CO_2, go via reflux condenser and scrubber to a stack. After leaching, the slurry is pneumatically transferred into a filter vessel where the leached graphite is caught on a bed of granular alumina which acts as the filter. The filter cake is washed 3 or 4 times with water and is stored as a solid waste or burned. The filtrate goes to feed adjustment and then to solvent extraction.

3.2 Grinding of the Fuel Elements

A specially designed roll mill is required for the grinding of the brittle graphite. The optimum angle for feeding fuel into the roll mill is about 12°.[32] A large amount of fine particles is undesirable in the filtration step. By proper

selection of grinding conditions and type of mill, it should be possible to have a very narrow range of particle size distribution. Techniques such as anodic disintegration[4,31] could possibly be used before the grinding step to separate the fuel particles from the bulk of the graphite matrix. Unirradiated elements gave a good filtration rate,[32] but it is not yet known how irradiated graphites will behave in the washing and filtration steps.

3.3 Dissolution of Ground Product--Oxide Particles

Dissolution of oxide particles was discussed above. Irradiated sol-gel particles dissolve more rapidly in Thorex reagent than unirradiated particles.[33,34] There have been no studies on the dissolution of ground fuel elements containing oxide particles.

3.4 Leaching of Ground Product--Carbide Particles

Ferris and Bradley studied the reaction of uranium carbides with nitric acid in the temperature range of 25 to 120°C.[35,36] The carbides are completely passive in boiling 0.001 to 0.5 M HNO_3. In 4 and 16 M HNO_3 the carbides give uranyl nitrate, soluble organic acids, NO_2, NO, CO_2, and traces of N_2O. The main nitrogen oxide from 4 M HNO_3 was NO; whereas NO_2 was the main product from 16 M HNO_3. Between 50 and 80% of the carbide carbon was converted to CO_2. The remaining carbide carbon was converted to oxalic acid (0 to 11%), mellitic acid (2 to 9%) and unidentified highly substituted aromatic compounds. Contrary to the hydrolysis in water, no hydrogen, CO, or hydrocarbons were formed. The concentration of CO_2 in the gas increases with increasing reaction time. Pauson et al.[38] found about the same CO_2 concentration and, on the average, about the same reaction products. The stoichiometry and kinetics of the reactions of uranium carbides with nitric acid are not yet fully understood. Pauson et al. postulate a reaction of the type $UC + 2O \longrightarrow UO_2 + C$. Ferris and Bradley think that because of the UC crystal structure (no C—C bonding, closest C—C distance 3.5 Å) that there could be free radicals such as HCO, CO,

and CH_2 which polymerize.

Goode et al.[34] dissolved irradiated (6,000 and 20,000 Mwd/tonne) UC in 60% HNO_3 (320% excess) to achieve a product that was 0.47 M $UO_2(NO_3)_2$ and 11 M in HNO_3. About 56% of the carbide carbon was volatilized as CO_2, and about 8.5 moles of HNO_3 were consumed per mole of UC dissolved. There is less information about the dissolution of the thorium carbides. Imai et al.[37,39] dissolved ThC_2 at 25 and 80°C and ThC at 25 and 100°C in 0.5 to 15 M HNO_3. At 25°C, 90% of the ThC_2 was dissolved in the range of 0.5 to 1 M HNO_3. The solubility of the dicarbide at 25°C reached a minimum at 6 M and increased with increasing HNO_3 concentrations up to 50% of a 1-gram sample in 30 ml of acid in 24 hr tests. The solubility in 8 M HNO_3 increases with an increase in temperature from 25 to 80°C, from 19% to 99%. Above 4 M HNO_3, a gray solid collodial precipitate, perhaps carbon, was formed. This reaction may be of the type $ThC_2 + 4HNO_3 \rightarrow Th(NO_3)_4 + (CH_2)_n$. Or,

$$ThC_2 + 4HNO_3 \rightarrow ThO_2 \cdot xH_2O + 4NO_2 + 2C,$$

$$ThO_2 \cdot xH_2O + 4HNO_3 \rightarrow Th(NO_3)_4 + 4H_2O.$$

In 24-hr tests, ThC was soluble at 25°C in 1 to 11 M HNO_3 to the extent of 15.5%.[39] With increasing concentration above 11 M, the solubility increases to 80%. Precipitation of the colloidal carbon is seen above 12 M HNO_3 together with the formation of NO_2. The reactions involved may be

$$ThC + 4HNO_3 \rightarrow Th(NO_3)_4 + (CH_2)_n + H_2 \ (<2M\ HNO_3)$$

$$ThC + 4HNO_3 \rightarrow ThO_2 \cdot xH_2O + 4NO_2 + C$$

$$ThO_2 \cdot xH_2O + 4HNO_3 \rightarrow Th(NO_3)_4 + 4H_2O.$$

Studies in the Dragon project[31] have shown that uranium and thorium can be recovered quantitatively from ground uranium/thorium particles coated with pyrolytic carbon-silicon carbide-pyrolytic carbon (triplex coating). The particles were hydrolyzed after the grinding step with steam or 4 M HNO_3 and then leached with 13 M HNO_3. The hydrolysis step makes the addition of HF to the leachant unnecessary.

Grind-leach processing studies are still in the laboratory and bench-scale stage, and, hence, are much less advanced on a technical scale than the burn-leach process. The optimum conditions for the dissolution of the ThC_2-UC_2 elements are not yet known. The soluble organic products which are present after dissolution of the carbide particles perhaps will cause problems in the following solvent extraction step, and will, therefore, probably have to be degraded prior to solvent extraction.[31,34] The correlation between carbide carbon and soluble organic acids is not yet well known. Fuel elements with oxide particles have not yet been examined. Grinding and dissolution of irradiated fuel elements may cause new problems, especially in the filtration step. Also, we can get new facts about fission product retention in the graphite only by experiments with irradiated fuel. If it is possible to vent the off-gases from the burn-leach process, after appropriate dilution, directly to the atmosphere, or, if it would be possible to separate the rare gases from the off-gas, the burn-leach process has fewer technical disadvantages than the grind-leach process.

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