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MASTER

**DESCRIPTION OF THE PILOT PLANT FOR THE HEADEND
REPROCESSING OF UNIRRADIATED ROVER FUELS**

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A B S T R A C T

A non-radioactive pilot plant facility for the headend processing of graphite matrix nuclear rocket (Rover) fuel is described. The graphite matrix is removed by burning in an inert alumina bed fluidized with oxygen; the uranium is converted to the nonvolatile oxide, U_3O_8 , and separated from the bed by elutriation from the burner. Solids scrubbed from the off-gas are dissolved in nitric acid, producing a solution suitable for solvent extraction of the uranium.

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SUMMARY

A pilot plant facility for developing a continuous headend process for recovery of uranium from Rover fuel has been installed at the Idaho Chemical Processing Plant (ICPP). The process involves burning the fuel in a two-stage fluidized bed burner to remove the graphite matrix; separating the nonvolatile U_3O_8 (produced by combustion) from the alumina bed by elutriation; scrubbing the elutriated solids from the off-gas; and dissolving the U_3O_8 in nitric acid to produce a solution suitable for solvent extraction of the uranium.

Preliminary studies in the pilot plant facility have shown that: (1) approximately 98 percent of the graphite charged to the fluidized bed can be burned, (2) greater than 99.9% of the uranium can be recovered from the graphite matrix, and (3) a negligible amount of the bed material is elutriated with the U_3O_8 from the burner.

I. INTRODUCTION

The immediate need for a process to recover uranium from graphite fuels has been precipitated by the large quantities of spent fuel generated from the static testing of the Rover Nuclear Rocket engines. Basically, the Rover fuels consists of uranium dicarbide microspheres, coated with pyrolytic carbon, and dispersed in a graphite matrix. Separation of the uranium from the massive graphite matrix is the first step in the reprocessing of Rover fuels. The pyrolytic carbon coating and the bulk graphite can be removed relatively easily by combustion in an oxygen atmosphere. Because of the magnitude of the heat generation rates, a fluidized bed with its excellent heat transfer characteristics is an ideal medium in which to carry out the highly exothermic reaction without fusing the metallic oxide products.

Process development studies, initiated at the Oak Ridge and Brookhaven National Laboratories⁽¹⁾⁽²⁾, are continuing at the Idaho Chemical Processing Plant⁽³⁾⁽⁴⁾⁽⁵⁾. In the headend process under development at ICPP, U_3O_8 particles are separated from the alumina bed by elutriation from the burner. The U_3O_8 particles are then scrubbed from the off-gas and dissolved in nitric acid.

Peach Bottom and Ft. St. Vrain High Temperature Gas Cooled Reactors (HTGRS) use a fuel similar in composition to the Rover fuel. Therefore, much of the technology resulting from the development studies using Rover fuel may also be applied to the design of a demonstrational facility for reprocessing HTGR fuels.

II. PILOT PLANT FLOWSHEET

The flowsheet of the Graphite Fuels Pilot Plant is shown in Figure 1. The pilot unit is located in the High Bay Chemical Engineering Laboratory at ICPP and consists of a burner and a leacher module. Components of the burner module are: (1) a fluidized bed for burning the fuel, and

FIGURE I

(2) a dry collection system for separating particulates (mainly U_3O_8 product) from the off-gas. The leacher module consists of: (1) a wet (HNO_3) scrubbing and off-gas cleanup system, (2) a concentric tube leaching vessel for dissolution of U_3O_8 in nitric acid, and (3) a fines generator for producing a U_3O_8 dust laden gas stream simulating the effluent from the burner. Collection of dry or wet uranium oxide, and independent operation of either module, are possible.

During operation, the graphite fuel is introduced into the first-stage fluidized bed through a charging tube located in the second stage. The reactant gas (oxygen and/or air) fluidizes the alumina bed which serves to transfer the heat generated during the graphite combustion. Normal operating temperature for combustion, $700^\circ C$, is maintained by cooling air circulating between the shroud and the burner.

Particles of unreacted graphite or uranium dicarbide, released from the fuel surface in the first-stage bed, are essentially completely reacted in the second-stage bed. Additional oxygen for the reaction is preheated in an annulus surrounding the fuel charging tube before being introduced at the bottom of the second stage. Baffles, in the upper stage, serve to: (1) increase the residence time of reacting particles, (2) prevent the formation of large gas bubbles, and (3) reduce the amount of bed material elutriated. Significant reactions occurring within the fluidized bed are:

The particulate matter (U_3O_8 and Nb_2O_5) in the off-gas from the burner may be collected in the dry or wet product collection systems.

For dry collection, the solids are collected by passing the off-gas through sintered metal filters. During wet collection, the solids are scrubbed from the off-gas by nitric acid prior to dissolution.

III. PILOT PLANT EQUIPMENT

A. Fluidized Bed Burner

The fluidized bed burner is a two-stage unit as shown in Figure 2. The inert bed material normally used is -40 +70 U.S. standard mesh refractory grade alumina. Fuel is charged into and ignited in the lower stage where a combination of thermal stresses and mechanical attrition causes small particles of graphite to be released from the intact fuel elements. These graphite particles continue to burn in the oxygen-rich fluidizing gas of the upper stage until they are either completely reacted or elutriated from the bed.

The lower stage is constructed of a 4'-6" long section of 4-inch stainless steel (Type 310) pipe fitted with flanges at each end. A mating flange and cap with a 1/8-inch thick sintered stainless steel disc placed between the flanges at the bottom serves as a combination fluidizing gas distributor and bed support plate. Fuel rods are supported on a grid located six inches above the gas distributor.

The upper stage consists of a 9'-2" long section of 6-inch (Type 310) stainless steel pipe connected to the lower stage by a flanged reducer. A charging tube and baffle assembly, contained in the upper stage, is designed for fuel charging and preheating of the oxygen introduced into the upper stage. The core of the charging assembly is a 1-1/4-inch diameter fuel charging tube. A 2-inch diameter tube concentric with the fuel charging tube provides an annular space for preheating the oxygen introduced at the bottom of the second stage. Two types of baffles are used in the upper stage as shown in Figure 2. Deflection baffles, located on six-inch centers from the top flange, reduce the amount of bed material (Al_2O_3) carried over with the product (U_3O_8) by providing a tortuous path for solids leaving the burner. The second type of baffle is made of 1/16" perforated stainless steel discs.

GRAPHITE BURNER AND BAFFLED CHARGING ASSEMBLY

FIGURE 2

Nine of these baffles, spaced at nine-inch intervals and welded to the two-inch tubing, reduce slugging by breaking up large bubbles of gas.

Heat for burner startup is supplied to both stages by external electrical resistance elements (2750 watts at 240 volts). Four of these elements surround the lower stage burner, and two vertical banks of six elements each surround the lower 6 feet of the upper stage burner. Stainless steel shrouds surrounding the electrical elements of both stages act as heat reflectors and provide an annular space for cooling air flow. Cooling air is supplied by a blower mounted at the bottom of the burner module. The entire burner is insulated with high-temperature, magnesia insulation (two inches thick on the lower stage, three inches thick on the upper stage).

Samples of the fluidized bed may be taken from five different bed heights, 3", 4'-3", 5'-8", 8'-6", and 12'-0". Each sample line extends from the burner to the sampling glove box located beneath the burner. A pneumatically operated ball valve controls the flow in each sample line. These valves are manipulated with air actuators from inside the glove box as shown in Figure 3.

B. Solids Collection Pot

The solids elutriated from the burner may be collected by diverting the off-gas stream to the solids collection pot shown in Figure 4. Solids are removed from the gas stream as it passes through two sintered metal filters located inside the collection pot. Four pneumatically operated valves mounted above the collection pot form a filter blow back system. Each filter is cleaned independently by blocking the off-gas flow through the filter (see Figure 1) and applying a pulse of air to the inside of the filter.

C. Fines Generator

The fines generator, designed to produce a U_3O_8 dust-laden gas stream simulating off-gas from the burner, consists of a variable speed screw-type feeder attached to a two-inch glass tee. Air enters the bottom of the glass tee where it mixes with solids (U_3O_8) metered in by the screw feeder. The suspended dust is carried in the gas stream from the top of the glass tee to the gas scrubbing system in the leacher module.

SAMPLING GLOVE BOX

Figure 3

SOLIDS COLLECTION VESSEL

Figure 4

D. Glove Boxes

To prevent the spread of alpha contamination into the laboratory, two glove boxes maintained at subatmospheric pressure are used for all sampling and fuel handling. Doors opening directly into the glove boxes are provided for material transfers. An air velocity of at least 150 ft/min is maintained through the door opening by an exhaust blower. Exhaust air from the glove boxes passes through a prefilter and an AEC-type high-efficiency filter before being discharged to the atmosphere.

Full length rods may be charged to the first-stage burner from the charging glove box located on top of the burner module. A 1-1/4-inch schedule 40 pipe connects the fuel charging port in the floor of this glove box with the fuel charging tube extending through the top flange of the upper stage. Two pneumatically operated ball valves located in this line form an air-lock into which one full length fuel rod can be charged. Alumina is introduced into the burner by gravity transfer from the upper glove box through a 5-liter vessel located between the glove box and the top of the burner. Ball valves located above and below the 5-liter vessel are used to form an airlock when transferring alumina to the burner.

The sample glove box, as shown in Figure 3, contains: (1) the sampling valve actuators, (2) the dry solids collection pot outlet and drain valve, (3) the burner sample line outlets, and (4) a prefilter for the glove box exhaust system.

E. Off-Gas Cleanup System

The components of the off-gas cleanup system (shown in Figure 5) are: (1) a venturi scrubber, (2) a single-stage cyclone, (3) a York mesh demister, (4) a steam ejector, (5) two steam condensers, (6) an off-gas heater, and (7) an AEC-type high-efficiency filter. Solids elutriated from the burner are contacted with nitric acid in the venturi scrubber. The liquid is de-entrained from the off-gas in a cyclone and gravitates into a leaching tank. The cyclone has a 1-1/2-inch diameter barrel having the general proportions of the simple cyclone presented in the Chemical Engineer's Handbook⁽⁶⁾. Mist contained in the effluent

OFF-GAS SCRUBBING SYSTEM

Figure 5

gas from the cyclone coalesces in the York Mesh demister and drains into the leaching tank below the cyclone. Off-gas from the demister passes through a steam jet ejector to raise the off-gas pressure above 12.3 psia (local atmospheric pressure). Normally the steam condensate from the off-gas is diverted to the cold waste drain, but it may be diverted to the leaching tank if the condensate contains a significant amount of uranium. Finally, the off-gas stream is heated above its dew point in a section of pipe wrapped with heating tapes before passing through a high-efficiency filter into the building off-gas system.

F. Off-Gas Sampling System

A particulate filter located in the charging glove box permits diversion of the full flow of off-gas from three locations in the off-gas cleanup system [(1) before the venturi scrubber, (2) after the York Mesh demister, and (3) between the condenser outlet and the absolute filter] through the filter. The efficiency of the off-gas cleanup system is determined by dividing the weight of solids collected on the sample filter per unit time by the weight of solids entrained in the burner effluent gas per unit of time.

G. Leaching Tank

The uranium dissolution tank is a steam jacketed pipe (15 feet long and 4 inches in diameter) with a pyrex glass pipe (18 inches long and 4 inches in diameter) flanged to the bottom. A two-inch O.D. tube welded to the top flange and centered inside the 4-inch pipe extends to the top of the glass section. Liquid enters the top of the tank, flows down the inside of the two-inch tube and then up through the annulus to the overflow line near the top of the vessel. Undissolved solids which settle out can be drained through a ball valve located in the bottom of the glass section. Liquid in the tank is heated by steam in the jacket surrounding the tank. Product solution from the leaching tank is collected continuously in two 30-gallon stainless steel drums.

H. Instrumentation and Control

Miniature instruments and multi-point recorders are used throughout the system. Pressure drops across the sintered-metal support plate, the fluidized bed, the filters in the dry collection vessel, and the

venturi scrubber are recorded continuously. Pressure drop indicators are located across the absolute filters. Bed level pressure probes are located at four bed heights--3'-2", 4'-8", 6'-2", and 7'-2" above the bottom of the second stage.

During dry collection of the product, the desired burner pressure is maintained by a control valve located in the process line downstream of the sintered metal filters. When the wet scrubbing system is operating, constant burner pressure is maintained by controlling the amount of steam entering the steam jet. This method of pressure control eliminates the need for a control valve in the process line and reduces the quantity of steam required.

The fluidizing gas flow rate to each stage of the burner is measured with an integral-orifice, differential-pressure, transmitter. The composition of the inlet fluidizing gas is controlled by using pure oxygen to maintain constant pressure upstream of the orifice while metering in the desired amount of air through a rotameter.

Electrical power to the heating elements surrounding both stages of the burner is normally controlled manually; however, automatic control is possible for the second-stage heaters.

Temperatures throughout the system are continually recorded on two multipoint temperature recorders.

IV. AUXILIARY APPARATUS

A. Two-Stage Glass Column

A glass column, identical in dimensions to the two-stage graphite burner, was constructed to provide a visual display of the action of the fluidized bed. The behavior of the bed material, the effects of baffles on the particle and gas movement, and the action of fuel pieces can be readily observed. The quantitative effect of the baffles on the residence time of elutriable reacting particles is useful in the scale-up to a plant size unit. Studies of the action of the fuel pieces within the fluidized bed have proved useful in detecting inadequacies in the design of the present fuel charging system.

B. Miniburner

Shown in Figure 6 is a small scale fluidized bed unit for making scoping studies. Many investigations can be made more rapidly and at a lower cost in this unit than in the large pilot plant unit. The miniburner is a single-stage unit (2-inches in diameter and 12-inches high). Product (U_3O_8) can be elutriated to a dry collection vessel or can be retained in the burner by an internally mounted sintered metal filter. Fluidizing gas is raised to an operating temperature of $700^{\circ}C$ by six, 250 watt, tubular elements, located in the heater box at the bottom of the burner. The bed temperature is controlled by passing cooling air between a shroud and the burner.

V. RESULTS OF PRELIMINARY FEASIBILITY STUDIES

Preliminary studies in the two-stage fluidized bed burner have demonstrated the feasibility of separating the uranium from the massive graphite by a fluidized bed process. Subsequent reports will be issued for specific runs, giving detailed objectives, data, and results. This descriptive report (IN-1181) will be referenced in those reports, and only future changes in the flowsheet will be described. The preliminary results are as follows:

1. Separation of the uranium from the graphite matrix by burning in a two-stage fluidized bed is feasible.
2. Approximately 98% of the carbon in the fuel elements is converted to volatile oxides of carbon in the burner.
3. Essentially all of the particulate U_3O_8 can be separated from the alumina bed by elutriation.
4. Fuel rods can be charged through a charging tube in the upper stage into the lower stage.
5. Greater than 99.9 percent of the uranium can be recovered using the wet off-gas cleanup system.

MINIBURNER

Figure 6

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