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FLUIDIZED-BED COMBUSTORS USED IN HTGR FUEL REPROCESSING*

by

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ABSTRACT

High-Temperature Gas-Cooled Reactors utilize graphite-base fuels. Fluidized-bed combustors are being employed successfully in the experimental reprocessing of these fuels. This paper presents a general discussion of the reprocessing method and describes the two types of fluidized beds being used.

INTRODUCTION

The High-Temperature Gas-Cooled Reactor (HTGR), as developed at Gulf General Atomic, is a helium-cooled, graphite-moderated reactor. The fuel in an HTGR consists of fissile microsphere particles containing U-235, recycle microsphere particles containing U-233, and thorium fertile particles contained in a hexagonal fuel element, shown in Fig. 1. The HTGR fuel recycle operation consists of shipping spent fuel to a recycle facility, reprocessing the fuel to recover the U-233 and U-235, refabricating the U-233 and U-235 into recycle fuel, shipping the refabricated fuel from the recycle facility to the reactor, and ultimately storing the radioactive fission product wastes.

The fuel reprocessing sequence starts with the head-end operation shown in Fig. 2, in which the fuel in the HTGR fuel element is separated from the graphite body by crushing and fluidized-bed burning. Subsequent head-end operations separate particles containing U-235 from ash containing U-233, thorium, and fission products. The metal oxide ash is dissolved to create a solution of uranium, thorium, and fission products; the silicon-carbide-coated U-235 is the residue. The U-235 is separated mechanically, and the uranium and thorium are recovered individually from the fission products. The recovered U-233 and thorium are stored for reuse as fuel. The radioactive wastes are disposed of in appropriate storage facilities.

HEAD-END REPROCESSING

Head-end reprocessing for HTGR fuel consists of a crush-burn-leach process.⁽¹⁾ Fuel element size reduction is the first step in head-end reprocessing. Two major criteria govern this step: (1) the fuel must be crushed to a suitable size for maintaining fluidization quality in the

Fig. 1. HTGR fuel element

Fig. 2. Head-end reprocessing simplified flow diagram

fluidized-bed burners, and (2) the crushing system must minimize fuel particle breakage to prevent undesirable crossover of fissile and fertile product uranium.⁽²⁾

A three-stage crushing system has been adopted for the reprocessing plant, based on the experimental testing of commercially available equipment using full-sized fuel elements. This crushing system is presently being tested.

Primary reduction is done in a large, overhead eccentric jaw crusher; secondary reduction in a small, overhead eccentric jaw crusher; and tertiary crushing in a double-roll crusher. The tertiary crusher product, nominally minus 3/16 in., is pneumatically conveyed to the fluidized-bed burner feed hoppers.

Crushed fuel is fed to the top or base of a continuous, exothermic* fluidized-bed burner, shown in Fig. 3, by an auger feeder. The feed rate is automatically controlled by the off-gas carbon-monoxide concentration, which has been shown to be proportional to the graphite surface area exposed in the bed.

Both the crushed graphite and the silicon-carbide-coated fissile particles serve as the fluidizing media. The heat generated by burning is removed by forced-air cooling in a clamshell jacket surrounding the burner and an off-gas heat exchanger. The fluidizing gas fed to the burner is oxygen with a small amount of inert gas (i.e., CO_2 , N_2), and the flow is automatically controlled to maintain the bed temperature. The burner product removal rate is automatically controlled by the bed pressure drop, which is proportional to the bed weight.

The burner off-gas with entrained fines is passed through a cyclone separator and a sintered metal filter for fines removal, before being

*The term exothermic is used to describe the burner that generates sufficient heat to maintain operating temperature.

Fig. 3. Exothermic fluidized bed burner

cooled and proceeding to off-gas treatment. Off-gas treatment removes the fission products including noble gases before release to the environment. Fines are presently being recycled to the burner in the experimental program.

If the exothermic burner is operated with top feed and no fines recycle, the elutriated fines from the burner (both TRISO/TRISO and TRISO/BISO flow-sheets) are also added to the feed stream for the last burning step. This mixture now constitutes the feed material for the endothermic* fluidized-bed burner.

The exothermic burner product is fed to a batch-operated endothermic fluidized-bed burner, shown in Fig. 4, where the remaining graphite is burned and the thorium and uranium-oxide kernels are exposed. The silicon-carbide-coated fissile particles serve as the inert fluidizing media. The feed stream to the endothermic burner will not sustain exothermic burning to the low carbon level required in the subsequent processing steps. The burning in the endothermic burner, therefore, proceeds from exothermic conditions, with the heat removed from a clamshell surrounding the burner, to endothermic conditions, with heat supplied by resistance heaters located in the clamshell. The off-gas from the burner is treated in the same manner as that from the exothermic burner. The product from the endothermic burner is pneumatically conveyed to the leaching system.

The thorium and uranium oxides are dissolved in acid thorex ($13 \text{ M HNO}_3 - 0.05 \text{ M HF} - 0.01 \text{ M Al(NO}_3)_3$) in a steam-jacketed cylindrical vessel with gas sparge mixing. This leaching vessel is run as a refluxing, batch leacher.

The insoluble silicon-carbide-coated fissile particles and the unburned carbon must be separated from the mother liquor before the solution can be fed to the solvent extraction system for uranium purification and thorium

*The term endothermic is used to describe the burner that requires heat input from a furnace to maintain operating temperature.

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Fig. 4. Endothermic fluid bed burner

recovery. A centrifugal separator receives the entire slurry from the leacher. Solids retained on the centrifuge screen are washed with fresh leach solution, which becomes the leach solution for the next batch of solids from the endothermic burner. The washed solids from the leacher are then air-dried and transferred to a screen classifier, where the fissile particles are separated from the silicon-carbide hulls. The waste solids are processed as wastes, and the fissile particles are stored for later processing, by a method similar to that for the fertile particles, to recover and purify the uranium.

The clarified leach solution is evaporated and steam-stripped to an acid-deficient condition for use as feed to an acid thorex^(3,4) extraction process. The acid thorex solvent extraction process is used for the decontamination and purification of the U-233 and thorium and for the separation of the U-233 and thorium from each other.

FLUIDIZED-BED BURNERS

EXOTHERMIC FLUIDIZED-BED BURNERS

Figure 3 depicts the exothermic fluidized-bed burner presently being used in the experimental program. Exothermic burners with both 4-in. and 8-in. diameters are being used; construction of a larger burner is planned for early next year. The large burner will become the full-sized commercial plant test unit. Preliminary nuclear criticality calculations have shown that this burner can be about 16 in. in diameter.

OPERABILITY

The exothermic burners have been operated on a routine basis for the last 18 months. Startup is initiated by heating a charge of coke (1200 g for the 8-in. and 400 g for the 4-in.) to ignition temperature (700°C) with a carbon monoxide-oxygen gas mixture. This gas mixture is introduced into the burner with a standard cutting torch that is ignited by two spark plugs. After the coke is ignited, fluidizing oxygen and the graphite-base feed are introduced.

The steady-state bed properties of the exothermic burners are listed in Table 1; the bed contains 2% to 5% burnable carbon. Steady-state operation is achieved for experimental purposes in about 4 hr by adding the estimated steady-state bed composition directly to the burner immediately after start-up. Product removed after 4 hr is also near steady-state and exhibits about the same properties as the bed. The present feeding method is a variable-speed auger controlled automatically by the carbon-monoxide concentration in the off-gas. The nominal off-gas concentrations are 1% to 3% carbon monoxide, 0% to 6% oxygen, and 60% to 95% carbon dioxide. This composition is a function of the inert fluidizing gas diluent (i.e., air versus CO₂). Product removal rate is automatically controlled using the pressure drop across the bed and regulating a variable-speed drive motor on the product removal auger. Bed temperature is automatically controlled by regulating the fluidizing oxygen supply to the bed. Temperature profiles, off-gas compositions, bed and filter pressure drops, and mass flows of important streams are monitored continuously.

Fluidization quality has been difficult to define. Normal operation is with a well-mixed bed that occasionally slugs. Distributor plates are not presently being used but will be fully tested in the near future. At present, operation is with a cone base and a ball check valve. Preliminary tests of perforated plates, bubble caps, and sintered metal screens were all successful to some extent, and the beds appeared to maintain good fluidization quality.

Feed

The feed to the burners is presently defined as minus 3/16-in. graphite-based material. This feed size was established by gradually increasing the size from minus 1/16 in. to minus 1/4 in. Poor fluidization occurred with minus 1/4-in. feed, as witnessed by local "hot spots" in the bed. Returning to minus 3/16-in. feed eliminated this problem. Table 2 shows the average properties of the exothermic burner feed.

TABLE 1
AVERAGE BED PROPERTIES
FOR EXOTHERMIC BURNERS

Burnable Carbon: 3.0%
Average Particle Size: 590 μ

<u>Size</u>	<u>Wt % of Total Sample</u>
-3/16 in. + 1/8 in.	0
-1/8 in. + 869 μ	0.2
-869 μ + 550 μ	67.9
-550 μ + 420 μ	4.9
-420 μ + 375 μ	0.2
-375 μ + 250 μ	1.0
-250 μ + 125 μ	5.5
-125 μ + 74 μ	4.3
-74 μ + 44 μ	2.8
-44 μ	13.2

TABLE 2
AVERAGE PROPERTIES OF EXOTHERMIC BURNER FEED

Top Density: 1.25 g/cm³
Bulk Density: 1.08 g/cm³
Angle of Repose: 35°
Average Burnable Carbon: 80%
Average Particle Size: 854 μ

<u>Size</u>	<u>Wt % of Total Sample</u>	<u>% Burnable Carbon in Fraction</u>
-3/16 in. + 869 μ	64.6	100
-869 μ + 550 μ	25.0	21
-550 μ + 420 μ	1.5	78
-420 μ + 375 μ	1.5	97
-375 μ + 250 μ	1.5	98
-250 μ	5.9	98

Heat Transfer

The heat transfer problem encountered is somewhat different from that occurring in most fluidized-bed work; because of nuclear criticality considerations, the cooling medium is limited to air. Figure 5 shows the heat balance for a typical 8-in.-diameter exothermic burner run. The overall heat transfer coefficients for the off-gas heat exchanger and clamshell cooler are also shown.

Fines Recycle

One of the major problem areas in exothermic burner operation is the burning of fines that have elutriated from the burner. Since burning efficiency and radioactive hot-cell constraints play a major role in the process, work has focused on burning the fines by recycle to the bed rather than in a separate fines burnup cell.

Fines carryover has been about 22% of the burn rate when the furnace has operated at normal conditions, i.e., at a burn rate of approximately 200 g/min and a superficial fluidization velocity of 3 to 4 ft/sec. Of these fines, about 98% is collected by a cyclone and 2% by a filter chamber. The elutriated fines are described in Fig. 6.

The present operating mode is to recycle the fines by blending them with the graphite-base feed stream. This composite is fed to the bottom of the fluidized bed, and the fines appear to burn successfully when steady-state is achieved.

The nominal burn rate for the 4-in. and 8-in. exothermic burners is about 33 g carbon/hr-ft², which corresponds to 50 and 200 g carbon/min, respectively. A burn rate of 50 g carbon/hr-ft² (corresponding to 75 and 300 g carbon/min, respectively) is planned for the 4-in. and 8-in. exothermic burners. Burn rates of 125 and 475 g carbon/min for the 4-in. and 8-in. burners (about 84 g carbon/hr-ft²) have been achieved for short periods.

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Fig. 5. Heat balance and heat transfer coefficients for a typical 8-in. exothermic burner run

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Fig. 6. Elutriated fines description

Operation at these high burn rates is not possible for long periods because of limitations with existing equipment. Future studies will be aimed at defining these values over long-term run conditions.

Scale-Up

The design considerations for the larger exothermic fluidized-bed burner include both a theoretical approach and scale-up factors from the 4-in. and 8-in. burners. To date, the problems encountered for scale-up have been in defining both a suitable transport disengaging height and exact heat transfer values. Although the theoretical and experimental predictions of heat transfer values in the area of the bed-wall-clamshell are in agreement, it is difficult to define the proper heat transfer coefficient in the transport disengaging height in which the overall heat transfer coefficient rapidly decreases with reactor height. These values will be determined experimentally in future experiments in which "sectioned" clamshell coolers will be utilized.

ENDOTHERMIC FLUIDIZED BED BURNERS

Figure 4 depicts the 4-in.-diameter endothermic fluidized bed burner presently being used in the experimental program. It is planned to convert the 8-in.-diameter exothermic burner to an endothermic burner by adding resistance heaters to the clamshell interior and moving the filter chamber to directly above the burner for future scale-up testing.

Operability

The endothermic fluidized-bed burner has been operated as a batch burner on a routine basis for the last 18 months and is presently being automated. The automation consists of the furnace temperature control loop and a burner control system. The burner control system is a repeating unit of (1) automatic temperature control (by regulating the fluidizing oxygen flow) and (2) a series of programmed events. The programmed events

control fluidizing gas flow, batch product dump valve, batch pneumatic feeder, and refluidizing oxygen flow until the automatic temperature control loop takes over and the sequence starts again. This control cycle is repeated automatically and provides a batch-continuous operation. Although the endothermic burner is similar to the exothermic burner, the fines are burned by containing them within the burner. The low fluidizing velocities used during the endothermic burn stage allow burning the bed to less than 1% carbon.

Feed

The feed to the endothermic burner is the product from the exothermic burner. The size distribution of this feed is highly variable, depending on the flowsheet being processed, but at all times it is easily fluidized. The average particle size of the feed varies from 200 to 400 microns for the various flowsheet varieties.

Heat Transfer

The burn rates achieved in the 4-in. endothermic burner are about one-half that of the 4-in. exothermic burner because the bulk of the heat transfer occurs in the transport disengaging height. Also, the operating period in the endothermic stage of burning is a slow burning process. Burn rates of 25 to 35 g carbon/hr-ft² are achieved during the exothermic burn period and 5 to 10 g carbon/hr-ft² in the endothermic burn period. An average burn rate of about 20 to 25 g carbon/hr-ft² or 20 to 25 g carbon/min is achieved during batch-continuous operation.

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