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THORIUM FUEL CYCLE DEVELOPMENT PROGRESS REPORT

AUGUST 1970

NO. 13

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CHEMICAL TECHNOLOGY DIVISION
METALS AND CERAMICS DIVISION

THORIUM FUEL CYCLE DEVELOPMENT PROGRESS REPORT

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II. REFABRICATION DEVELOPMENT

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SEPTEMBER 1970

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THORIUM FUEL CYCLE DEVELOPMENT PROGRESS REPORT

AUGUST 1970

No. 13

Compiled by

The Staff of the Chemical Technology and
Metals and Ceramics Divisions

INTRODUCTION

The thorium fuel cycle development at ORNL is directed almost solely at HTGR fuels. These fuels consist of large blocks of graphite containing coolant channels and fuel and blanket holes. The fuel and blanket are made of microspheres of uranium or thorium compounds separately, or of mixtures of them in a single microsphere. The microspheres are coated with layers of pyrolytically deposited carbon and in some cases silicon carbide. The microspheres are retained in the holes in the graphite blocks in a bonded state.

Development work on all aspects of HTGR fuel recycle is in progress at ORNL. In addition, a major recycle development facility, the Thorium-Uranium Recycle Facility (TURF), has been built at ORNL, and the Coated Particle Development Laboratory (CPDL) has been put into operation in Building 4508. TURF is intended to be used as a development facility for fuel recycle. The CPDL is for engineering development studies leading to design of the equipment to be used in TURF.

I. HEAD-END PROCESSING DEVELOPMENT

The objective of this program is to evaluate head-end processes for converting irradiated HTGR fuels to a form suitable for recovery and decontamination of the thorium and uranium by a solvent extraction process. Small samples of irradiated and unirradiated fuel are studied to determine effects of irradiation on fuel reprocessing steps and to correlate with metallographic studies. An important objective is the determination of the amount of breakage of particle coatings and the resultant amount of cross-contamination of the fertile Th-^{233}U and fissile ^{235}U components in alternative reprocessing steps. Mechanical systems are being developed for degrading the fuels and providing a material suitable for use in studies of the burn-leach steps using fluidized-bed or fixed-bed burners. The mechanical and burn-leach engineering development work is carried out using full-scale unirradiated fuel of the type to be used in PSC, and is designed to provide scale-up data for use in the design of pilot or full-scale processing plants.

1. Studies with Irradiated Fuels

(V. C. A. Vaughen, J. H. Goode, G. Davis)

Procurement of Irradiated Dragon Fuels

We have been in negotiations with the UKAEA for the procurement of 9 irradiated Dragon compacts of the $(Th/U)C_2$, ThC_2, UC_{10}, UC_{20}, and $(U/Th)O_2$ varieties, and 7 unirradiated controls for our head-end studies. We have not yet received either unirradiated or irradiated compacts. The paperwork for the irradiated Dragon fuel samples is still in the British Ministry of Transport. We are hopeful that the unirradiated compacts will be sent shortly.

2. Head-End Engineering Studies

(R. S. Lowrie)

The head-end engineering studies comprise two principal areas of investigation — "mechanical" and "burn-leach." The general approach followed has entailed the comminution of a fuel element to produce

suitable feed material for the burn-leach process. A number of changes have occurred in the overall HTGR program during the past year and some of them, such as the recent decision of GGA to use bonded fuel sticks instead of loose particles in HTGR fuel elements, has led to changes in our ideas of the mechanical equipment required to reprocess spent reactor fuel. The effects of these changes are reflected in the generalized head-end reprocessing diagram shown in Fig. 1.

2.1 Comminution Studies (R. S. Lowrie, C. H. Tipton)

Unirradiated graphite fuel blocks (1/6 of a fuel log) containing resin bonded fuel sticks with triso coated UO_2 and ThO_2 particles were crushed in a hammer mill using 3/4-in. and 3/8-in. grate spacings. The size distribution of the crushed product was determined by sieving (Table 1). No fuel particles were found in the +4 mesh or larger size fractions of material crushed using a 3/4-in. grate spacing or in the +12 mesh or large size fractions obtained with a 3/8-in. grate spacing. Apparently the increased time in the hammer mill freed any fuel particles adhering to the graphite in the +12 mesh fraction obtained when using −3/8-in. grate spacing so that the total amount of barren graphite (graphite free from any fuel particles) is about the same for either grate spacing (∿ 45%). Cost will determine the ultimate disposal plan for the slightly contaminated barren graphite waste.

2.2 Fluidized-Bed Burning (R. S. Lowrie, C. H. Tipton)

Burning tests were made to determine the effect of increasing oxygen concentration in the reagent gas on triso particle coating breakage. Operating conditions are shown in Table 2. No plugging phenomena or wall "hot spots" were observed. Further, oxygen utilization was > 95% during the major burning period and the free-flowing burner ash contained < 1% unburned graphite. Some evidence of sintered Al_2O_3 was noticed in the +20 mesh fraction of the burner ash from Run BT-16, probably due to the higher localized temperatures resulting from the use of 100% oxygen. The ThO_2 particles reported to the +35 mesh fractions, the UO_2 particles to the −35 +50 mesh fractions.

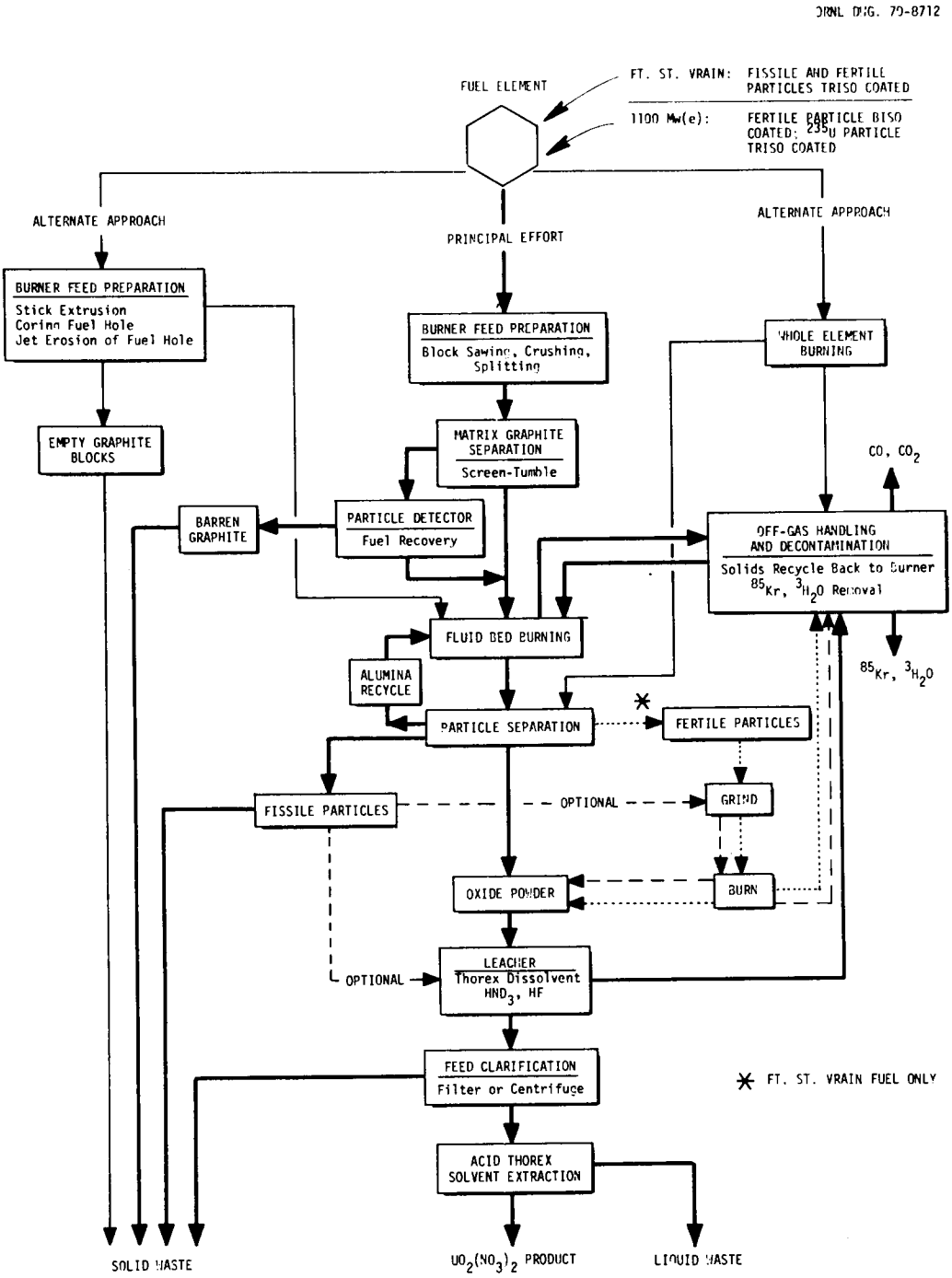

Fig. 1. Generalized Head-End Reprocessing Diagram.

Table 1. Size Distribution of Graphite Fuel Crushed in a Hammer Mill

Size Fraction	Hammer Mill[a] Product (wt %)			
	3/4-in. Grate Spacing		3/8-in. Grate Spacing	
+3/4 in.	2.2[b]	3.2[b]	--	--
–3/4 +1/2 in.	9.0[b]	12.2[b]	--	--
–1/2 +4 mesh	33.0[b]	30.8[b]	14.4[b]	23.4[b]
–4 +12 mesh	14.5	14.6	27.6[b]	19.83[b]
–12 +20 mesh	11.1	11.7	14.5	11.72
–20 +35 mesh	19.8	18.0	10.7	19.7
–35 +50 mesh	3.6	2.6	8.5	6.6
–50 +80 mesh	2.5	2.5	7.4	5.4
–80 mesh	4.8[c]	4.4[c]	16.7[c]	13.2[c]

[a]Type A - Jeffrey Swing Hammer.

[b]No fuel particles were found in these fractions.

[c]Contained only a few pieces of broken UO_2 and ThO_2 particles.

Table 2. Fluidized-Bed Burning Tests

Run No.	BT-14	BT-15	BT-16
Charge to Burner			
Alumina bed,[a] g	1600	1600	1600
Crushed fuel,[b] g	2000	2038	1835
Reagent	20% O_2 Air	70% O_2 30% N_2	100% O_2
Operating Conditions			
Superficial velocity, fps	1.25	1.25	1.25
Temperature, °C	750 ± 25	750 ± 25	750 ± 25
Burning time, hr	14.75	7.25	4.25
Burner Residue, g	1894	1954	1879

[a]–90 Mesh Norton RR Alumina.

[b]Product from hammer mill set to produce –3/4-in. product.

The percentages of particles with broken coatings are shown in Table 3. Decreased breakage of the UO_2 particle coatings from 6.09 to 2.9% as oxygen concentration increased from 20 to 100% was primarily due to the decreased time in the fluidized bed. Breakage of the ThO_2 particle coatings dropped from 6.9 to 3.6% as the oxygen concentration increased from 20 to 70%, then jumped to 11.3% with 100% oxygen. However, since pure oxygen reagent can lead to sintering of the bed, a 100% oxygen feed would probably not be used. If a 70% oxygen reagent is used, the overall coating breakage for both the crushing and burning step is 5.7% for UO_2 and 4.67% for ThO_2 particles, slightly less than the arbitrarily chosen value of 5% crossover of ^{233}U to the reject ^{235}U stream.

Table 3. Breakage of Particle Coatings in Fluidized-Bed Burning Tests

Run No.	Percentages of Particles with Broken Coatings		
	BT-14	BT-15	BT-16
U particles as charged	2.17	2.17	2.17
Th particles as charged	1.07	1.07	1.07
U particles in burner ash	8.26	5.7	5.4
Th particles in burner ash	7.97	4.67	12.37
U particles broken in test	6.09	3.53	2.97
Th particles broken in test	6.90	3.6	11.30

2.3 Flowsheet Calculations (R. S. Lowrie)

Material balance calculations are being made for reprocessing irradiated Ft. St. Vrain fuel through the head-end, solvent extraction, sol formation, and microsphere preparation steps of the thorium recycle flowsheet. They are based on the following assumptions:

1. 12 kg of $(^{233}U,Th)O_2$ microspheres will be produced per day. (After allowance is made for losses in the various fabrication process operations, it is estimated that there will be 10 kg of coated product.)

2. All of the ^{233}U required will come from reprocessing irradiated Ft. St. Vrain fuel elements.

The amount of uranium and fission products in the fertile particles is being calculated with the ØRIGEN code using the Ft. St. Vrain Reactor core characteristics supplied by GGA. Preliminary results indicate that it will be necessary to process ∿ 10 spent fuel elements per day (2-yr exposure in the reactor) to supply the necessary uranium.

2.4 Visit to GGA (R. S. Lowrie, V. C. A. Vaughen)

Topics of general interest were discussed during a recent trip to GGA. (This visit will be reported in a joint trip report.) The RTE fuel blocks and assembly were especially interesting. It appears now that the fuel blocks would be better divided into quarter sections by milling a slot on the outer diameter rather than by hacksawing. Some empty GGA reject 30-in. elements will probably be available for our tests.

II. REFABRICATION DEVELOPMENT

1. Particle Preparation

The fuel material of primary interest for ORNL recycle studies is the ThO_2-UO_2 particle, which has a thorium-to-uranium ratio of about 4.2. The uncoated fuel particles are to be microspheres 350 ± 100 μ in diameter, made by the sol-gel process. Fuel preparation includes development and demonstration of all process steps involved in making remotely the ThO_2-UO_2 microspheres. The steps include demonstrating reliable, remote processes for reproducibly mixing $Th(NO_3)_4$ and $UO_2(NO_3)_2$ solutions in the desired thorium-to-uranium ratio, preparing the mixed, stable ThO_2-UO_3 sol in concentrations exceeding 1 g-mole of oxides per liter, forming ThO_2-UO_3 gel microspheres, and converting them to dense ThO_2-UO_2 in good yield.

Experimental studies of processes and equipment for preparation of sols and microspheres are reported here. The present emphasis is on processes, procedures, and prototype equipment for preparation of oxide microspheres in the Thorium-Uranium Recycle Facility (TURF). Tests of flowsheets and procedures to be used for test materials preparation are also included.

1.1 Preparation of Test Materials (C. C. Haws)

Fired UO_2 spheres containing 234 g of uranium (93% ^{235}U) were delivered to supply an additional 200 g of 100 micron spheres for RTE fabrication. Cleanup was completed giving 29 g U as waste solution and leaving 7 g unaccounted for.

2. Fueled Graphite Fabrication Development

(F. J. Furman, W. H. Pechin, C. B. Pollock, J. M. Robbins, and J. D. Sease)

We are developing processes and equipment for the refabrication of HTGR fuel as detailed in the National HTGR Recycle Development Program Plan. The fuel consists of microspheres of thorium and/or uranium as the oxide or carbide, coated with multiple layers of pyrolytic carbon and silicon carbide. These particles are loaded into hexagonal graphite logs which contain both fuel and coolant holes.

Our work is divided into particle coating, particle handling and inspection, fuel stick making, and fuel element assembly. Last report period we concentrated on particle coating development. We continued the work on modifying the prototype coating furnace, experimented with coating furnace cone reclamation and began a series of tests to establish load limits of a 5-in.-diam coating furnace, paying particular attention to coating HTGR reference recycle particles with a minimum of soot inclusions and faceting.

2.1 Particle Coating

We are taking advantage of a forced shutdown of prototype coating furnace operations, necessitatated by modifications to the laboratory, to improve the coating furnace electrodes, particle unloading system, and exhaust system. The electrodes are being modified so that the contact pressure between the electrodes and the heating element can be increased to minimize the contact resistance. The unloading system is being modified to allow fluidization of the particles in the water cooled hopper to increase heat transfer. The soot filter on the exhaust system is being changed from a bag filter to a flat filter so that the geometry will be critically safe. In addition, the furnace pressure control valve

is being moved to the downstream side of the soot filter. With the valve on the upstream side of the filter, soot would deposit in the valve, restricting the exhaust flow.

We are attempting to devise a method to reclaim the furnace cones after coating operations with propylene have built up a pyrolytic carbon deposit in the throat of the cone. Initially, we attempted to deposit on the cone an anisotropic layer of pyrolytic carbon just prior to loading the particles to be coated. However, with the lowest gas flows possible with our present equipment, the anisotropic deposit formed above the throat of the cone. Not wanting to resort to devices to cause the coating gas to impinge on the area desired, we shifted our approach to applying a layer of graphite mold release on the cone. Since we applied the mold release by hand before the cone was placed in the furnace, we could control where the deposit was made.

We set the coating parameters to achieve a heavy coating on the cone. Visual comparison of the results showed that the pyrolytic coating did not adhere to the treated cone as strongly as to the untreated cone. However, in the transition region between the straight walls of the cone inlet and the cone itself, some deposit firmly adhered, possibly because of mechanically locking into cutting-tool marks. Although we were not able to remove this coating by hand, we expect to be able to remove it with a special tool rigidly mounted in a drill press. We are fabricating this tool now.

We are planning a series of coating runs to investigate coating quality. Specifically, we shall be attempting to minimize soot inclusions and faceting on HTGR reference recycle coatings. We shall vary the coating parameters and furnace loads, keeping the properties of the coatings within density, anisotropy, and

thickness specifications, and shall study the change in soot inclusions, metallography, and the change in faceting by a qualitative technique we are developing. Presently we are preparing large uniform quantities of buffer coated microspheres to use in this study.

2.2 Development of Bonded Beds of Coated Particles for HTGR Fuel Elements

We have continued to prepare small diameter UO_2 particles (100μm) for use in the Recycle Test Elements. We received 264 g of sol-gel UO_2 particles and, after processing, were able to use 135 g for coating. All of the particles have been coated with low density (1.2 g/cm^3) pyrolytic carbon and half of the particles have been coated with high density (1.85 g/cm^3) pyrolytic carbon. We anticipate that all of the particles will be ready for stick forming shortly.

III. TURF EQUIPMENT DESIGN AND ENGINEERING

(A. L. Lotts, J. W. Anderson, J. D. Sease, and J. W. Snider)

The objective of this part of the program is to design, construct, and to perform operational testing of three pilot plants to demonstrate recycle of HTGR fuels. These pilot plants are the Head-End Reprocessing Pilot Plant, Solvent Extraction Pilot Plant, and the Refabrication Pilot Plant. The Head-End Pilot Plant and the Refabrication Pilot Plant will be installed in Bldg. 7930 and the Solvent Extraction Pilot Plant will be installed in Bldg. 3019. The design of these pilot plants will include processing equipment, fuel material handling equipment, and modifications to the facilities of Bldgs. 7930 and 3019. These engineering efforts will include major processes and equipment involved in recycle Head-End, Solvent Extraction, Sol-Gel, and Refabrication, as well as Special Tasks such as waste and material handling and shipping. These are reported subsequently under their respective subsections.

Our general project work this report period was concentrated on revision of the National HTGR Recycle Development Program Plan and on the updating of the Project Schedule for various major subtasks.

1. Head-End Reprocessing Equipment

Design of the pilot plant will not get under way to any significant degree for some time; however, during the report period substantial effort was expended in preparing more definitive plans for designing, constructing, and operational testing of the pilot plant. Chemical flowsheets proposed for the pilot plant were updated. Some initial studies were made to better define the waste drain streams from the pilot plant and the requirements for receiving, handling, storing, and shipping materials to support pilot plant operation. This information was coordinated with details of the program plan to better define the activities of this subtask.

2. Solvent Extraction Equipment

Design of the Solvent Extraction Pilot Plant relates largely to design of offgas handling equipment and to additions to the Thorex Pilot Plant in Bldg. 3019. As is the case with the head-end equipment design, little of this design will be undertaken in the near future. During the report period efforts were directed toward preparing a better definition of the work required in this subtask. Work was initiated on updating flowsheets for this part of the recycle process.

3. Sol-Gel Equipment

This area of the program includes the particle preparation equipment to be installed in the Refabrication Pilot Plant. At the present time there is no design in progress. However,

updating of flowsheets for this part of the recycle process was initiated during this report period. Efforts were also directed toward preparing a better definition of the activities to be performed in this subtask.

4. Refabrication Equipment

Included in this subtask is the remainder of the processing equipment to be installed in the Refabrication Pilot Plant, not included in subsection 3 above. General work in this area during the report period included developing a better definition of the activities to be performed in this subtask.

4.1 Refabrication Equipment Design
(W. A. Pate, F. C. Davis, and D. D. Cannon)

A preliminary process flowsheet was prepared. This flowsheet shows information such as flow rate, equipment capacity, special instrumentation readout, and fuel particle quantities. These preliminary flowsheets were distributed for comment and will be finalized for issue when comments are resolved.

Discussions were held with personnel involved in the fuel stick fabrication development activity. These discussions were intended to familiarize design personnel with the present status of fuel stick fabrication development and to establish communication channels such that development information can feed back to the design effort. From these discussions, it was evident that several alternative fabrication approaches exist and should be exploited in keeping with the requirement for a relatively high speed, remotely operated process. Conceptual designs of proposed fuel stick fabrication equipment are being prepared at this time. These designs will ultimately be evaluated with project personnel to arrive at a design for equipment that will be fabricated and used in the laboratory development activities.

5. Special Tasks

5.1 Fuel Material Handling
(J. M. Chandler)

A brief study was conducted during the report period to better establish the requirement for fuel material handling with regard to receiving, handling, storing, and shipping fuel material in support of recycle pilot plant demonstrations. The results of these brief studies show that some additional equipment and certain facility modifications will be necessary in order to accommodate some material handling requirements in support of these subtasks. More definitive requirements in this area are being developed for incorporation into the HTGR Recycle Program Plan.

IV. MATERIALS IRRADIATION

1. HTGR Recycle Fuels Irradiation
(T. N. Washburn, R. B. Fitts, and A. R. Olsen)

The irradiation tests on the HTGR recycle program have two main objectives: (1) to provide irradiated fuel for head-end process studies, and (2) to provide irradiation proof tests of the products of coated particle process development for the Thorium-Uranium Recycle Facility. The test conditions of interest include fuel temperatures between 600 and 1300°C, burnup to 20% FIMA in the $(Th,U)O_2$ particles, and fast fluence exposures up to 8×10^{21} neutrons/cm^2.

The first two stages in this program were implemented this year. They are: (1) the start of accelerated burnup rate capsule irradiation and (2) irradiation of eight test fuel elements in the Peach Bottom Reactor.

1.1 Capsule Irradiation

(A. R. Olsen)

Fabrication of capsule components is continuing. During the report period we modified the capsule design for the "mockup capsules" required for each in-core test in the ETR. If this design modification is acceptable to the ETR we will be able to measure, with fission monitors, the fission rate at three locations along the capsule mockup axial centerline in the ETR Critical Facility at the start of each irradiation cycle. These measurements will be used to normalize our fission rate calculations, help to define the effectiveness of the thermal neutron shrouds, and permit us to observe the effects of changing core loadings from cycle to cycle.

1.2 Large Scale Irradiations

(R. B. Fitts)

The initial set of six RTE's has been inserted into the Peach Bottom Reactor. The reactor went to power on July 14, 1970 and has operated without incident since that date. The accumulated 42 days of irradiation (to August 25, 1970) is 14% of that planned for the low burnup elements and 4.2% of that planned for the highest burnup elements. Similar fuel elements, in a companion GGA test element series, are instrumented with thermocouples and data from these indicate that the RTE's are operating under the planned conditions.

It now appears that the remaining two RTE's will be loaded into the reactor in early 1971.

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