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THORIUM FUEL CYCLE DEVELOPMENT PROGRESS REPORT

JULY 1970

NO. 12

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ORNL-TM-3112

Contract No. W-7405-eng-26

CHEMICAL TECHNOLOGY DIVISION
METALS AND CERAMICS DIVISION

THORIUM FUEL CYCLE DEVELOPMENT PROGRESS REPORT

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I. HEAD-END REPROCESSING DEVELOPMENT

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II. REFABRICATION DEVELOPMENT

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W. D. Bond, J. W. Snider, J. D. Sease

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AUGUST 1970

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THORIUM FUEL CYCLE DEVELOPMENT PROGRESS REPORT

JULY 1970

No. 12

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 REPROCESSING DEVELOPMENT

(R. E. Blanco, W. E. Unger)

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)

1.1 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. We are ready to resume hot cell studies immediately upon their receipt. In the meantime we are analyzing the data from our studies with the other Dragon compacts we have had for some time, and are preparing a report on this work.

2. Head-End Engineering Studies

(C. D. Watson, R. S. Lowrie)

The head-end engineering studies are comprised of two principal areas of investigation — "mechanical" and "burn-leach." The general approach followed has entailed the mechanical dissection, by sawing, of full-sized graphitic blocks containing fuel, followed by comminution of size fractions suitable for the burn-leach process. However, the recent GGA decision to use bonded fuel sticks rather than loose particles as the reference form has necessitated changes in the approach.

Work on the head-end engineering studies program has resumed. Efforts during this report period continued on rewriting the National HTGR Recycle Program Plan to reflect changes which have occurred in the overall HTGR program.

II. REFABRICATION DEVELOPMENT

1. Particle Preparation
(R. G. Wymer - Coordinator)

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.

1.1 Sol and Microsphere Preparation Development (P. A. Haas)

Experimental engineering 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.

Preparation of Test Materials (C. C. Haws)

A batch of enriched urania sol prepared by the precipitation-peptization flowsheet is being formed into gel spheres, dried, and calcined to supply an additional 200 g of 100-micron UO_2 spheres for RTE fabrication. The fired spheres should be ready for delivery before the end of July.

Sol and Microsphere Preparation Development

A second revision was completed of the ORNL report draft for the demonstration runs for ThO_2-UO_3 sol and ThO_2-UO_2 spheres. This draft should be typed and ready for reviews by August 1.

1.2 Sphere-Forming Column Chemistry (W. D. Bond, J. W. Snider, P. A. Haas, C. C. Haws)

Our previous studies have shown that pH, surfactant concentration and water content of 2-ethyl-1-hexanol (2EH) are important variables in the microsphere forming process. We are presently examining methods for improving the control of pH and surfactant concentration during continuous operation of sphere-forming columns. Our present system for controlling the water content is adequate. For water content control, a portion of the used solvent is heated at 150-160°C to remove most of the water, and the desired water content is obtained by mixing this relatively dry 2EH with the remaining wet solvent.

In the past, sphere-forming columns have been operated continuously by making periodic additions of surfactants to offset the losses of surfactant caused largely by reaction of the nitric acid (extracted from the sol) with the surfactants during removal of water by distillation. In addition to reactions during distillation, a relatively small amount of reaction occurs in the forming column as evidenced by slow discoloration of the solvent when the forming column is batch operated without a distillation system. The pH or acid contents of the 2EH that have been employed in continuous operation have been mainly those governed by the amount of acid extracted from the sol which can subsequently be maintained in the 2EH with the distillation system operating. When higher acid contents are required, nitric acid must be added to the 2EH, and reactions with surfactants and 2EH are increased.

We are studying a total 2EH purification method in which the 2EH is recovered by distillation; the used surfactants are discarded, and the 2EH is recycled to the forming column after surfactants, water, and nitric acid are added. Studies are continuing on the effect of surfactant and water content, and the pH of 2EH on sphere forming using different sols in batch operated laboratory forming columns.

Purification of 2EH by Distillation for Recycle (W. D. Bond, C. W. Greene C. C. Haws)

The purification of 2EH by distillation from recycle 2EH has continued. Solutions of 2EH and surfactants that had been recycled in the UNOP sphere-forming column for several weeks were used as the feed source. The known added surfactant concentration of the feed solution was >5%. After saturation of the feed solution with water (2.5 w/o), 2EH and water was flash evaporated at 180° leaving the surfactants in the evaporator. Water was removed from the 2EH in a 160°C distillation step. The reclaimed 2EH contained less than 0.005 v/o surfactants. A material balance showed that 95-98% of the alcohol was recovered. We have purified approximately 100 liters of 2EH in our laboratory distillation unit, and will reuse this alcohol in sphere-forming studies in the UNOP column.

1.3 Microsphere Column Studies (P. A. Haas, C. C. Haws)

Studies are continuing on the effects of alcohol recycle systems on yield and quality of microspheres from the forming column. Large volumes of waste from the regeneration cycle of the ion exchange system have necessitated further laboratory studies aimed at reducing the waste volumes. It appears that a significant volume reduction may be possible.

2.0 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. The particles are bonded together into fuel sticks and these sticks 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 month we concentrated on the modifications to the prototype remotely operated coating furnace. These modifications are intended to correct design defects which were discovered during the operations of the last few months and also to move the equipment out of the way of forthcoming modifications to the laboratory.

2.1 Particle Handling

The prototype coating furnace control panel and part of the gas supply piping were in the way of the wall to be constructed between the prototype coating system and the plutonium laboratory being planned for the remainder of the room. The panel was moved and the piping was relocated during the last month. All electrical and gas systems have now been reconnected.

On the furnace itself the electrodes were found to be inadequately designed. These electrodes have been redesigned and material ordered for fabrication.

The exhaust system of the furnace, while operational, would present a criticality problem in TURF during operations with ^{233}U. In the

exhaust system the gas must first be filtered to remove soot. Previously we have been using a bag filter but this geometry of filter could potentially go critical if sufficient amounts of particles were blown into it and allowed to accumulate. To elminate this problem a small diameter bag or sock filter could be used but the maximum diameter recommended for a critically safe cylinder of ^{233}U solution is only 3.7 in., too small for even a sock filter. We have been able to design a flat filter which will meet the recommended thickness for a critically safe slab—1.7 in. This filter, with an effective filter area of approximately 9 ft^2, will also allow for the accumulation and removal of soot. The soot will be blown or vibrated off the vertical filter cloth into a horizontal tube. A piston will travel the length of the tube compressing and pushing out the accumulated soot. This filter is now being fabricated.

In addition to the above modifications to the prototype coater, we began an investigation of methods to reclaim the graphite furnace cones. These cones are quite expensive (approximately $90 each) and must be discarded after a few high density pyrolytic carbon coating runs. The pyrolytic carbon deposits on the cone as a hard, firmly attached layer which changes the coating characteristics of the system. Previous attempts to remove this layer have resulted in damage to the cone.

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

Additional coated particles and fuel sticks are needed for the Recycle Test Element (RTE) project to furnish the fuel for the RTE-3 and to provide fuel for the HTGR recycle capsules.

Approximately 102 in. of fuel stick type "g" (UO_2 + ThO_2) is required for the RTE-3. This represents eight stacks of bonded fuel sticks, each 12.82 in. long. In addition to the bonded sticks, we

are to furnish GGA with 500 g of loose coated UO_2 particles. The fuel stick requirement for the HTGR recycle capsules is listed in Table 2.1.

The UO_2 kernels have been received from the Chemical Technology Division. Other particles and materials are on hand so stick fabrication can now be started.

Table 2.1 Fuel Sticks[a] for HTGR Capsules

Combination	Fissile	Fertile	Number of Sticks Required[b]
a	$(Th-20\%\ U)O_2$	ThO_2	7
c	$(Th-33\%\ U)O_2$	ThO_2	7
f	UC_2 (TRISO)	ThC_2	9
g	UO_2	ThO_2	7

[a]Sticks 0.490 ± 0.010 in. diam × 2.125 ± 0.125 in. long loaded with U (93% enriched) at 0.260 ± 0.001 g/in.

[b]Includes three extra of each type for archive and cold reprocessing studies.

III. TURF EQUIPMENT DESIGN AND ENGINEERING

(J. W. Anderson, J. W. Snider, and J. D. Sease)

The objective of this part of the program is to design and construct equipment that will be used in TURF to demonstrate the recycling of HTGR fuels. Some TURF design tasks were resumed this month after a substantial period of inactivity due to fund restrictions. The design effort will include the major processes involved in recycle, head-end, solvent extraction, sol-gel, and refabrication

as well as special tasks such as waste handling and shipping. These are reported subsequently under their respective sections.

Our general project work this month was concentrated on revision of the "National HTGR Recycle Development Program Plan" and on the updating of the various project schedules.

1. Head-End Reprocessing Equipment

There will be relatively little equipment design related specifically to TURF equipment in the near future under this heading. However, some design decisions needed for critical path scheduling will be made, and some equipment design needed for development work will also be relevant to this section of the report from time to time. Such items will be reported here when appropriate. There is nothing of that nature this month.

2. Solvent Extraction Equipment

The equipment design needed for the solvent extraction process (Thorex Pilot Plant) is largely related to off-gas handling and to additions to the Thorex Pilot Plant. As is the case with the head-end equipment design, little of this design will take place in the near future, but some critical path decisions and off-gas handling equipment for development studies will be carried out, and these items will be reported as appropriate. There is nothing to report this month.

3. Sol-Gel Equipment

This is an area where TURF equipment design can and will proceed as soon as it is clear that such action is consistent with the overall Thorium Fuel Cycle Program. At the present time there is no design work in progress.

4. Refabrication Equipment

The objective of this task is to design, fabricate, and operate the refabrication equipment that will be installed in the recycle of HTGR fuel. The engineering design is broken down into conceptual design, detailed conceptual design, and detailed design and fabrication. This work was initiated this report period and has been confined to updating process and equipment flowsheets.

5. Special Tasks

Nothing to report.

IV. MATERIALS IRRADIATION

1.0 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) fabrication of eight test fuel elements to be irradiated in the Peach Bottom Reactor.

1.1 Capsule Irradiation

Two capsules are being prepared for irradiation of HTGR fuel in the Engineering Test Reactor (ETR). The capsules have been designed and the thermal neutron shielding material has been received from Bettis Atomic Power Laboratory. Fabrication of all capsule components has been initiated.

1.2 Large Scale Irradiations

Eight Recycle Test Elements (RTE's) are to be irradiated in the Peach Bottom Reactor. Six of these have been placed under irradiation in Peach Bottom; two are yet to be fabricated.

The UO_2 kernels necessary for the preparation of the fuel sticks for the two remaining RTE's have been requested and delivery is anticipated on August 1, 1970. All other required materials are on hand. The last two RTE's are scheduled for insertion in the Peach Bottom Reactor in November 1970.

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