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THORIUM FUEL CYCLE DEVELOPMENT PROGRESS REPORT

MARCH 1970

NO. 8

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CHEMICAL TECHNOLOGY DIVISION
METALS AND CERAMICS DIVISION

THORIUM FUEL CYCLE DEVELOPMENT PROGRESS REPORT

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JUNE 1970

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THORIUM FUEL CYCLE DEVELOPMENT PROGRESS REPORT

MARCH 1970

NO. 8

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 either unbonded or bonded states.

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 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)

Additional work with irradiated Dragon fuel compacts is being carried out to further elucidate effects of fuel crushing operations on particle breakage. The Dragon compact reported on below contains 0.25 g of TRISO-coated $(U,Th)C_2$ microbeads per gram of compact. The remainder of the compact is graphite. The compact was irradiated for ~15,000 MWD/T in the Dragon Reactor, and has now cooled about 3 years.

Irradiated Dragon compact #8375 was sawed into two roughly equal cylindrical pieces. The half compact left over from sawing studies (58.32 g) was crushed in the wide open jaw position of our crusher (~5/8 in.). The resultant 9 pieces were combined into four samples of 11-16 grams each, labelled R-5 through R-8. The recovered fines weighed 0.85 gram (1.46%). The crusher handling loss was 0.12 g (0.21%).

If the entire loss weight and fines (0.97 g) were composed of broken beads and graphite in the ratio of weights of the original fuel compact, the crusher breakage of beads at wide jaw spacing would be less than 1.7%. Since not all loosened beads are crushed, actual breakage will be determined after the fines are leached.

1.1 Burn-Grind-Burn-Leach Head-End Studies

Sample R-5 (16.35 g, 2 pieces) was burned in moist oxygen at 750°C in a static bed. Burner product (4.12 g) appeared to be free flowing SiC-coated $(Th\text{-}U)C_2$ beads free of bulk carbon. The beads were ground in a Waring Blender purged with 0.2% H_2 in He. Blender product (4.04 g, 98%) was collected. The ground beads were burned to obtain an oxide weighing 3.58 g. This was leached 3 times with Thorex Reagent (13 M HNO_3 - 0.1 M $Al(NO_3)_3$ - 0.05 M HF). Two water rinses were combined with each leach solution. The final residue, which weighed 0.76 g, is being analyzed for U, Th, and selected fission products. The calculated weight of leacher products and residue summed to 97% of the leacher feed weight.

Recoveries are given in Table 1.

Sample R-6 (14.55 g) was burned in moist oxygen at 750°C in a static bed. Burner product (3.7 g) was again free flowing SiC beads with no apparent bulk carbon. The product from grinding in a Waring Blender as before was 3.63 g (98%). The oxide obtained from burning the ground beads weighed 3.05 g. The sum of the weights of leacher products and residue was 90% of the leacher feed weight. The material balances on this run are poor and indicate that R-6 data are probably in error. These runs will be repeated.

1.2 Release of Volatile Fission Products During Head-End Steps

Noble Gas (^{85}Kr) Releases

As mentioned in 1.1 above, fractions R-5 and R-6 were burned, ground, reburned, and leached. Collected off-gas purge streams from each step were analyzed for ^{85}Kr. Several (4-6) small samples of each gas composite were counted. A 95% confidence level "Student's t" statistical range based

Table 1. Compositions of Samples R-5 and R-6 from Dragon Compact 8375 Corrected by Material Balance

	R-5	R-6
U, g	0.227	0.167
Th, g	2.28	1.86
IPYC,* g	0.56	0.66
SiC, g	0.80	0.80
SiC coated particles, g	4.12	3.70
OPYC* and matrix graphite, g	12.23	10.85
Sample weight, g	16.35	14.55

*IPYC refers to the inner pyrocarbon coating;
OPYC refers to the outer pyrocarbon coating.

on the 4-6 samples, was calculated for each composite. Ranges were between 2.2 to 4.4% of the average value. These are good checks. The results are given in Table 2. As observed in the past, between 97-99% of the krypton was released when the SiC particles were ground and burned. In R-5 the major ^{85}Kr fraction was released during burning. In R-6 the major fraction was released during grinding. We do not understand the reason for this difference.

Tritium Releases

The tritium released in these studies was mixed with H_2 (diluted with He), or with moist oxygen purge gas, and collected on molecular sieves after it had been passed over a hot CuO converter to produce HTO. Tritium release in sample R-5 seems more consistent with previous work than sample R-6 (Table 2). About 32% of the tritium apparently was outside the SiC fuel particles. (Roughly 40% was found before.) The major tritium release occurred on burning the ground beads.

Table 2. Comparisons of Samples R-5 and R-6 Off-Gas Releases in Head-End Steps

Sample Analyzed	Fraction of ^{85}Kr Found		Fraction of ^{3}H Found	Total Activity	
	R-5 (%)	R-6 (%)	R-5 (%)	R-5 Totals (d/m)	R-6 Totals (d/m)
Off-gas from the first burning step	>0.4[a]	1.04	32.3	2.40×10^{8}	5.03×10^{8}
Off-gas from the grinding step	27.6	81.1	0.3	1.9×10^{6}	4.0×10^{6}
Off-gas from the second burning step	71.7	15.7	60.6	4.52×10^{8}	1.32×10^{7}
Off-gas from the leaching step	0.3	2.1	3.8	2.84×10^{7}	2.18×10^{7}
First leach liquor	--	--	3.0	2.25×10^{7}	2.7×10^{6}
Totals, d/m	4.13×10^{10}	3.34×10^{10}	--	7.45×10^{8}	5.44×10^{8}
Sample wt, g	16.35	14.55	16.35	16.35	14.55

[a]Collected on dry ice-charcoal trap; CO_2 displaced some of the ^{85}Kr.

[b]Second burning recovery of ^{3}H abnormally low.

Total amounts of tritium recovered from each step indicate that the sample R-6 values for the second burning and the first leach liquot are anomalous compared to sample R-5 results. The values for the other steps are in better agreement. These experiments will be repeated.

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 sticks, followed by comminution to size fractions suitable for the burn-leach process. However, the recent GGA decision to use loose fuel particles rather than fuel sticks as the reference form has necessitated changes in the approach.

There is nothing to report in this area this month.

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 Microspheres 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 Gel Spheres Without Fluidization

Some of the problems associated with microsphere forming are directly related to particle-particle and particle-column wall interactions in the fluidized-bed sphere-forming columns. These problems can be reduced or eliminated by gelling sol droplets in a free-fall column which is not fluidized and non-turbulent.

The column heights required for gelation of ThO_2 sol to form microspheres were studied as a function of temperature, 2-ethyl-1-hexanol—isoamyl alcohol (2EH-iAA) mixture composition and other variables as an MIT Practice School problem. The mass transfer correlations developed by Clinton for 2EH were extrapolated to the alcohol mixtures and to higher temperatures.

The maximum gel particle diameters were determined experimentally for a 2 $\underline{M}$ ThO_2 sol using 5- and 11-ft column heights and 2EH-iAA mixtures of 0 to 70 mole percent iAA. Agreement between the calculated and experimental diameters was acceptable for both column lengths and 0 to 30 mole percent iAA. Above 30 mole percent iAA, the experimentally determined maximum diameters were larger than the calculated values and the particles were distorted in shape. Experimentally determined viscosities and water solubilities were used for the calculations for 2EH-iAA mixtures as the two properties are not linear with mole fraction.

From the MIT Practice School study a computer program was developed and used to calculate column heights required to achieve gelation for a range of initial sol molarities and for two sol densities at gelation. For a given initial sol droplet size, the column height required for gelation

varies little with the initial sol molarity. As the initial sol molarity (and the final sphere size) increases at a fixed initial droplet size, the droplet gels in a shorter time, but settles faster, and two effects are mutually compensating. The column height required is a maximum at an initial ThO_2 or UO_2 sol molarity of about 3. The molarity required for gelation only has a small effect on the column length. Starting with a 1 M ThO_2 sol, a 12 M final concentration requires 93% of the column length required for 16 M. Some examples of the results are:

Initial sol droplet diameter, microns	400	400	400	400	400
Initial sol concentration, M	1	2	4	8	1
Final gel concentration, M	16	16	16	16	12
Time required, seconds	471	341	219	105	449
Column height, cm	283	339	359	287	263

Microspheres Preparation Development (C. C. Haws)

The engineering development system for preparing gel spheres in a fluidized bed is being modified to improve it for column chemistry studies. A single tank with mixing and a single pump is being installed. This simplified flow arrangement makes the calculations of and interpretation of concentration changes vs time much simpler.

1.2 Preparation of Test Materials

(J. R. Parrott, F. G. Kitts, P. A. Haas, F. L. Daley, R. J. Shannon)

In order to fabricate the eight recycle test elements (RTE's) that will be irradiated in the Peach Bottom Reactor we need several kilograms of ThO_2-UO_2 microspheres containing ^{235}U and thorium in the Th/U ratio of 2/1. Specifications call for the microspheres to be 350±100 μ in diam with a BISO coating of pyrolytic carbon.

The preparation of this material continued through the month. Six one-kg batches of sol were produced in the extraction equipment using the standard flowsheet for extracting nitrate from the previously mixed nitrate solution with Amberlite LA-2 amine in an n-paraffin diluent.

The first four batches of sol were formed into microspheres by an indirect method (addition of 4 moles of carbon per mole of oxide sol) described in last month's report. These runs yielded 2.7 kg of microspheres, bringing our total produced to date to 4.7 kg as UO_2-ThO_2. This material will be calcined to 1400°C in another facility to attain the desired density. One 600-g batch was screened and rounded, giving a 60% yield of material suitable for coating.

A photograph of this material was submitted to Gulf General Atomic (GGA) for comments regarding its use in irradiation tests. Although the microspheres have a rough surface and are not perfectly round, GGA felt they would be adequate.

Problems with handling the carbon sols, primarily frequent plugging of the disperser, led to a more concerted effort to determine the effect of operating variables on microspheres forming and hopefully a method to produce microspheres directly from the sol, without carbon addition.

A series of nine experimental sphere-forming runs, varying in scale from 10 g to 100 g of heavy metal were made in the development laboratory. These runs utilized feed prepared by the solvent extraction process. The results of these runs showed that by using 2EH with an Ethomeen S/15 content of 0.2 v/o, a water content of 1.5 v/o, and a pH between 3.5 and 4.5 (maintained with HNO_3), a satisfactory yield (71%) of microspheres could be obtained. Also, the microspheres must be steam dried, as previously observed, to prevent the temperature excursion which sometimes leads to destruction of the microspheres in the dryer. Modifications to the production sphere-forming facility were made to incorporate the necessary controls to meet the above requirements and the facility was placed in operation using carbon-free sol at the end of this period.

A high reject rate (60%), as discussed in detail last month, of the first batches of 100 μ $^{235}UO_2$ microspheres for the eight RTEs required the preparation during this report period of two additional batches.

1.3 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. Engineering studies of acid removal by caustic scrubbing reported previously have been discontinued because of intolerable and unavoidable caustic entrainment.

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

Last month, studies were continued on the purification of 2EH from the used 2EH and surfactant solutions that had been used in microsphere-forming columns. We determined the maximum, initial rates of distillation of 2EH free of surfactants that could be attained with our laboratory equipment at several temperatures, and the volume of water required per volume of the 2EH that was purified. The results are shown for the two-phase distillation at 99-100°C in Table 3 and for single-phase distillations at 150 and 180°C in Table 4.

The 100°C, 150 and 155°C distillations were carried out by metering the used 2EH and water into a distilling pot, collecting the condensate and then separating the 2EH and water phases in the condensate. A 180° distillation was carried out in the same manner except the 2EH was given a second distillation at 180°C. All of the 2EH recovered was free of surfactants (< 0.005 v/o). The rate of distillation was varied by varying the applied voltage to the electrical heaters via a variac. The rates attained at 100°C cannot be directly compared to those obtained at 150° and 180°. A 5-liter round-bottomed (R.B.) flask heated by a 600-watt heater was used in the 100°C distillation whereas a 1-liter R.B. flask heated by a 380-watt heater was used at the other temperatures.

Simple prorating of the volume-power ratio of the 5-liter flask to the 1-liter flask indicates that rates of 2EH recovery in the 100° distillation would be about a factor of 0.55 smaller had the experiment been performed in the same equipment as the 150 and 180° distillations.

Table 3. Two-Phase Distillation of Used 2EH and H_2O at 100°C

Conditions: Feed was 2EH containing surfactants that had been used in sphere-forming columns. An electrically heated (600 watt, max), 5-liter stillpot was used.

Rate of Collection of Distillate (ml/min)	2EH in Distillate (vol %)	Rate of 2EH Recovery (ml/min)
3.55	28	0.99
3.55	20	0.71
3.60	20	0.72
3.40	24	0.82
4.80	21	1.00
8.30	19	1.58
12.6	16	2.02

Table 4. Single-Phase Distillation of Used 2EH-Water Solutions

Conditions: Feed was 2EH containing surfactants that had been used in microsphere-forming columns. Electrically heated (380 watt, max), 1-liter stillpot used.

Temperature (°C)	Rate of Distillate Collection (ml/min)	2EH in Diphasic Distillate (vol %)	Rate of 2EH Recovery (ml/min)
150[a]	8.0	68	5.4
	8.0	73	5.8
	11.0	79	8.7
	12.0	77	9.2
	12.5	76	9.5
155[a]	15	78	11.7
180[b]	15	~ 100	15.0
	27.5	~ 100	27.5
	45.0	~ 100	45.0

[a]Water and used 2EH were metered into the distillation pot at volume ratios of 2EH/H_2O of ~4.

[b]Used 2EH saturated with water at room temperature (2.5% water) was metered into the distillation pot.

2.0 Fueled Graphite Fabrication Development

(F. J. Furman, C. B. Pollock, J. M. Robbins, W. H. Pechin, R. B. Fitts, 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, particle blending and bonding, fuel element assembly, and fuel element inspection. This month we continued those activities associated with the fabrication of the Recycle Test Elements (RTE's) which are to be irradiated in the Peach Bottom Reactor.

2.1 Particle Handling and Inspection

The microspheres prepared for the RTE's must be screened, shape separated (nonspherical particles eliminated), sampled, and batched prior to coating. The types of particles which will be produced at ORNL are BISO coated (low-density plus high-density pyrolytic carbon) ThO_2, $(0.8\ Th\text{–}0.2\ U)O_2$, $(0.67\ Th\text{–}0.33\ U)O_2$, and UO_2. The ThO_2 particles are being prepared by Ceramics Laboratory personnel as they are received from the Chemical Technology Division. Both the $(0.8\ Th\text{–}0.2\ U)O_2$ and the $(0.67\ Th\text{–}0.33\ U)O_2$ particles were contaminated with 10 parts per billion ^{238}Pu during their fabrication. Hence, special handling of these microspheres is necessary. A glove box was set up for the particle handling prior to coating and the $(0.8\ Th\text{–}0.2\ U)O_2$ particles have been processed.

The $(0.67\ Th\text{–}0.33\ U)O_2$ particles require high temperature (1400°C) sintering to reach densities greater than 95% of theoretical. Chemical Technology did not have equipment rated for this temperature so we are installing a furnace adjacent to the processing glove box. This

furnace will be in a hood which will have a filtered exhaust to prevent the spread of contamination in case the furnace tube should crack during operations. As soon as the installation is complete we will begin processing the $(0.67\ Th\text{-}0.33\ U)O_2$.

After the microspheres are coated they must be processed before the second coat is applied and before they are shipped. Although we hope the contamination will be contained by the coatings, we have installed a glove box for this processing. The particles will be transferred pneumatically from the furnace cooling hopper directly to the glove box. This transfer device, mentioned in the January monthly report, has continued to work well, transferring both low- and high-density pyrolytic carbon coated microspheres without damage.

2.2 Particle Coating

During test runs in preparation for coating thorium-bearing RTE microspheres, the electrodes in the 5-in.-diam prototype coater shorted to ground, requiring extensive repairs. The short was caused by a Micarta insulator heating to a higher temperature than expected. This problem has been corrected and the furnace repaired. Since some changes in the heat distribution in the furnace occurred when the problem was corrected, the coating parameters are being rechecked prior to coating the RTE microspheres.

The changes to the furnace appear to have had little effect on the results of low-density pyrolytic carbon coating with acetylene and helium, but the results of high-density pyrolytic carbon coating with propylene appear to have shifted considerably. We are presently adjusting our parameters to achieve the high-density coatings specified for the RTE's.

During the past month we have continued to coat small UO_2 particles for the Peach Bottom recycle test elements. At the present time we have coated in excess of 400 g of UO_2 kernels. These represent approximately 1.8 kg of coated particles. One additional batch of UO_2 kernels is required to complete the job.

Several interesting points have been observed in the UO_2 coating operation. We have found that it is possible to deposit rather thick buffer coatings on small particles by utilizing larger "knocker" particles and lower gas flows. Also, we have been able to vary the density of the buffer coatings by varying the gas mixtures. For example, we found with a ratio of 7000 cm^3/g of C_2H_2 and 3000 cm^3/g of He, the coating density was approximately 1.3 g/cm^3. If the ratio was changed to 7500 cm^3/g of C_2H_2 and 3000 cm^3/g of He, then the density of the coating was 1.15 g/cm^3. The very small particle size in this experiment has created a problem in that faceting of the particles is common. But we have learned how to minimize this problem and are producing spherical particles. It was found that a larger gas flow could be used if the amount of "knocker" was increased. The larger gas flow of course decreased the number of faceted particles.

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

The results of HFIR Target Experiment HT-4 show that we have developed a bonded bed which potentially meets the requirements of HTGR fuel sticks. However, additional work is required to find the most economical raw materials and refabrication technique.

A series of new bonded-bed specimens is being prepared for irradiation testing in the spine positions of the Recycle Test Elements. All samples will contain identical coated particles, and each will be 0.4 in. in diameter by 0.7 in. long and supported in a small H-327 graphite crucible. The compositions selected for testing are listed in Table 5. Also, these new filler materials will subsequently be tested in a HFIR target experiment similar to HT-4.

The Santa Maria flour was obtained through the Los Alamos Scientific Laboratory (LASL) and the Y-12 Chemical Engineering Development Department. It is reported to result in bodies similar in unirradiated properties to the Poco grades of graphite.[1] When mixed

[1]LASL Progress Report 9, LA 4171-MS, Feb.—April, 1969.

Table 5. Bonding Materials Selected for Irradiation Testing in the Peach Bottom Recycle Test Element Experiment

Binder[a]	Filler	Amount Filler[b] (wt %)	Number Specimens
Varcum[c]	Poco AXM[d]	40	2
Varcum	Robinson[e]	45	2
Varcum	JOZ[f]	42	2
Varcum	Santa Maria[g]	50	2
Varcum	Asbury[h]	35	2

[a]Compositions will be duplicated using Allied Chemicals 15V coal tar pitch as binder in place of Varcum.

[b]Represents the practical maximum of each filler material.

[c]A prepolymerized furfuryl alcohol from Varcum Chemical Company.

[d]−40 μ size graphite flour from Poco Carbon Company.

[e]−40 μ size graphite flour from Carbon Products Division, UCC.

[f]−40 μ size graphite flour from Great Lakes Carbon Corporation.

[g]−40 μ size graphite flour from Collier Carbon and Chemical Corp.

[h]−40 μ size graphite flour furnished by Asbury Graphite Company.

with Varcum in concentrations up to 50 wt %, the Santa Maria flour made a smooth appearing matrix material that injected easily through the 0.7-in.-long bed of the test piece. The JOZ grade of graphite flour is derived from a petroleum coke and is the base material for Great Lakes Grade H-337 graphite. Our interest in this material was evoked by the encouraging irradiation behavior of H-337 in HFIR experiments at 750°C, and the availability of the material in large quantities. Only 42 wt % of the JOZ flour could be mixed with Varcum and still be usable. The mix showed a slight tendency toward dilatancy;[2] however, injection was not difficult through the 0.7-in.-long test specimen. The Robinson grade flour is an air-blown graphite powder which does not form a needle-like structure and therefore may

[2]A dilatant mix is one that seems reasonably fluid when at rest but becomes more viscous when pressure is applied or when otherwise agitated.

form an isotropic matrix. We were able to inject a mixture of 45 wt % of the Robinson flour in Varcum through the 0.7-in.-long test piece. When 50 wt % of the flour was mixed with Varcum, the mix showed a marked degree of dilatancy[2] and could not be injected more than approximately one-half inch into the test bed.

The Asbury flour is the natural flake graphite that was used in HT-1 and HT-3. Only 35 wt % could be added to Varcum and injected through the test piece. One mix was evaluated where only Thermax could be added to Varcum and still be usable. This is the same amount of total filler material that can be used when a mixture of equal amounts of Thermax and Poco graphite flour is the filler. Test pieces, 0.7 in. long have also been fabricated using 15V coal tar pitch as the binder with each of the above described filler materials in the same concentrations as given for the Varcum binder.

It should be emphasized that these test pieces are only 0.7 in. long, as described above. More work is needed to determine the practical maximum length that can be fabricated using these bonding materials.

3.0 Coated Particle Fuels Quality Assurance Program for RTE's

W. H. Pechin, F. J. Furman, R. B. Fitts

We are writing the quality assurance procedures to be used in the preparation of fuel for the Recycle Test Element to be irradiated in the Peach Bottom Reactor.

Last month we listed the analyses to be performed at the various stages of the coating process. Experience since that time indicates that at least one change is advisable. After the application of the isotropic coating it is difficult to separate the oversize marker particles from the main coating batches without including some buffer coated material. The buffer coated material in the samples results in high values for carbon and low values in the mercury density. As an alternative we will crack the isotropic coatings off of the particles to determine the density in a gradient density column. In the

interests of economy we have developed a sink-float test in which coating samples are dropped into a series of solutions of various densities. This test will tell us if the coating density is within the tolerance limits after each coating run and we can submit samples in groups of the most economical size for final density measurement.

III MATERIAL IRRADIATION

(A. L. Lotts, T. N. Washburn, J. D. Sease, J. H. Coobs)

1. HTGR Recycle Fuels Irradiation

(T. N. Washburn, R. B. Fitts, 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 are being implemented this year. They are (1) accelerated burnup rate capsule irradiations, and (2) eight test fuel elements to be irradiated in the Peach Bottom Reactor.

1.1 Capsule Irradiation

The review of the available information on the ORR "partial element" tests was completed. The data indicate that we would have to remove at least two additional fuel plates to have adequate room for cooling the capsules and that the resulting fast flux-to-thermal flux ratio would be only slightly better than that advertised for ETR J-8 positions where we have adequate coolant capacity. Therefore, we have requested up-to-date flux monitoring of the J-8 positions

and have begun the design of capsules for these irradiation facilities. The current conceptual design involves two temperature regimes in each capsule with five 0.5-in.-diam x 2-in.-long fuel bed increments in each regime. Preliminary heat generation calculations indicate linear heat rates as high as 18 kw/ft will result from a bed loaded with equal numbers of 4.2:1 $(Th,U)O_2$ and ThO_2 coated particles. Detailed physics and heat transfer calculations are being made. These together with the program needs for recycle separation tests and particle evaluation tests will be used to finalize the design.

1.2 Large Scale Irradiations

Eight Recycle Test Elements (RTE's) will be operated in the Peach Bottom Reactor starting in June 1970. These fuel elements will contain samples of fuels proposed for use in 1100 Mw(e) HTGR's. The preparation of the ORNL portion of the fuels for RTE's is behind schedule, although not seriously, and is progressing as outlined below and described in detail as indicated.

The ORNL specifications for kernels, coatings, and fuel sticks are being put in final form. The data sheets required to document the quality of the fuel are undergoing review. Progress on defining the analytical techniques to be employed are reported under Section 2 of this report.

The coating of the ThO_2, 2:1 $(Th,U)O_2$, and 4:1 $(Th,U)O_2$ kernels (three of the four types to be coated) is described in Section 2 of this report.

IV. URANIUM-233 REPROCESSING

(J. R. Parrott, R. G. Nicol, W. A. Shannon)

ORNL serves as a national distribution center for ^{233}U. The facility, which contains a small batch leacher, a batch dissolver, and a single-cycle solvent extraction system, is capable of purifying ^{233}U at the rate of 25 kg per week. It includes storage system for liquids and solids, with capacities of 500 and 120 kg of ^{233}U respectively.

1. Dissolution and Purification

The dissolution and purification equipment were not operated during the month. Except for 9 kg of uranium (84% ^{233}U) remaining in the vault, all the scrap has been dissolved and accounted for.

Process piping was revised to permit the transfer of water in storage-tank U-3 to the evaporator feed tank for concentration and storage. This water was used to re-calibrate the tank to determine the density of glass in the tank which would indicate breakage and settling of the rings and had stripped 500 g of ^{233}U off the rings while standing over a several-months period.

2. Storage and Distribution

The facility presently contains 226 kg of ^{233}U which varies in isotopic purity between 84 and 98% and ^{232}U content between 4 and 250 ppm. A total of 68 g of ^{233}U was received and 103 g of ^{233}U was shipped during the month.

Present plant criticality requirements specify that all ^{233}U or ^{235}U in solution being transferred to the tank farm must be mixed with or followed by 100 times as much ^{238}U as uranyl nitrate solution. To meet this a 150-gal tank was installed in the penthouse and was connected to a "hot" drain line going directly to the tank farm. The storage tank was calibrated, charged with 100 kg of ^{238}U, and placed in operation.

We also have a facility (TRUST-Thorium Reactor Uranium Storage Tank) in which we store 1047 kg of highly enriched uranium (76.5% ^{235}U, 9.7% ^{233}U) in the form of a uranyl nitrate solution. This material is the uranium product from the Indian Point reactor fuel, which was purified by solvent extraction at the Nuclear Fuels Services Plant. The solution will

be stored indefinitely since the ^{232}U content (120 ppm) prohibits its direct refabrication into fuel elements, and the low ^{233}U content makes it of little interest for reactors demonstrating the thorium fuel cycle.

The pressure control valve, purchased for use in the new vessel off-gas line for storage tank P-25, was installed, but instrumentation was not completed. When instrumentation is complete, sparging tests will be performed to determine the effectiveness of the recently added de-entrainment separator and filter.

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