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HOT-CELL EVALUATION OF THE GRIND-LEACH PROCESS. 1. IRRADIATED HTGR CANDIDATE FUELS: PYROCARBON-COATED $(Th,U)C_2$ PARTICLES DISPERSED IN GRAPHITE

J. R. Flanary
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Price: Printed Copy $3.00; Microfiche $0.65

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ORNL-4117

Contract No. W-7405-eng-26

CHEMICAL TECHNOLOGY DIVISION
Chemical Development Section B

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AUGUST 1967

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ABSTRACT

The grind-leach process, an aqueous head-end method developed for HTGR candidate fuels, was evaluated in hot-cell experiments with irradiated fuel specimens composed of pyrocarbon-coated $(Th,U)C_2$ particles dispersed in graphite. Fine-grinding of the GAIL-3A and GAIL-3B fuel compacts (supplied from General Atomic's In-Pile Loop irradiation studies) and ORNL experimental AVR fuel spheres to -100 mesh, followed by multiple leaching with boiling 13 M HNO_3, failed to produce near-quantitative recovery of the fuel values; as much as 4.5% of the uranium and 6.9% of the thorium appeared as nonleachable losses in the graphite waste. (The grind-leach method normally provided 99.7% recovery of heavy metals from similar unirradiated fuel samples.)

Retention of uranium, thorium, and fission products in the residue was uniformly high for all leached fuel specimens that had been irradiated to burnups of 9600 to 41,200 Mwd per metric ton of (U + Th). Although the nonleachable fractions of heavy metals and fission products appeared to be independent of the known irradiation histories of the test fuels, the results implied that the fraction resistant to leaching may have been bound in the pyrocarbon coating, perhaps by the mechanisms of diffusion and fission recoil.

Because of incomplete recovery of the fuel values, and the large fraction of heat-generating fission products that remains unleached from the graphite waste, the grind-leach method does not appear promising for processing fuels that contain pyrocarbon-coated carbide particles. However, it may prove useful in processing other coated-particle fuels that are currently being developed.

1. INTRODUCTION

The principal objective of the hot-cell studies was to evaluate the grind-leach process as a means for both quantitatively recovering uranium and thorium and removing fission products from graphite-base

reactor fuels; in addition, the effects of various methods of fuel fabrication and irradiation histories on the process performance were investigated.

Graphite-base fuels currently favored for high-temperature, gas-cooled reactors are composed of pyrolytic-carbon-coated $(Th,U)C_2$ particles dispersed throughout a graphite matrix. This type of fuel is being developed by General Atomic (a Division of the General Dynamics Corporation) for use in the Peach Bottom and Public Service Company of Colorado reactors. Spherical graphite fuel elements that contain pyrolytic-carbon-coated $(Th,U)C_2$ microspheres are being used in the AVR at Juelich, West Germany. Fuel for the O.E.C.D. Dragon Reactor is composed of pyrocarbon-coated $(Th,U)C_2$ microspheres (having an intermediate layer of silicon carbide) dispersed in a graphite matrix.

In the grind-leach process that has been developed at Oak Ridge National Laboratory,[1-5] the fuel elements are coarsely crushed to -4 mesh and then ground to -140 mesh in a double-roll mill to ensure that the chemically inert pyrocarbon coating of every fuel particle is broken. The exposed carbide or oxide kernel is leached from the graphite with boiling nitric acid or fluoride-catalyzed nitric acid, producing a solution from which the uranium and thorium can be recovered by solvent extraction.

The experiments described in this report were performed with fuel specimens [pyrocarbon-coated $(Th,U)C_2$ particles in a graphite matrix] that were prepared by General Atomic and irradiated by General Atomic and ORNL. Similar experiments are planned for investigations of other fuel specimens [silicon carbide-coated $(Th,U)C_2$ particles] to be supplied by the Dragon Project.

The authors wish to thank J. W. Ullmann for his assistance in obtaining generous quantities of irradiated fuel specimens from General Atomic. M. F. Osborne, D. R. Cuneo, and H. E. Robertson, ORNL Reactor Chemistry Division, furnished the AVR spheres. L. A. Byrd, O. L. Kirkland, and G. E. Woodall performed the hot-cell manipulations. Analytical services were provided by the groups of C. E. Lamb, W. R. Laing, and E. I. Wyatt of the ORNL Analytical Chemistry Division.

2. DESCRIPTION OF MATERIALS

2.1 Equipment

For hot-cell tests of the grind-leach process, we needed a simple, compact device that would effectively size-reduce 10- to 20-g batches of coarsely crushed HTGR fuel [265- to 465-μ-diam pyrocarbon-coated $(Th,U)C_2$ particles dispersed in graphite] to -140 mesh (particles 105 μ and smaller). The Unit Operations Section had developed a double-roll mill with such a capability; however, since it was considered unwieldy for small-scale hot-cell work, and since designing and building special equipment for this type of service would have been costly and time-consuming, we investigated the possibility of using a commercial food blender.

Tests with the latter device (model 1037, Waring Products Co.) showed that 10-g batches of coarsely crushed, unirradiated HTGR fuel could be completely pulverized to -140 mesh, 95% in the first 20-min pass and the remainder in two additional 10-min passes. The blender was modified for remote operation by sealing the lid with a neoprene gasket and fitting the shredder with sealed roller bearings to replace the original bronze sleeve bearing that had overheated while grinding the dry, abrasive powder. Each batch of pulverized fuel was passed through a sieve to ensure that all the fuel particle coatings were shattered. A small electrically driven, vibrating plate was built to aid the in-cell sieving operations.

In the leaching experiments, each of the samples of powdered fuel (10 to 20 g) was placed in a standard 125-ml Erlenmeyer flask that was mounted on a hot plate and connected to an updraft condenser. Leach liquors were withdrawn from the flask by applying vacuum to a Pyrex tube fitted with an M-porosity filter, and the leached graphite was transferred onto a Büchner funnel, where it was washed several times. Each wash was removed by vacuum filtration. The last traces of water were removed from the residue by washing with acetone; after air-drying, the powder was transferred to a sample bottle, and then submitted for analysis.

For the final series of runs designed to determine the reproducibility of the results, the equipment was modified to minimize the exposure of the leached graphite to extraneous contamination in the hot cell. Special Erlenmeyer flasks were fabricated with built-in M-porosity frits. The leach and wash liquors were removed by vacuum filtration through a sidearm, and all the finely divided graphite was retained in the flask. The leached graphite was dried in the flask with acetone and then transferred directly into a sample bottle.

2.2 Irradiated Fuel

Irradiated fuel compacts were obtained from the General Atomic GAIL-3A and GAIL-3B In-Pile Loop experiments. These compacts consisted of 2.75-in.-OD, 1.75-in.-ID, by 1.5-in.-thick annular rings of hot-pressed graphite interspersed with 30 vol % $(Th,U)C_2$ coated particles. During fabrication, a size range of 150 to 350 μ was carefully maintained for the $(Th,U)C_2$ kernels, upon which single, laminar 55-μ coatings of pyrolytic carbon were applied by cracking C_2H_2 at 1400°C. The overall fuel composition of the GAIL-3A compacts before irradiation was 6.8% uranium (93% enriched), 15.5% thorium, and 77.7% carbon; the GAIL-3B compacts contained slightly more thorium.[6]

The GAIL-3A compacts were irradiated to a reported average burnup of 8850 Mwd per metric ton of (U + Th), whereas the GAIL-3B compacts received a more severe treatment, attaining an average burnup of 41,500 Mwd per metric ton, with fuel core temperatures reaching 1750°C. Detailed irradiation data are given in Table 1. Postirradiation examination[7] of the GAIL-3A fuel by General Atomic indicated that some of the fuel particle coatings had ruptured during irradiation. Subsequent examination[8] of the GAIL-3B material revealed that about 98% of the pyrolytic carbon coatings had failed in compacts Nos. 6 and 7, which had reached the maximum fuel temperature (1750°C) and burnup (5.5%). GAIL-3B compact No. 7 was chosen for the initial hot-cell grind-leach experiments because it had attained the highest burnup.

Experimental AVR fuel in the form of 1.5-in.-diam spheres was also used to evaluate the grind-leach process. These specimens, which were

Table 1. Fabrication and Irradiation Data for GAIL-3A and GAIL-3B Compacts and AVR Fuel Spheres

	Fuel		
	GAIL-3A[a]	GAIL-3B[a]	AVR Spheres[b]
Flux (thermal)	~ 4×10^{13}	~ 4×10^{13}	2.1×10^{13}
Irradiation time, sec	10^7	3.75×10^7	9.06×10^6
Discharge date	2/4/62	3/18/64	8/25/63
Mw per compact	0.067	0.067	0.017
Average burnup, Mwd per metric ton of (Th + U)	8850	41,500	23,200
Burnup (Th + U), atom %	0.9	2.8 to 5.5	---
Range of in-pile temperatures, °C	1370 to 1480	1300 to 1420	540 to 790
Approximate composition of compact			
Uranium (93% enriched), g	13 ± 0.15	13	1.96
Thorium, g	29.5 ± 0.3	32.7	1.20
Carbon, g	Balance	Balance	Balance
Total weight, g	190	190	54.6
Thorium:uranium ratio	2.27	2.5	0.614
Moles of ^{235}U per mole of thorium	0.436	0.395	1.497
g of ^{235}U per g of compact	0.0646	0.0655	0.0333
Particles	50- to 60-μ-thick coatings 150- to 420-μ-diam substrate	55-μ-thick coatings 150- to 350-μ-diam substrate	87- to 125-μ-thick coatings 177- to 250-μ-diam substrate

[a]Irradiated in the GETR, Vallecitos, California; data supplied by General Atomic.

[b]Irradiated in the ORR, Oak Ridge National Laboratory.

prepared for ORNL by General Atomic, consisted of 177- to 250-μ-diam $(Th,U)C_2$ particles, with single, laminar 87- to 125-μ-thick pyrocarbon coatings, dispersed in hot-pressed graphite; they were irradiated by ORNL to a burnup of 23,000 Mwd per metric ton at temperatures reaching 790°C (Table 1). This material was selected for further process studies because (1) its composition and fabrication were comparable to the GAIL-3B compacts and (2) postirradiation examination[9] had shown that the fuel particle coatings remained intact throughout the irradiation.

3. LABORATORY STUDIES USING UNIRRADIATED PROTOTYPE HTGR FUELS

3.1 Peach Bottom Fuel

Leaching experiments were performed, using unirradiated Peach Bottom fuel (3% uranium, 14% thorium, 83% carbon) that was composed of 150- to 420-μ-diam $(Th,U)C_2$ microspheres, with 55-μ-thick pyrocarbon coatings, dispersed in a graphite matrix. Batches (about 40 g) of the coarsely crushed fuel (-6 mesh as obtained from the Unit Operations Section) were pulverized completely to -100 mesh in a Waring blender; each batch of ground fuel was passed through a 149-μ sieve.

Samples (about 10 g) of the pulverized fuel were leached for 5 hr with 25 ml of boiling 13 M HNO_3--0.05 M HF. The refluxing acid appeared to provide adequate solid-liquid contact. The cooled leach liquor was removed by filtration, and the graphite residue was leached for 2 hr with a second 25-ml aliquot of boiling reagent. After the second leach liquor was removed, the graphite residue was washed with three 25-ml aliquots of water at 25°C (Table A-1 in Appendix) or with five 25-ml aliquots of leachant at 25°C (Table A-2 in Appendix).

In run PB-2, 99.85% of the uranium and 99.99% of the thorium were recovered; analysis of the leach and wash solutions indicated that the small amount of uranium (0.42 mg) found in the 7.75-g residue was the result of incomplete leaching. In runs PB-3 and -4, 99.98% of the uranium and 99.99% of the thorium were leached. Since the five washes removed almost negligible amounts of uranium, the nearly quantitative

recovery of the heavy metals is attributed to effective leaching at the reflux temperature.

3.2 Experimental Graphite Fuel Spheres

Since the initial evaluation of the grind-leach process, using GAIL-3B fuel compacts (Sect. 4.1), indicated that recoveries of the heavy metals were incomplete and that the leaching of fission products was inadequate, we made additional tests using other HTGR-type fuel samples with different irradiation histories. We obtained quantities of both irradiated and unirradiated 1.5-in.-diam spheres of experimental coated-particle fuel that had been manufactured by General Atomic for ORNL, using the same techniques that were employed for the GAIL-3A and -3B fuels (Sect. 2.2). The unirradiated spheres were coarsely crushed and then ground to -100 mesh for leaching tests to serve as controls for the irradiated specimens that were to be tested in subsequent experiments.

Samples (about 20 g) were leached two to four times with 50-ml aliquots of boiling 13 M HNO_3--0.05 M HF; then the residue was washed three to five times with 25-ml aliquots of water at 25°C following each leach. The results (Table A-10 in Appendix) showed that a 3-hr leach dissolved 95 to about 99% of the heavy metals but that the remainder was leached very slowly and incompletely. About 99.7% was recovered in two leaches (total contact time, 7 hr); no improvement in recovery could be achieved with four leaches (total contact time, 9 hr).

3.3 Leaching Experiments with Unirradiated Peach Bottom Fuel to Determine Reproducibility of Results

In the first hot-cell trials of the grind-leach process, inconsistencies in the results, particularly regarding the leaching of fission products ^{106}Ru and ^{144}Ce, raised doubts as to the adequacy of analytical techniques and our experimental procedures. In an attempt to resolve these inconsistencies, we refined the original leaching procedure to minimize operational errors, particularly contamination of samples that were to be analyzed. Another objective was the development of a standard leaching technique for all types of HTGR fuels to be examined in the hot cells.

Four runs were made to determine whether results obtained by using this revised procedure were reproducible. About 128 g of coarsely crushed, unirradiated, prototype Peach Bottom fuel was ground in a blender and sieved to obtain a -100 mesh product; this product was then passed repeatedly through a powder sample splitter, which subdivided the batch into eight, near-homogeneous samples of nearly equal weight (~ 16 g).

Four of the samples were leached for successive periods of 3, 2, 2, and 2 hr using fresh boiling 13 M HNO_3 in each leach. Each residue was washed with water three times to remove the dissolved uranium and thorium.

Average total recoveries of 99.84% of the uranium and 99.71% of the thorium were obtained (Table A-3 in Appendix); however, even greater recoveries (99.98% of the uranium and 99.99% of the thorium) had been attained in previous tests using the original leaching procedure and the same type of fuel. This discrepancy cannot be adequately explained at the present time.

The results obtained with the revised procedure show a high degree of reproducibility (Table 2), demonstrating that (1) the procedure is devoid of large, random operational errors and (2) the precision of the analytical techniques used to determine uranium and thorium is satisfactory.

The results (Table A-3 in Appendix) show clearly that most of the $(Th,U)C_2$ was dissolved rapidly in the first 3-hr leach with boiling 13 M HNO_3. In the following three leaches (total contact time, 6 hr), only 0.26% of the uranium and 0.86% of the thorium were leached from the residue. The amount leached diminished with each successive leach as the heavy-metals "heel" became smaller. The overall dissolution rate of the "heel" was about three orders of magnitude lower than that of the bulk of the $(Th,U)C_2$, which suggests that the "heel" may have been a solid solution of the heavy-metal carbides and pyrolytic carbon, formed during coated-particle fabrication, that was difficult to dissolve.

The revised procedure was also used with GAIL-3B fuel (Sect. 4.4) that had been irradiated to an average burnup of 36,400 Mwd per metric ton, according to our measurements.

Table 2. Averaged Results Obtained by Leaching Prototype Peach Bottom Fuel According to the Revised Procedure[a]

Recovered in	Uranium (%)	Standard Deviation	Thorium (%)	Standard Deviation
First leach	99.58	± 0.026	98.85	± 0.044
Second leach	0.16	± 0.039	0.70	± 0.052
Third leach	0.06	± 0.015	0.10	± 0.016
Fourth leach	0.04	± 0.007	0.06	± 0.005
Average total recovered	99.84		99.71	
Residue	0.16	± 0.018	0.29	± 0.027

[a]Note that these results are for four identical experiments (Table A-3 in Appendix).

4. HOT-CELL STUDIES WITH IRRADIATED PROTOTYPE HTGR FUELS

4.1 GAIL-3B Fuel

The first hot-cell trials of the grind-leach process were made with compact No. 7 of the GAIL-3B fuel (Sect. 2.2). This compact was selected because it had the highest reported burnup: 55,000 Mwd per metric ton of (U + Th). It was first crushed to -4 mesh lumps in a small ore crusher (Denver Fire Clay Model 1). Then approximately 40-g batches were individually size-reduced to -100 and -140 mesh in a commercial food blender; each batch was sieved to meet the desired particle size specification. No difficulties were encountered in the fine-grinding procedure.

In eight experiments, 10-g samples of the pulverized fuel were leached two to three times with refluxing 13 M HNO_3 or 13 M HNO_3--0.05 M HF and washed five times with fresh leachant (or water) at 25°C; the graphite residue was dried with acetone. In spite of extensive leaching and washing, the residues retained excessive amounts of the thorium

(2.02% average) and the uranium (1.14% average) in some form that was inert to the leachant (Tables A-4 through A-7 in Appendix). Grind-leach processing of unirradiated fuel of this type normally results in recovery of 99.7% of the uranium and thorium (Sect. 3.2). Retention of fission product gamma and beta emitters by the residues averaged 8.8 and 20.6%, respectively, the major contaminants being ^{106}Ru and ^{144}Ce.

Analyses of the leach and wash solutions indicated that the excessive amounts of uranium, thorium, and fission products retained by the graphite were caused by incomplete leaching rather than by inefficient washing. However, no significant improvement was noted in the recovery of heavy metals or the leaching of fission products from the residues when any of the following treatments was incorporated in the original procedure: (1) making the 13 M HNO_3 leachant 0.05 M in HF, (2) using three leaches instead of two, (3) grinding the fuel to a -140 mesh product, and (4) washing of the leached residue several times with various agents (13 M HNO_3, 13 M HNO_3--0.05 M HF, water, C_2H_5OH).

Postirradiation examination[8] of the GAIL-3B fuel by General Atomic showed that up to 98% of the pyrocarbon-coated particles had ruptured in compact No. 7, which had attained a maximum burnup of 36,400 Mwd per metric ton (average of our burnup calculations based on cesium analysis) and a maximum core temperature of 1750°C. Consequently, we decided to perform additional hot-cell experiments with other coated-particle carbide fuels having different irradiation histories to determine whether the retention of thorium, uranium, and fission products by the graphite may be associated with the fact that the particles had been ruptured.

4.2 Experimental Graphite Fuel Spheres

We used experimental graphite fuel specimens (1.5-in.-diam spheres; Sect. 2.2) that had been fabricated for ORNL by General Atomic in essentially the same way as the GAIL-3B compacts but had been irradiated to 23,000 Mwd per metric ton at temperatures reaching 790°C without suffering rupture of particle coatings.[8] Samples from ORNL experiment No. 08A-6 were ground to -100 mesh; after three leaches (total contact time, 9 hr) with refluxing 13 M HNO_3--0.05 M HF followed by exhaustive water washing,

0.7 to 2.5% of the uranium and 1.2 to 4% of the thorium were found in the residue (Table A-11 in Appendix). [In control experiments with unirradiated samples of this fuel, 99.7% of the heavy metals was recovered (see Sect. 3.2).] From 19 to 35% of the fission product gamma emitters were also retained in the residues, the principal contaminants being ^{106}Ru and ^{144}Ce (Table A-11 in Appendix).

4.3 GAIL-3A Fuel

Two other experiments (3A-1 and -2) were made using the previously described leaching and washing procedure with GAIL-3A fuel that had been irradiated to a measured burnup of 9600 Mwd per metric ton at in-pile temperatures reaching 1480°C. Postirradiation examination[7] had revealed some cracked particle coatings. The leached residues from this particular fuel retained as much as 0.9% of the uranium, 4% of the thorium, and 19% of the gamma activity (Table A-9 in Appendix).

4.4 Leaching Experiments with GAIL-3B Fuel to Determine Reproducibility of Results

Three runs, which were made to determine the reproducibility of results obtained from leaching, were attempted using GAIL-3B material that had been irradiated to a measured burnup of 36,400 Mwd per metric ton. The revised procedure was employed, which, in previous tests with unirradiated prototype fuel, had given excellent reproducibility of results (Sect. 3.3). Approximately 80 g of GAIL-3B compact No. 9 was ground and sieved to -100 mesh, and the pulverized product was subdivided into four ~20-g samples using a powder sample splitter. Each sample was leached with aliquots of refluxing 13 M HNO_3 for successive periods of 3, 2, 2, and 2 hr. After each leach, the residue was washed three times with boiling water. All leach liquors and wash solutions were removed from the flask at about 80°C by vacuum filtration, and the residue was dried with acetone and sampled as a powder.

Even with the carefully controlled leaching procedure, recovery of the heavy metals was rather low; the residues retained 1.88 to 4.46% of the uranium and 4.45 to 6.89% of the thorium. (In these experiments, the

retention of heavy metals by the leached graphite was the highest encountered in our hot-cell studies.) Retention of principal fission products was comparable to that observed in previous experiments; as much as 42% of the ^{144}Ce, 8.3% of the ^{137}Cs, and 3.6% of the ^{106}Ru remained in the graphite.

The complete data (Table A-8 in Appendix) show variable results for the leaching of uranium and thorium. Somewhat better reproducibility was demonstrated in leaching ^{144}Ce and ^{137}Cs than for the uranium and thorium, probably because of the refined leaching procedure and the greater precision provided by gamma scanning. Although adequate techniques were developed for analyzing uranium and thorium in unirradiated graphite residues, these techniques are undoubtedly subject to error when applied to the "hot" analytical facility. Also, it is possible that the fraction of heavy-metal carbides "fixed" in the pyrocarbon coating by diffusion or fission recoil may vary significantly within a single irradiated fuel compact; in this case, grinding the fuel to only -100 mesh (149 μ) could produce segregation of the dense $(Th,U)C_2$ kernels from the relatively light graphite fines. Also, passage of the pulverized fuel through a sample splitter may be insufficient to produce completely homogeneous samples with regard to the distribution of the shattered pyrocarbon coatings.

5. DISCUSSION AND CONCLUSIONS

For convenience in reviewing the pertinent results of this study, all grind-leach experiments are summarized in Table 3. The data in this table indicate that two leaches (total contact time, 7 hr) will recover all of the readily dissolvable $(Th,U)C_2$; additional leaching is quite unproductive. The recovery of heavy metals is about the same whether the leached graphite is washed with water or with fresh leachant; however, washing with boiling water is preferred because vacuum filtration in this instance is about three times as fast as with concentrated nitric acid. The recovery of heavy metals is not increased by making the leachant 0.05 M in HF; however, leachants containing HF were used in most of the hot-cell runs on the

Table 3. Summary of Grind-Leach Experiments

Run No.	Burnup[a] (Mwd/metric ton)	Leaches		Washes		Amount Unleached from Graphite Residue (%)			
		Number	Reagent	Number	Reagent	U	Th	Gr γ	Gr β
			Peach Bottom Fuel - Unirradiated						
PB-1	0	2	HNO_3-HF	3	H_2O	0.44	2.74		
PB-2	0	2	HNO_3-HF	3	H_2O	0.15	0.01		
PB-3	0	2	HNO_3-HF	5	HNO_3-HF	0.02	0.002		
PB-4	0	2	HNO_3	5	HNO_3	0.02	0.001		
			Peach Bottom Fuel - Unirradiated - Reproducibility Runs						
PB-6	0	4	HNO_3	3	H_2O	0.16	0.26		
PB-7	0	4	HNO_3	3	H_2O	0.14	0.27		
PB-8	0	4	HNO_3	3	H_2O	0.16	0.29		
PB-9	0	4	HNO_3	3	H_2O	0.19	0.33		
			GAIL-3B Fuel						
3B-1	33,500	2	HNO_3	5	HNO_3	0.54	0.90	2.48	8.25
3B-2	41,200	2	HNO_3	5	HNO_3	2.15	3.29	8.99	18.82
3B-3	34,100	2	HNO_3-HF	5	HNO_3-HF	0.44	3.82	7.38	22.52
3B-4	33,400	2	HNO_3-HF	5	HNO_3-HF	1.40	0.92	8.21	26.27
3B-5	41,000	2	HNO_3-HF	5	HNO_3-HF	1.29	2.83	10.22	26.97
3B-6	38,500	2	HNO_3-HF	5	HNO_3-HF	1.37	2.91	14.43	-
3B-7	36,200	3	HNO_3-HF	5	H_2O	0.54	0.40	8.17	-
3B-8	33,300	3	HNO_3	2	C_2H_5OH	1.38	1.07	10.53	-
				3	H_2O				
			GAIL-3B Fuel - Reproducibility Runs						
3B-9	36,800	4	HNO_3	3	H_2O	2.54	4.45	18.22	
3B-10	30,600	4	HNO_3	3	H_2O	4.46	6.89	8.80	
3B-11	25,100	4	HNO_3	3	H_2O	1.88	6.27	0.93	
			Experimental Graphite Fuel Spheres - Unirradiated						
GA-1	0	2	HNO_3-HF	3	H_2O	0.90	1.04		
GA-2	0	2	HNO_3-HF	3	H_2O	0.33	0.27		
GA-3	0	4	HNO_3-HF	3	H_2O	0.38	0.24		
			Experimental Graphite Fuel Spheres - Irradiated						
AVR-1	23,200	3	HNO_3-HF	5	H_2O	0.70	4.07	19.01	22.74
AVR-2	26,400	3	HNO_3-HF	5	H_2O	2.50	1.19	34.86	25.10
			GAIL-3A Fuel						
3A-1	8,800	3	HNO_3-HF	5	H_2O	0.90	1.11	19.22	15.66
3A-2	10,400	3	HNO_3-HF	5	H_2O	0.96	4.0	9.54	10.43

[a]Based on cesium measurements at ORNL.

premise that the leaching of fission products might be more effective. (Nitric acid containing fluoride as a catalyst would be essential for leaching coated-particle fuels containing thoria.)

In summary, the hot-cell processing, by the grind-leach method, of pyrocarbon-coated $(Th,U)C_2$ dispersed in graphite resulted in unleachable losses of from 0.4 to 4.5% of the uranium and from 0.4 to 6.9% of the thorium. These losses are not reproducible for 10- to 20-g samples of the same fuel compact, a peculiarity that is charged to inadequate mixing of pulverized samples. Also, the losses appear to be independent of the fuel irradiation level (9600 to 36,400 Mwd per metric ton), in-pile temperature (790 to 1750°C), and extent of particle coating failure. The data obtained on fission product leaching also fail to correlate with irradiation conditions. As much as 35% of the gross gamma activity was not leached; ^{106}Ru was regularly the most resistive to leaching. As much as 42% of the ^{144}Ce and 8.3% of the ^{137}Cs were also held by the graphite residue.

Our results suggest that the unleachable heavy metals had become fixed in the relatively inert pyrolytic carbon coatings. Although fission recoil is one possible mechanism for their presence, diffusion at the high temperatures prevailing during the particle coating process is a more likely source. Further diffusion of the thorium and uranium, as well as the migration of fission products, appears to take place during irradiation.

Thus, we conclude that the grind-leach head-end method is an unattractive means for processing first-generation HTGR fuels [i.e., those containing pyrocarbon-coated $(Th,U)C_2$ particles] owing to the poor recoveries of the fuel values and the extensive holdup of heat-generating radioelements by the graphite waste. In addition, the process would present formidable engineering problems in radiochemical plant design and operation, such as safe management of radioactive dust generated by fine-grinding of the fuel, powder-liquid mixing and separation problems, and a relatively low processing capacity, which would necessarily be imposed by criticality limitations.

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7. APPENDIX

Table A-1. Runs PB-1, -2.[a]

	Recovered from Residue (%)					Left in Graphite Residue (%)
	Leach		Wash			
	1	2	1	2	3	
PB-1						
U	68.43	20.53	8.68	1.42	0.51	0.44
Th	72.27	17.88	5.71	1.08	0.76	2.74
PB-2						
U	83.89	13.41	2.55	0.15	0.02	0.15
Th	82.75	14.52	2.58	0.16	0.02	0.009

[a]Conditions:

Fuel: 10 g of unirradiated GAIL-3A; avg. composition 2.6% U, 13% Th, 84.4% C.

Size Reduction: -100 mesh.

Leaches: Two 25-ml aliquots of 13 M HNO_3--0.05 M HF, refluxed successively 5 hr and 2 hr.

Washes: Three 25-ml aliquots of water; 30 min to 1 hr at 25°C.

Table A-2. Runs PB-3, -4.[a]

	Recovered from Residue (%)							Left in Graphite Residue (%)
	Leach		Wash					
	1	2	1	2	3	4	5	
PB-3								
U	76.07	20.34	3.53	0.003	0.02	0.003	Nil	0.02
Th	78.65	17.47	3.57	0.20	0.03	0.06	0.01	0.002
PB-4[b]								
U	73.42	22.43	4.12	Nil	0.002	0.002	0.001	0.02
Th	73.87	23.22	3.59	0.27	0.09	0.02	0.01	0.001

[a]Conditions:

Fuel: 10 g of unirradiated GAIL-3A; avg. composition 2.6% U, 13% Th, 84.4% C.

Size Reduction: -100 mesh.

Leaches: Two 25-ml aliquots of 13 M HNO_3--0.05 M HF, refluxed successively 5 hr and 2 hr.

Washes: Five 25-ml aliquots of 13 M HNO_3--0.05 M HF; 30 min to 1 hr at 25°C.

[b]13 M HNO_3 used for leaching and washing in PB-4; other conditions same as in PB-3.

Table A-3. Runs PB-6, -7, -8, -9.[a]

	Recovered from Residue (%)				Left in Graphite Residue (%)
	Leach				
	1	2	3	4	
PB-6					
U	99.55	0.22	0.04	0.03	0.16
Th	98.80	0.79	0.09	0.06	0.26
PB-7					
U	99.61	0.15	0.06	0.04	0.14
Th	98.91	0.67	0.09	0.06	0.27
PB-8					
U	99.60	0.13	0.07	0.04	0.16
Th	98.86	0.67	0.12	0.06	0.29
PB-9					
U	99.56	0.12	0.08	0.05	0.19
Th	98.81	0.67	0.12	0.07	0.33

[a]Conditions:

Fuel: Approximately 15 g of unirradiated GAIL-3A; avg. composition 2.6% U, 13% Th, 84.4% C.

Size Reduction: -100 mesh.

Leaches: Four successive leaches with 50-ml aliquots of 13 M HNO_3, refluxed 3, 2, 2, and 2 hr. Following each leach, residue washed with three 25-ml aliquots of water, 10 min each at 100°C.

Table A-4. Runs 3B-1, -2.[a]

	Recovered from Residue (%)							Left in Graphite Residue (%)
	Leach		Wash					
	1	2	1	2	3	4	5	
3B-1								
U	83.94	12.70	1.77	0.57	0.23	0.16	0.07	0.54
Th	79.70	15.50	2.66	0.59	0.30	0.22	0.15	0.90
Gr γ	81.3	13.5	1.5	0.67	0.29	0.24	0.03	2.48
Ru γ	69.34	10.83	1.63	0.69	0.26	0.22	0.02	16.14
Zr γ	95.57	3.09	0.42	0.18	0.06	0.06	--	0.60
Cs γ	83.21	13.27	1.52	0.56	0.28	0.25	0.02	0.90
Gr β	81.96	14.30	1.65	0.69	0.33	0.25	0.03	8.25
TRE β	76.21	12.42	1.60	0.65	0.30	0.23	0.02	8.56
3B-2								
U	78.26	14.67	4.46	0.27	0.11	0.05	0.08	2.15
Th	82.54	11.05	2.60	0.32	0.13	0.04	0.08	3.29
Gr γ	79.40	8.84	2.25	0.30	0.09	0.07	0.04	8.99
Ru γ	47.37	5.36	1.94	0.01	0.06	0.03	0.01	45.21
Zr γ	79.80	8.98	3.24	0.37	0.12	0.17	0.10	7.23
Cs γ	83.48	9.04	3.04	0.30	0.09	0.06	0.03	3.91
Gr β	70.59	5.65	1.93	1.84	0.54	0.38	0.21	18.82
TRE β	58.99	6.54	2.30	1.84	0.65	0.51	0.28	29.03

[a]Conditions:

Fuel: 10 g HTGR GAIL-3B fuel, irradiated to 33,500 Mwd per metric ton of (U + Th), decayed 2 years.

Size Reduction: -100 mesh.

Leaches: Two 25-ml aliquots of 13 M HNO_3, refluxed successively 5 hr and 2 hr.

Washes: Five 25-ml aliquots of 13 M HNO_3; 30 min to 1 hr each at 25°C.

Table A-5. Runs 3B-3, -4.[a]

	Recovered from Residue (%)							Left in Graphite Residue (%)
	Leach		Wash					
	1	2	1	2	3	4	5	
3B-3								
U	56.25	33.60	7.91	0.97	0.44	0.25	0.14	0.44
Th	56.90	32.47	5.83	0.46	0.19	0.19	0.19	3.82
Gr γ	57.34	26.06	7.38	1.13	0.35	0.23	0.10	7.38
Ru γ	43.61	22.33	6.44	1.04	0.31	0.21	0.10	25.96
Zr γ	57.29	29.54	7.52	1.25	0.04	0.04	0.02	4.30
Cs γ	60.71	26.93	7.34	1.22	0.34	0.23	0.09	3.13
Gr β	48.51	21.66	5.89	0.87	0.28	0.17	0.10	22.52
TRE β	45.04	20.11	5.63	0.86	0.27	0.16	0.05	27.88
3B-4								
U	57.93	27.03	13.05	0.43	0.10	0.04	0.02	1.40
Th	76.87	17.47	4.05	0.49	0.14	0.05	0.03	0.92
Gr γ	65.30	19.43	6.11	0.70	0.17	0.06	0.03	8.21
Ru γ	49.67	12.15	5.53	0.76	0.22	0.11	0.11	31.45
Zr γ	51.53	19.39	7.16	1.10	0.29	0.04	0.04	20.45
Cs γ	69.23	19.85	5.09	0.68	0.16	0.06	0.03	4.89
Gr β	53.89	15.16	3.94	0.54	0.13	0.03	0.03	26.27
TRE β	54.33	5.77	3.07	0.43	0.12	0.06	--	36.22

[a]Conditions:

Fuel: 10 g HTGR GAIL-3B, irradiated to 34,100 Mwd per metric ton of (U + Th), decayed 2 years.

Size Reduction: -100 mesh.

Leaches: Two 25-ml aliquots of 13 M HNO_3--0.05 M HF; refluxed successively 5 hr and 2 hr.

Washes: Five 25-ml aliquots of 13 M HNO_3--0.05 M HF; 30 min to 1 hr each at 25°C.

Table A-6. Runs 3B-5, -6.[a]

	Recovered from Residue (%)							Left in Graphite Residue (%)
	Leach		Wash					
	1	2	1	2	3	4	5	
3B-5								
U	71.40	21.58	4.32	0.72	0.54	0.22	0.05	1.29
Th	75.54	19.28	2.03	0.11	0.07	0.11	0.04	2.83
Gr γ	64.88	20.51	3.58	0.49	0.04	0.02	0.01	10.22
Ru γ	40.80	15.80	2.91	0.40	0.04	0.03	0.01	39.99
Zr γ	74.49	18.95	3.05	0.50	0.11	0.09	0.02	2.79
Cs γ	64.42	22.13	4.15	0.51	0.04	0.01	0.01	4.70
Gr β	51.62	17.87	3.08	0.40	0.03	--	--	26.97
TRE β	48.45	15.15	2.80	0.36	0.03	0.01	0.01	33.20

	Recovered from Residue (%)								Left in Graphite Residue (%)
	Leach			Wash					
	1	2	3	1	2	3	4	5	
3B-6									
U	69.23	16.98	11.43	0.77	0.16	0.03	0.02	0.01	1.37
Th	73.00	17.52	6.05	0.35	0.07	0.03	0.03	0.03	2.91
Gr γ	61.54	15.24	7.69	0.84	0.16	0.05	0.02	0.01	14.43
Ru γ	38.30	11.66	7.71	0.91	0.17	0.06	--	--	41.25
Cs γ	68.72	16.19	8.31	0.84	0.13	0.06	0.02	--	5.73
Ce γ	40.20	9.46	5.67	0.55	0.09	0.03	0.01	--	43.99

[a]Conditions:

Fuel: 10 g of HTGR GAIL-3B, irradiated to 41,000 (3B-5) and 38,500 (3B-6) Mwd per metric ton of (U + Th); decayed 2 years.

Size Reduction: -140 mesh.

Leaches: Three 25-ml aliquots of 13 M HNO_3--0.05 M; refluxed successively 5 hr, 2 hr, 2 hr.

Washes: Five 25-ml aliquots of 13 M HNO_3--0.05 M HF; 30 min to 1 hr at 25°C.

Table A-7. Runs 3B-7, -8.[a]

	Recovered from Residue (%)								Left in Graphite Residue (%)
	Leach			Wash					
	1	2	3	1	2	3	4	5	
3B-7									
U	77.13	19.62	2.49	0.17	0.03	0.01	--	0.01	0.54
Th	81.59	14.17	3.42	0.38	0.03	0.01	--	0.01	0.40
Gr γ	75.15	14.37	2.11	0.20	0.03	0.01	--	0.02	8.17
Ru γ	44.56	12.63	10.56	0.52	0.07	0.02	--	0.01	31.62
Cs γ	74.73	16.19	3.84	0.18	0.04	0.02	--	0.01	4.98
Ce γ	46.28	10.33	3.35	0.16	0.02	0.01	--	0.01	39.79
3B-8									
U	64.79	27.57	5.51	0.65	0.09	0.01	--	--	1.38
Th	70.30	24.06	4.11	0.40	0.03	0.01	0.01	0.01	1.07
Gr γ	60.08	24.53	4.28	0.41	0.04	0.16	0.05	0.02	10.53
Ru γ	33.56	18.30	4.42	0.63	0.12	--	--	0.04	42.97
Cs γ	79.93	31.18	5.04	0.42	0.05	0.25	0.07	0.02	6.12
Ce γ	41.43	16.05	2.81	0.39	0.06	0.01	0.01	0.01	39.22

[a]Conditions:

Fuel: 10 g of HTGR GAIL-3B, irradiated to 36,200 (3B-7) and 33,300 (3B-8) Mwd per metric ton of (U + Th), decayed 2 years.

Size Reduction: -140 mesh.

Leaches: Three 25-ml aliquots of 13 M HNO_3, refluxed successively 5 hr, 2 hr, 2 hr.

Washes: 3B-8: Two 25-ml aliquots of C_2H_5OH; three 25-ml aliquots of water; 30 min to 1 hr at 25°C.

3B-7: Five 25-ml aliquots of water.

Table A-8. Runs 3B-9, -10, -11.[a]

	Recovered from Residue (%)				Left in Graphite Residue (%)
	Leach				
	1	2	3	4	
3B-9					
U	96.54	0.77	0.09	0.06	2.54
Th	94.63	0.80	0.08	0.03	4.45
Gr γ	79.78	1.47	0.30	0.20	18.22
Ru γ	77.75	13.87	3.50	1.30	3.58
Cs γ	91.68	2.39	0.36	0.13	5.44
Ce γ	59.37	2.50	0.87	0.37	36.89
3B-10					
U	94.47	0.96	0.07	0.04	4.46
Th	91.80	1.17	0.08	0.04	6.89
Gr γ	88.97	1.55	0.44	0.24	8.80
Ru γ	87.77	7.70	1.78	1.03	1.72
Cs γ	89.81	1.42	0.28	0.19	8.30
Ce γ	55.76	1.25	0.48	0.34	42.17
3B-11					
U	97.93	0.27	0.08	0.05	1.88
Th	93.05	0.56	0.10	0.02	6.27
Gr γ	97.25	1.13	0.40	0.28	0.93
Ru γ	89.81	4.42	1.82	1.26	2.69
Cs γ	90.35	0.83	0.33	0.20	8.29
Ce γ	57.31	0.66	0.49	0.38	41.16

[a]Conditions:

Fuel: ~20 g of HTGR GAIL-3B fuel, irradiated to 36,800 (3B-9), 30,600 (3B-10), and 25,100 (3B-11) Mwd per metric ton of (U + Th), decayed 2 years.

Size Reduction: -100 mesh.

Leaches: Four successive leaches with 50-ml aliquots of 13 M HNO_3, refluxed 3, 2, 2, and 2 hr. Following each leach, residue was washed with three 25-ml aliquots of water, 10 min each at 100°C.

Table A-9. Runs 3A-1, -2.[a]

	Recovered from Residue (%)								Left in Graphite Residue (%)
	Leach			Wash					
	1	2	3	1	2	3	4	5	
3A-1									
U	77.65	17.69	3.25	0.48	0.01	0.02	--	--	0.90
Th	77.41	14.80	6.05	0.51	0.10	0.02	--	--	1.11
Gr γ	62.31	14.53	3.11	0.51	0.10	0.02	--	--	19.22
Ru γ	40.48	18.41	13.14	0.21	--	--	--	--	27.76
Cs γ	71.65	15.42	3.00	0.44	--	--	--	--	9.48
Ce γ	48.12	11.06	24.13	2.61	--	--	--	--	14.04
Gr β	65.70	15.09	3.09	0.46	--	--	--	--	15.66
3A-2									
U	82.11	13.70	2.89	0.21	0.10	0.02	0.01	--	0.96
Th	79.75	11.51	4.0	0.45	0.22	0.03	0.02	0.01	4.0
Gr γ	71.24	15.53	3.11	0.44	0.11	0.01	0.01	--	9.54
Ru γ	21.58	68.44	3.73	0.58	--	--	--	--	5.67
Cs γ	75.49	12.62	1.27	0.42	--	--	--	--	10.20
Ce γ	80.47	12.87	2.99	0.46	--	--	--	--	3.21
Gr β	74.19	12.15	2.82	0.41	--	--	--	--	10.43

[a]Conditions:

Fuel: 20 g of General Atomic GAIL-3A fuel, irradiated to 8,800 (3A-1) and 10,400 (3A-2) Mwd per metric ton of (U + Th), decayed 4.5 years.

Size Reduction: -100 mesh.

Leaches: Three 50-ml aliquots of 13 M HNO_3--0.05 M HF; refluxed successively 5, 2, and 2 hr.

Washes: Five 50-ml aliquots of water; 30 min to 1 hr each at 25°C.

Table A-10. Runs GA-1, -2, -3.[a]

	Recovered from Residue (%)				Left in Graphite Residue (%)
	Leach-Wash				
	1	2	3	4	
GA-1					
Leach, hr	5	3			
U	98.82	0.28	--	--	0.90
Th	98.36	0.60	--	--	1.04
GA-2					
Leach, hr	3	4			
U	99.40	0.27	--	--	0.33
Th	98.63	1.10	--	--	0.27
GA-3					
Leach, hr	3	2	2	2	
U	95.49	2.26	0.91	0.95	0.38
Th	94.49	4.93	0.31	0.02	0.24

[a]Conditions:

Fuel: 20 g of unirradiated General Atomic experimental graphite fuel; avg. composition 3.6% U, 2.2% Th, 94.2% C.

Size Reduction: -100 mesh.

Leach-Wash: Fuel sample leached with 50-ml aliquot of boiling 13 M HNO_3--0.05 M HF for time indicated, then given three to five 25-ml water washes at 25°C. Procedure repeated one to three times.

Table A-11. Runs AVR-1, -2.[a]

	Recovered from Residue (%)								Left in Graphite Residue (%)
	Leach			Wash					
	1	2	3	1	2	3	4	5	
AVR-1									
U	65.88	28.12	4.22	0.51	0.25	0.06	0.01	0.05	0.70
Th	53.78	32.05	7.95	1.58	0.45	0.13	--	--	4.07
Gr γ	52.67	20.99	5.54	1.07	0.32	0.12	0.08	0.20	19.01
Ru γ	12.13	5.72	1.75	0.33	--	--	--	--	80.09
Cs γ	61.23	24.92	5.98	1.14	--	--	--	--	6.75
Ce γ	55.88	21.50	5.13	0.93	--	--	--	--	16.55
Gr β	51.21	20.23	4.90	0.92	--	--	--	--	22.74
AVR-2									
U	71.61	19.56	4.72	1.33	0.20	0.04	--	--	2.50
Th	58.88	28.41	8.42	2.84	0.26	--	--	--	1.19
Gr γ	45.44	12.25	5.04	1.86	0.39	0.16	0.02	--	34.86
Ru γ	69.97	19.88	9.97	--	--	--	--	--	0.17
Cs γ	69.10	18.25	6.08	--	--	--	--	--	6.53
Ce γ	61.91	18.29	5.50	--	--	--	--	--	14.31
Gr β	55.35	14.70	4.84	--	--	--	--	--	25.10
TRE β	54.73	14.17	4.74	--	--	--	--	--	26.36

[a]Conditions:

Fuel: 20 g of experimental graphite fuel, irradiated to 23,200 (AVR-1) and 26,400 (AVR-2) Mwd per metric ton of (U + Th) and decayed 3 years.

Size Reduction: -100 mesh.

Leaches: Three 50-ml aliquots of 13 M HNO_3--0.05 M HF; refluxed successively 5, 2, and 2 hr.

Washes: Five 50-ml aliquots of water; 30 min to 1 hr at 25°C.

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