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Determination of Fission Product and Heavy Metals Inventories in FTE-4 Fuel Rods by a Grind-Burn-Leach Flowsheet

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DETERMINATION OF FISSION PRODUCT AND HEAVY METALS INVENTORIES IN FTE-4
FUEL RODS BY A GRIND-BURN-LEACH FLOWSHEET

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ABSTRACT

Experiments using High-Temperature Gas-Cooled Reactor (HTGR) fuel material, TRISO-coated $(2.75 \text{ Th/U})\text{C}_2$ --TRISO-coated ThC_2 and TRISO-coated UO_2 --BISO-coated ThO_2 , were performed in Building 4507 (the High-Level Chemical Development Facility) to determine the inventory and transport behavior of fission products and heavy metals from a grind-burn-leach process flowsheet. In addition, values calculated by the ORNL Isotope Generation and Depletion Code (ORIGEN, a computer program used for predicting quantities of activation products, actinides, and fission products from irradiation data and nuclear data libraries) are compared with values derived by chemical analyses (CA) and those measured by a gamma-scan nondestructive analytical (NDA) technique. Reasonable agreement was obtained between ORIGEN and NDA results for one of the tests, but the values obtained by chemical analysis were lower than either of the two other sets of values. With the exception of ^{234}U , isotopic uranium values determined by chemical analysis (mass spectrometry) agreed within 15% of the ORIGEN prediction.

1. INTRODUCTION

These experiments are part of a continuing study of the combustion of irradiated HTGR fuel test specimens and behavior of the heavy metals and fission products in head-end reprocessing steps. Specially fabricated Recycle Test Elements (RTE) with fuel rods of known composition¹ were irradiated under controlled conditions in the Peach Bottom reactor. Additional Fuel Test Elements (FTE) were also irradiated by General Atomic Company (GAC). In these experiments, the major goal was to compare starting inventories by three independent means: (1) the normal method using "wet analytical chemistry" techniques, (2) calculation of materials using the ORIGEN code,^{2,3} and (3) nondestructive gamma spectrometry.⁴ The need for this comparison grows out of a concern for knowing the degree of surface deposition ("plate-out") of materials on the walls of the processing equipment (burner, filters, lines, etc.), which may be determined by the difference between starting and final inventories.

A complete FTE-4 fuel rod was used in each experiment. The fuel rods were ground to a fine powder before burning to minimize the number of processing steps, and hence, losses. After burning, the ashes were leached, and the residues were fused for analysis. The equipment (except for the burner) was disassembled for destructive analysis. Throughout each run, precautions were taken to minimize losses and avoid cross-contamination of samples during handling.

2. FUEL DESCRIPTION

The HTGR fuel rods used in the experiments were coded FTE-4-3-5-7 [TRISO-coated (2.75 Th/U)C₂--TRISO-coated ThC₂] and FTE-4-3-1-8 (TRISO-coated UO₂--BISO-coated ThO₂). They were selected from many rods in a series of irradiation experiments, Fuel Test Elements (FTE), conducted to test candidate fuels. The average rod weight was 11.25 g.⁵ The FTE-4-3-5-7 fuel rod weighed 11.177 g on receipt. It contained 0.5529 g of uranium and 2.378 g of thorium (predirradiation).⁶ The FTE-4-3-1-8 fuel rod weighed 13.925 g and contained 0.5528 g of uranium and 5.184 g of thorium (preirradiation).⁶ Detailed fabrication data were not available.

3. IRRADIATION DATA

The FTE-4-3-5-7 fuel rod was irradiated at the 74.5-in. height in the Peach Bottom Reactor (Core II) to an exposure of 1.126E21 (E > 0.18 MeV) fast fluence and 1.516E21 (E < 2.38 eV) thermal fluence at 878 to 1215°C (av. 1056°C) over a period of 499 effective full-power days (EFPD) from day 252 (July 8, 1971) to day 701 (September 14, 1973).

Radiochemical analyses were performed between December 1975 and July 1976; activities were corrected for decay to April 1, 1975 to coincide with the date of the gamma scan.⁴ Post-irradiation examination data are to be published by General Atomic Company (GAC).⁶

While results of ORIGEN and nondestructive analysis (NDA) by gamma-scanning were in relatively good agreement ($\pm 20\%$) for FTE 4-3-5-7, there was difficulty in interpreting the scan data for FTE-4-3-1-8 due to fractures in the fuel rod. Less emphasis will be placed on the results of FTE-4-3-1-8 in the comparison analysis in this report due to the lack

of agreement between ORIGEN and NDA for FTE-4-3-1-8. However, all of the results are included for completeness.

4. SYSTEM CONFIGURATION

The hot-cell system is shown in Fig. 1. The feed gases (Ar, Ar-4% H_2 , N_2 , O_2 , and/or CO_2) are metered into the hot cell through a water bubbler and then to the appropriate apparatus (crusher, burner, or leacher system). Between the burner and the gas receiver, the gases flow through an absolute filter assembly, detectors measuring CO , CO_2 , and radiation level, a CuO bed, and a molecular sieve adsorber (to remove 3H_2O from the stream). The entire gas volume was accumulated and sampled at the end of each run. The CuO oxidizer serves to convert 3H_2 to 3H_2O as well as CO to CO_2 (to facilitate ^{14}C determinations). The burner, off-gas line (minus top flange), and absolute filters are shown in Fig. 2. The burner is a slightly tapered stainless steel cylinder approximately 30 cm long, which is placed in a well-type furnace. The base of the burner is fitted with a stainless steel tube to supply preheated inlet gas. A small perforated plate is welded into the burner base about 1 cm above the gas supply tube. The plate serves to diffuse the incoming gas and support the fuel material during combustion.

The burner has a flange on the top with annular recesses to hold a sintered-nickel filter with 5- μm -diam pores and a gold-plated gasket. Two thermocouples are in thermowells attached to the burner body (one inside the burner and one on the outside) to monitor the temperatures during the reaction. One additional thermocouple is mounted in the top

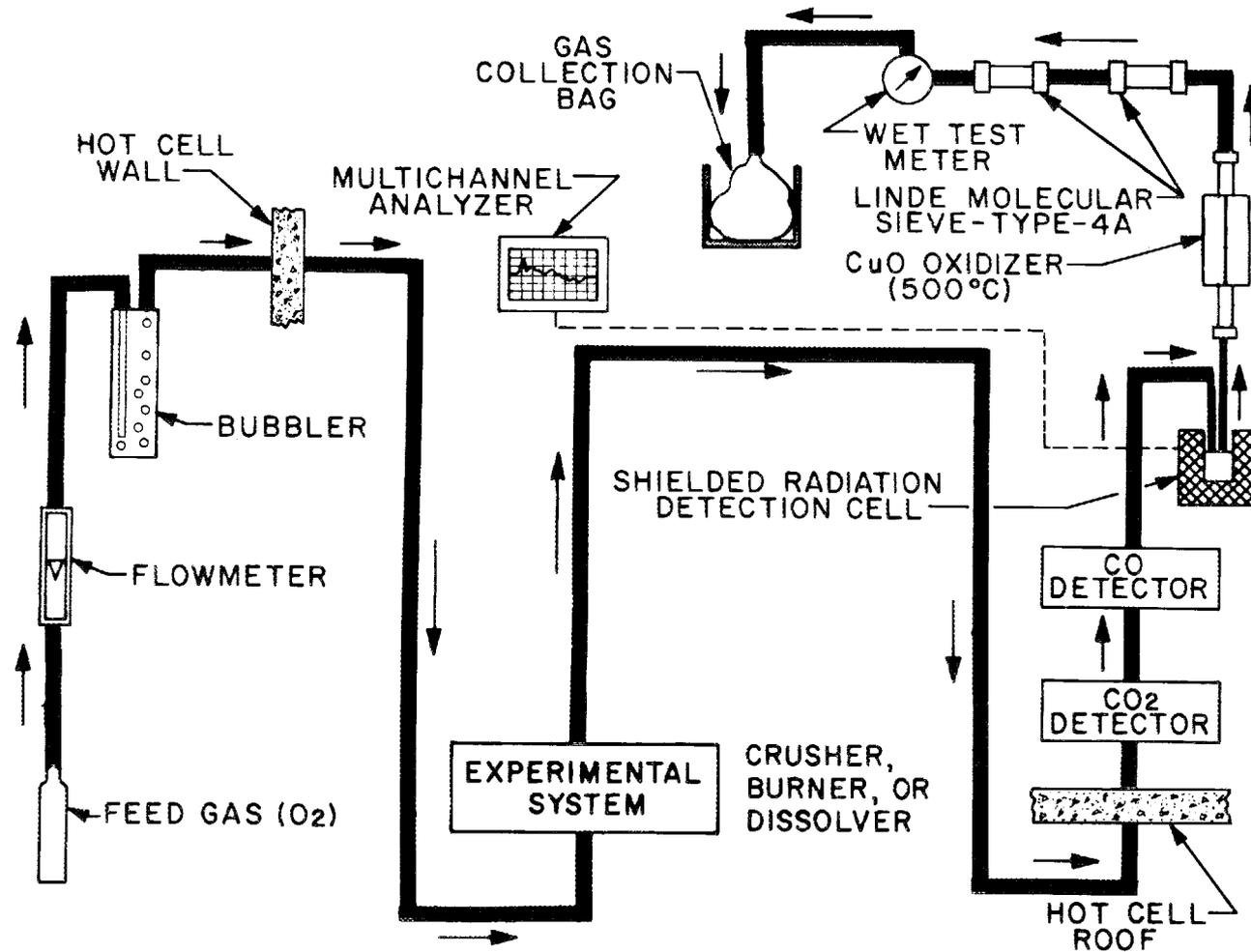


Fig. 1. Instrumentation and off-gas monitoring systems.

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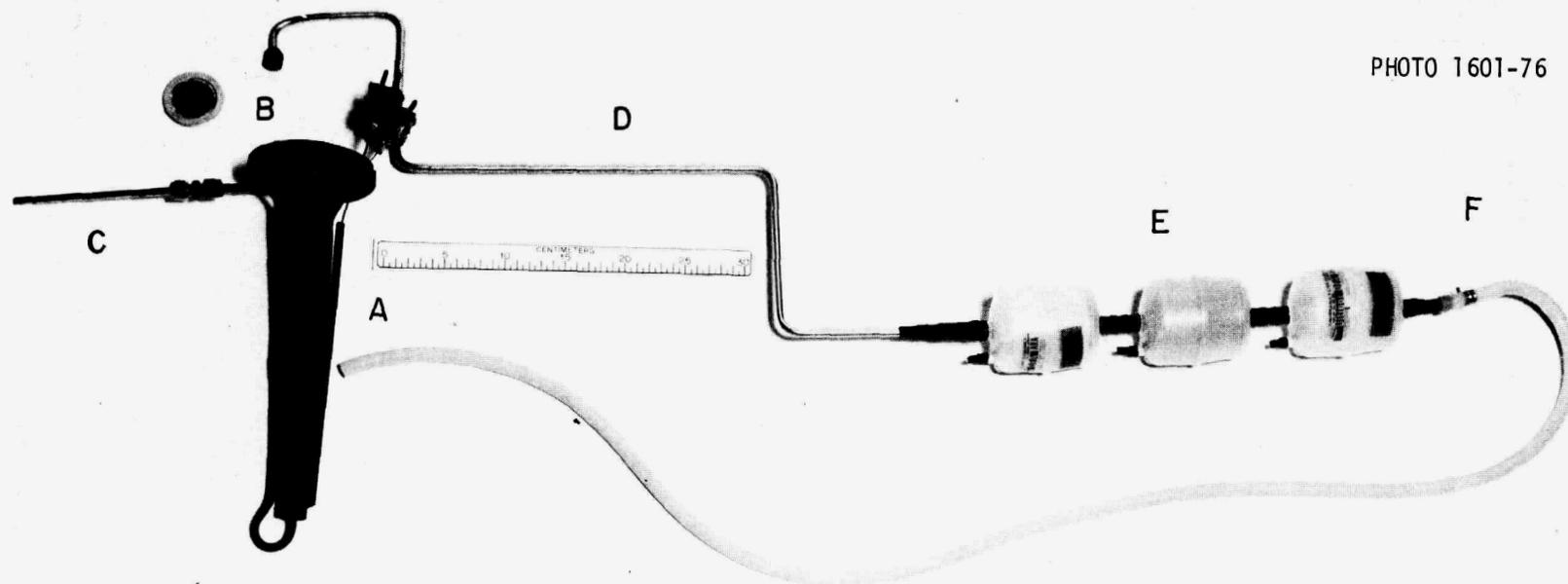


Fig. 2. Burner and off-gas system. A, burner; B, sintered-nickel filter; C, inlet gas line; D, off-gas line; E, absolute filter assembly; F, line to gas collection bag.

flange (not shown) to measure the temperature of the sintered-nickel filter. The off-gas line is a 0.635-cm-OD x 91.4-cm-long stainless steel tube attached to the top flange of the burner and the absolute-type filter assembly (Fig. 2). The filters are provided to collect entrained and volatilized radionuclides from the burner off-gas. The absolute filters used in this experiment were pretested, disposable, polypropylene-encased HEPA filters, No. MBY 3001 URA, made by Pall, Trinity, Inc.

5. OPERATING PROCEDURES, CONDITIONS, AND RESULTS

The FTE-4-3-5-7 and FTE-4-3-1-8 fuel rods were processed by the flowsheets in Figs. 3 and 4, respectively, described below.

5.1 Blending

5.1.1 Blending procedure

Each fuel rod was pulverized in a blender (Fig. 5) fitted with sintered-metal filters and connectors for attaching gas purge lines. Argon was used for the carbide-type fuel, while air was used to purge the oxide-type fuel. The blending cycle consisted of a 2-min mixing period followed by a 2-min resting period in order to prevent overheating of the unlubricated blender motor bearings. The blending cycle was repeated for a total mixing time of 30 min. A final purge was made for 30 min at a rate of 200 cc/min. The blender jar was emptied into the burner. A dry argon atmosphere was maintained to prevent exposure of the pulverized fuel to air and moisture and to minimize the oxidation of the carbides. To aid in the quantitative transfer of the fuel, the blender jar was flushed three times with ~ 1 g (by weight) of fresh carbon. After each addition of carbon, the blender was activated for

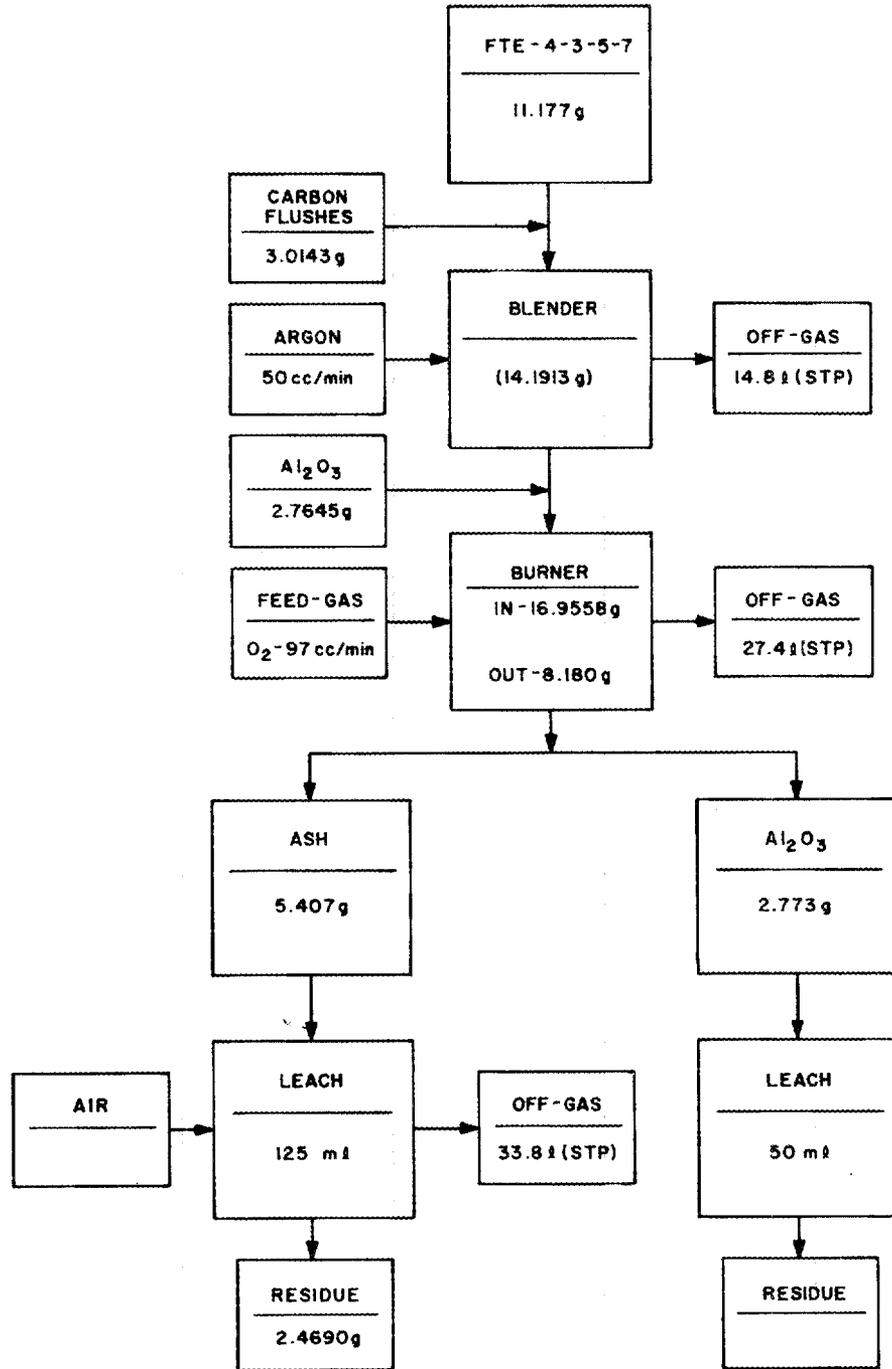


Fig. 3. Flowsheet for FTE-4-3-5-7.

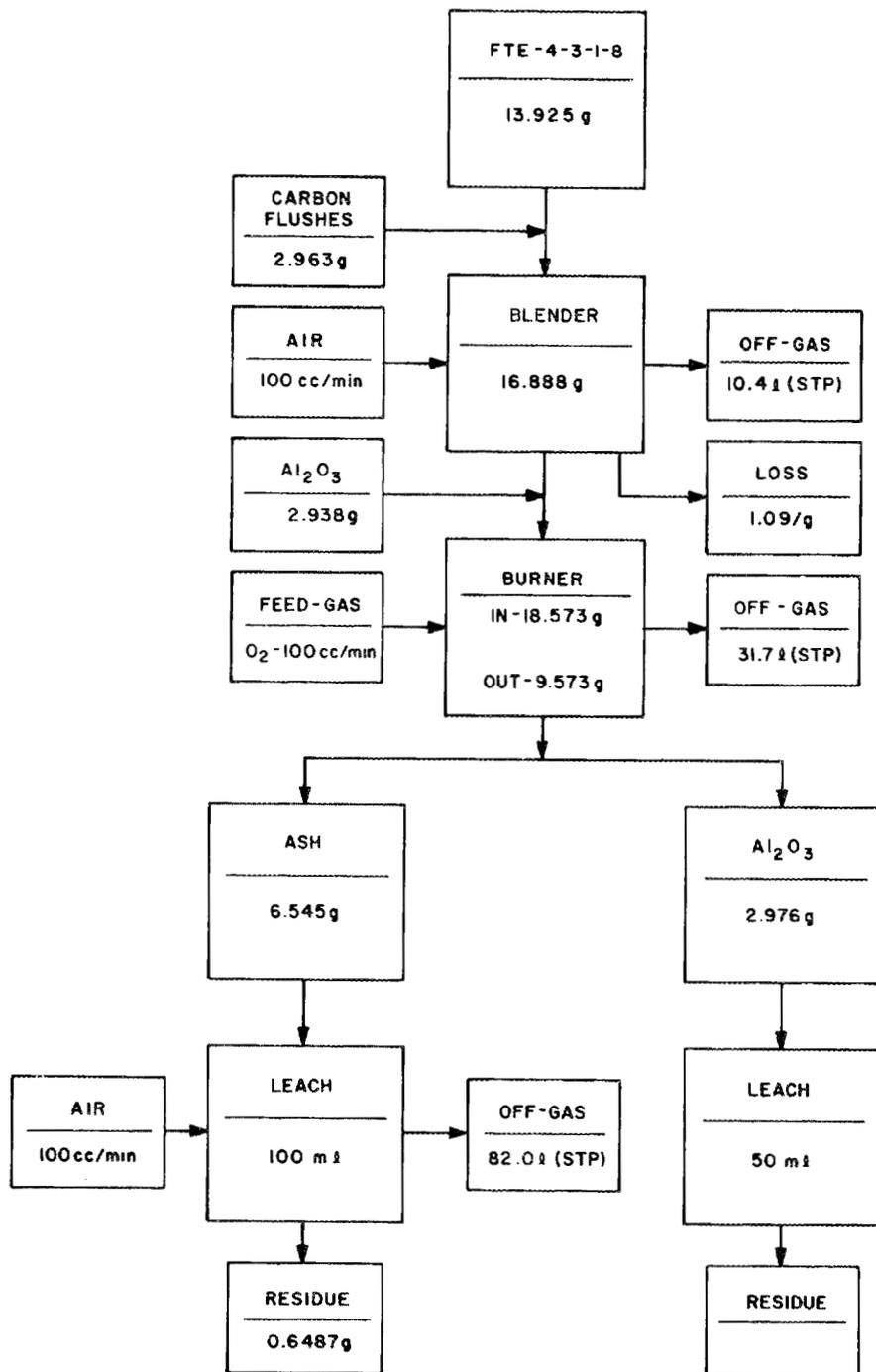


Fig. 4. Flowsheet for FTE-4-3-1-8.

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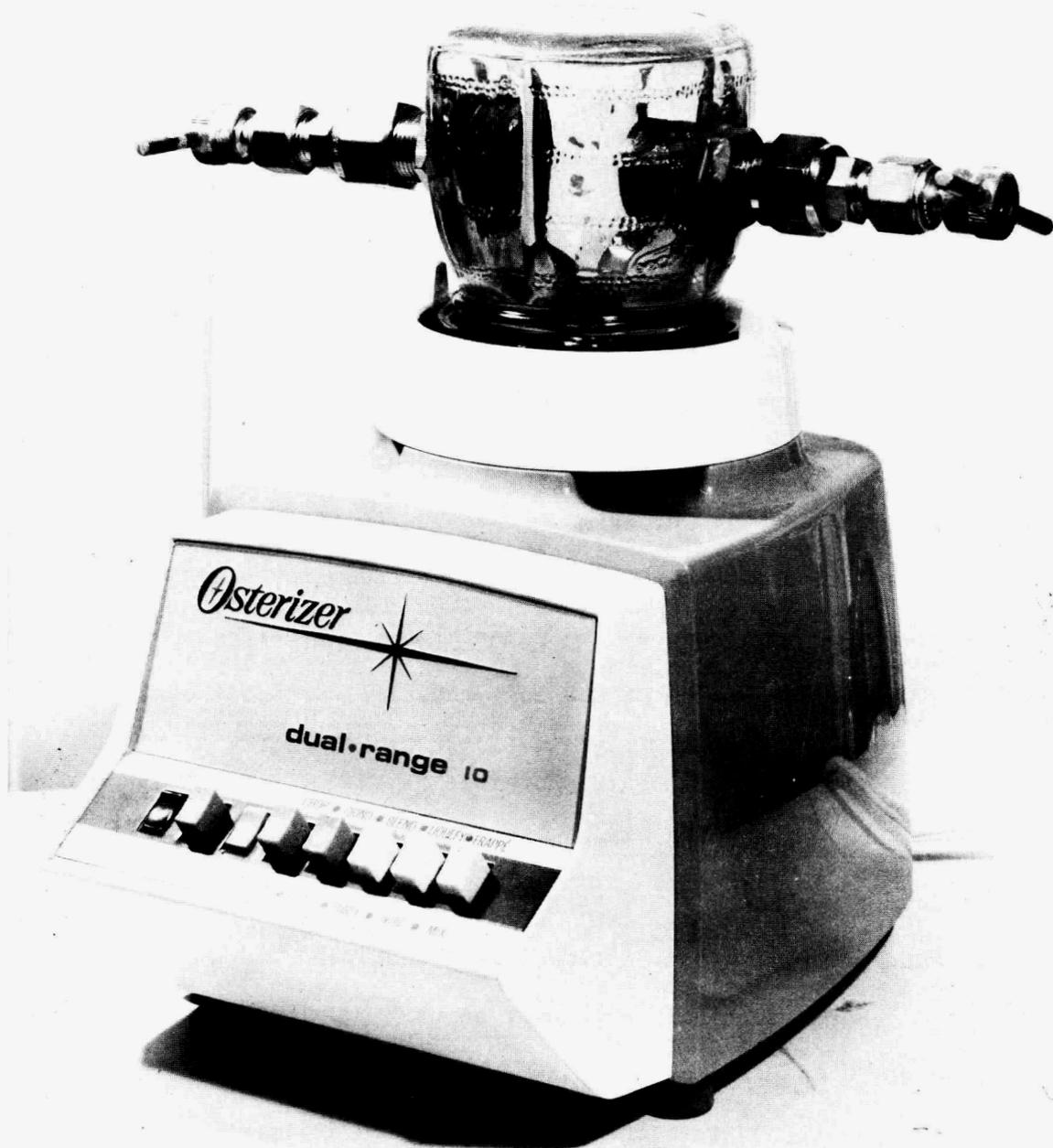


Fig. 5. A photograph of the blender that has been fitted with filters and connectors.

22 min. The blender was then brushed as clean as possible to transfer the fuel and carbon adhering to the inner walls of the burner.

5.1.2 Results

The blending operation for FTE-4-3-5-7 proceeded smoothly with no apparent problems. The need to control the atmosphere with argon precluded both sieving (to determine the adequacy of particle crushing) and weighing of the blender product. The collected purge gas containing the released ^{85}Kr had a radiation level of ~ 500 mr/hr at contact with a beta-gamma "cutie pie" survey meter.

The blending operation for FTE-4-3-1-8 was marred by an unusually large spill of powder (~ 1.1 g) during one of the carbon flushes. To avoid cross-contamination, no attempt was made to recover the powder. The 1.1-g loss represents 6.5% of the total blender inventory or 7.8% of the fuel-rod weight; however, no material balance correction was made because the composition of the lost material was not known.

5.2 Burning

5.2.1 Burning procedure and disassembly

The burner was prepared to receive the fuel by placing 15 alumina beads (Al_2O_3) in the bottom of it. Each bead had a 1/8-in. OD (3.175-mm OD), and the total weight was about 3 g. The beads were added to protect the supporting metal plate and to effect an even dispersion of the inlet gas supply into the powdered fuel during the burn.

The burner was sealed and tested for leaks after all the crushed fuel and carbon flushes were added. After its integrity was verified, heat was applied. Oxygen was admitted into the system at about 100 cc/min

(STP) during which the temperature was raised slowly to 750°C and maintained at this level for the duration of the burn. It was assumed that the fuel rod was completely burned when the concentration of CO₂ in the off-gas decreased to zero. The system was then purged with an additional 6 liters of oxygen at a rate of 200 cc/min.

The equipment was remotely disassembled and separated into the following components: (1) burner, (2) off-gas line, (3) filter assemblies, and (4) off-gas collection bag.

The burner assembly was dismantled, and the 5- μ m sintered-nickel frit was removed, packaged, and transferred to the analytical laboratory, where it was dissolved for chemical analysis. The ashes and the alumina beads were poured from the burner into a beaker. The beads were removed from the beaker, weighed, and leached to recover any adhering fuel ash.

The off-gas line, composed of a length of 91.4-cm stainless steel tubing, was separated from the assembly and cleaned to remove any outer surface radioactive contamination. It was sectioned into about 2.5-cm segments, which were collected into three samples and submitted for gamma analyses at the Radiochemical Analyses Laboratory.

The absolute filter assembly was taken apart by separating the three filter holders at the rubber connections. The gas entry and exit port on each filter holder was sealed to prevent entry of solution, and the outer surface of each filter holder was washed to remove any radioactive contamination. The filters were transferred to the Radiochemical Analyses Laboratory for gamma spectral analysis and returned to the Chemical Development group. Each filter holder was cut open, and the

paper filter was removed, repackaged, labeled, and then transferred to the Fission Product Surveillance Laboratory for gamma measurements using extended counting.

A special plastic bag was sealed onto the downstream end of the equipment line to receive all gases exhausted during the operation. The gases were accumulated, and samples were withdrawn for ^{14}C and ^{85}Kr at the conclusion of each operational step. (The gases were a mixture of combustion products, liberated gases, and the cover gas.)

5.2.2 Burning results

The burning operations proceeded smoothly with good control of temperature for both rods (Figs. 6 and 7). Burning lasted ~ 3 hr at an average temperature of 750°C . Off-gas compositions of CO_2 ranged from 75 to 80%, with CO concentrations varying between 6 and 23% for the carbide fuel. For FTE-4-3-1-8, the concentration of CO_2 exceeded 90% near the end of the run; however, the CO concentration never exceeded about 5%. These results represent typical burner performances. The radiation levels of the gas bag (about 30 liters) for the carbide fuel exceeded the range of the cutie pie (10 R/hr) at contact. The burner product was composed of 5.407 g of ash (48.4% of the fuel rod) and 2.773 g of alumina beads for FTE-4-3-5-7, and 6.545 g of ash (47.0% of the fuel rod) with 2.976 g of alumina beads for FTE-4-3-1-8. (This latter percentage could have been as high as about 50% if there had been no loss in blending.)

5.3 Leaching

5.3.1 Leaching procedure

The material removed from the burner consisted of the alumina beads

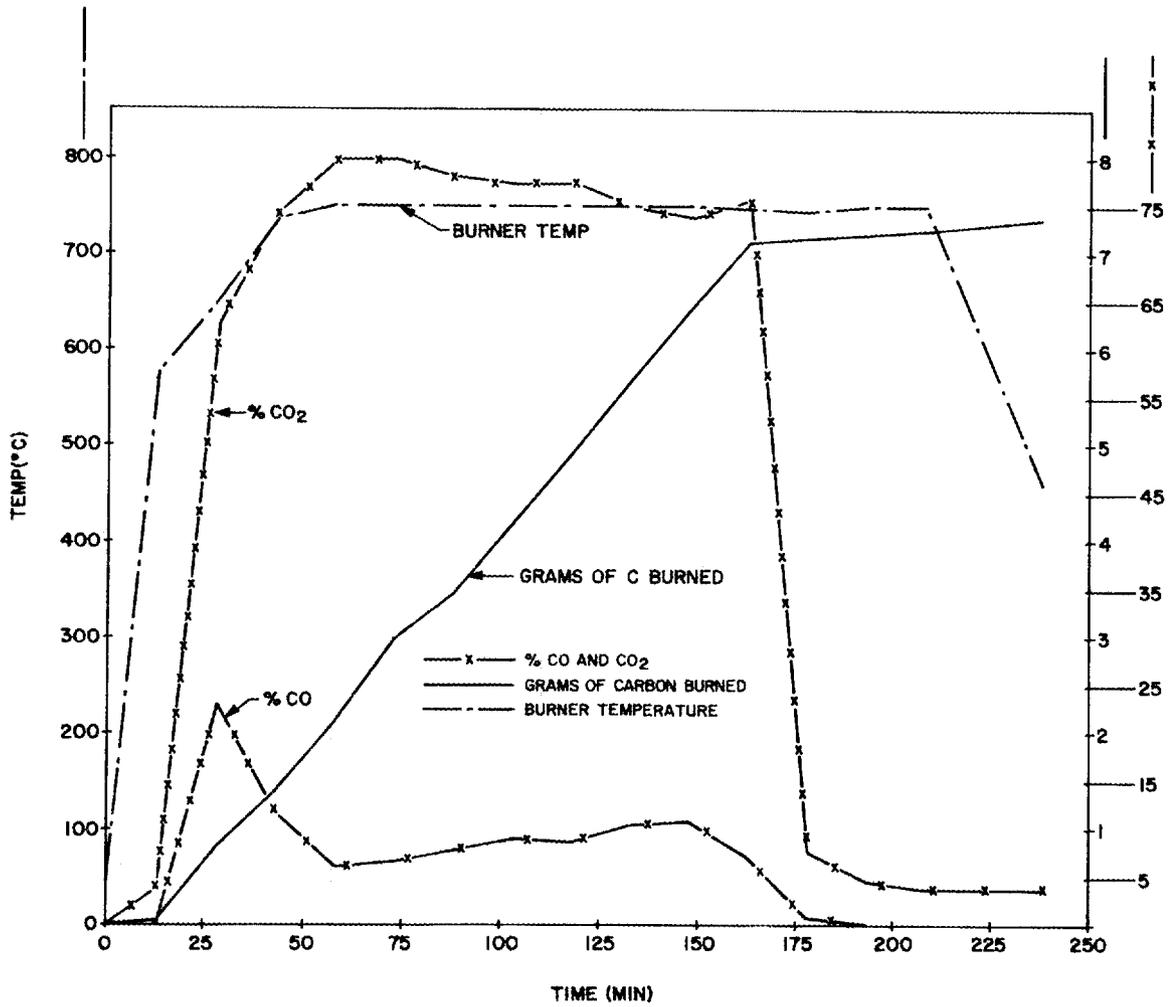


Fig. 6. History of the burner run for FTE-4-3-5-7.

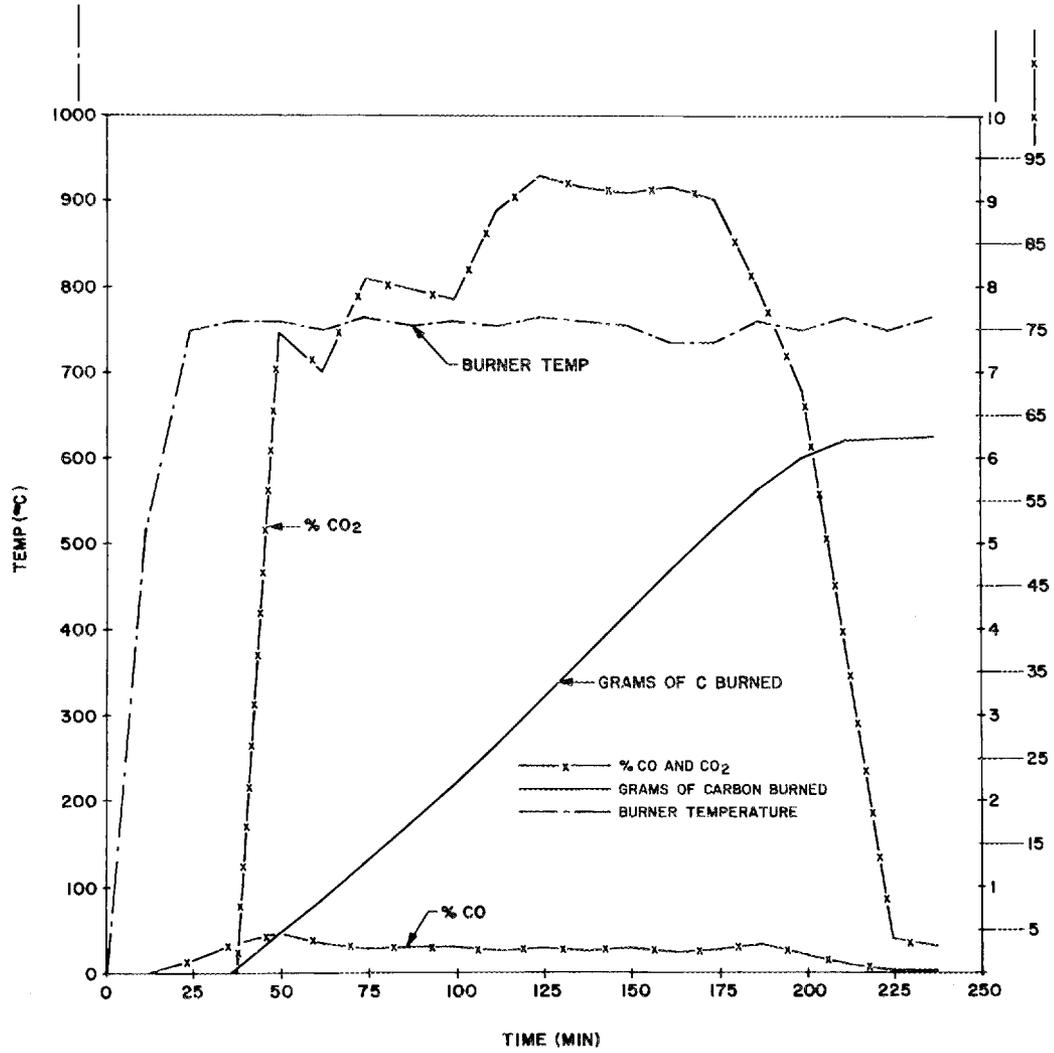


Fig. 7. History of the burner run for FTE-4-3-1-8.

and the ashes resulting from the burned fuel stick. Each of the products was separately leached under reflux conditions in about 50 ml of Thorex reagent (13 M HNO_3 --0.1 M Al^{3+} --0.05 M F^-). A flow of water-saturated air was maintained at a rate of 50 cc/min through the system during each leach operation. After 12 hr of leaching, the insoluble residue was separated from the Thorex leach solution by centrifugation and decantation. The ash was rinsed with 20 to 25 ml of Thorex reagent and the rinsings combined with the first leach solution. The solution was then diluted with Thorex reagent to a volume of 125 ml. The insoluble residue was dried, weighed, and packaged for analysis. The alumina beads were leached for 2 hr in a refluxing Thorex reagent and then separated, rinsed, and dried. The leach solution was made to a 50-ml volume with Thorex reagent.

Samples of both the leach solutions and the entire insoluble residue were submitted to Analytical Chemistry Division's High-Radiation-Level Analytical Laboratory (HRLAL) where they were treated and redistributed to other specialized groups in the Analytical Chemistry Division for specific determinations.

5.3.2 Leaching results

The burner ash, alumina beads, and sintered-nickel frit were leached separately and the results are shown in Tables 1 and 2.

Dissolution of the burner ash. The dissolver solution normally contains the bulk of the heavy metals and soluble fission products. The residues typically are SiC hulls and finely divided insoluble fission products.

Dissolver solution. The ashes from the burner were leached for 12 hr with Thorex reagent (Tables 1 and 2). The quantities of gamma-emitting radionuclides found in the two leach solutions were about equal.

Table 1. Leach results for FTE-4-3-5-7 fuel: TRISO-coated (2.75 Th/U)C₂--
TRISO-coated ThC₂

	Leach solution	Insolubles			Total leach plus insolubles	% of total as insolubles in burner ash
		First fusion	Second fusion ^a	Total in-solubles		
U, g	0.361	0.008	0.010	0.018	0.379	4.7
Th, g	2.100	0.006	0.041	0.047	2.147	2.2
⁹⁹ Tc, mg	1.338	0.095	0.019	0.114	1.452	7.8
¹²⁹ I, mg	0.010	<0.001	<0.001	<0.001	0.010	~10
<u>Nuclide, dpm</u>						
⁸⁵ Kr	6.13E08 ^b	c	c	c	6.13E08	
⁹⁰ Sr	6.60E11	7.50E08	7.62E08	8.37E09	6.68E11	1.3
⁹⁵ Zr	4.75E10	2.79E09	4.94E08	3.28E09	5.08E10	6.5
⁹⁵ Nb	<1.28E09	<5.52E06	<4.50E07	<5.05E07	<1.33E09	3.8
¹⁰⁶ Ru	3.79E10	5.15E07	2.09E10	2.10E10	5.89E10	35.7
¹¹⁰ Ag	3.81E07	1.73E07	2.20E06	1.95E07	5.76E07	33.9
¹²⁵ Sb	1.07E10	1.50E07	7.26E08	7.41E08	1.14E10	6.5
¹³⁴ Cs	1.98E11	5.80E06	1.83E09	1.84E09	2.00E11	0.9
¹³⁷ Cs	4.65E11	1.86E07	5.53E09	5.55E09	4.71E11	1.2
¹⁴⁴ Ce	3.50E12	1.02E10	1.07E09	1.13E10	3.51E12	0.3
¹⁵² Eu	<1.05E10	<1.08E07	<8.40E06	<1.92E07	<1.05E10	0.2
¹⁵⁴ Eu	1.28E10	5.13E07	8.52E07	1.37E08	1.29E10	1.1
¹⁵⁵ Eu	3.85E09	2.26E07	2.99E07	5.25E07	3.90E09	1.3

^aPart of melt was lost.

^bA shorthand notation for showing exponents; it will be used in subsequent tables.

^cNot analyzed.

Table 2. Leach results for FTE-4-3-1-8 fuel: TRISO-coated UO₂--BISO-coated ThO₂

	Leach solution	Insolubles			Total insolubles	Total leach plus insolubles	% of total as insolubles in burner ash
		R-1-A	R-1	R-1-NFR			
U, g	0.445	0.004	0.001	-	0.005	0.450	1.2
Th, g	4.495	0.017	0.002	-	0.019	4.514	0.4
⁹⁹ Tc, mg	1.15	0.042	0.452	-	0.494	1.644	30.0
¹²⁹ I, mg	0.048	<0.001	-	-	<0.001	0.048	<2.1
<u>Nuclide, dpm</u>							
⁸⁵ Kr	4.68E09	a	a	a	a	4.68E09	
⁹⁰ Sr	7.14E11	3.18E10	1.10E08	-	3.19E10	7.46E11	4.3
⁹⁵ Zr	5.53E10	9.78E08	2.46E09	-	3.44E09	5.87E10	5.9
⁹⁵ Nb	<1.38E09	<8.40E07	<2.65E08	2.61E08	<6.10E08	<1.99E09	30.6
¹⁰⁶ Ru	1.62E10	6.72E11	1.21E11	7.12E08	7.94E11	8.10E11	98.0
¹¹⁰ Ag	1.62E08	4.12E06	<1.35E06	-	<5.47E06	<1.67E08	3.3
¹²⁵ Sb	7.68E09	2.59E09	<4.75E09	3.25E08	<7.67E09	<1.53E10	50.1
¹³⁴ Cs	1.94E11	3.52E09	3.41E10	-	3.76E10	2.32E11	16.2
¹³⁷ Cs	5.05E11	9.00E09	7.88E10	-	8.78E10	5.93E11	14.8
¹⁴⁴ Ce	3.35E12	3.54E10	3.03E10	-	6.57E10	3.42E12	1.9
¹⁵² Eu	<6.00E09	<1.14E08	<9.15E08	-	<1.03E09	<7.03E09	14.6
¹⁵⁴ Eu	1.29E10	8.34E07	8.84E07	-	1.72E08	1.31E10	1.3
¹⁵⁵ Eu	4.15E09	4.97E07	3.54E07	-	8.51E07	4.24E09	2.0

^aNot analyzed.

Insoluble residues. The residue from FTE-4-3-5-7 was separated, dried, and weighed (2.469 g as indicated in Table 1). It was treated by Na_2CO_3 fusion in the HRLAL. The flux was dissolved and prepared for chemical analysis, and the remaining insolubles were subjected to a second Na_2CO_3 fusion. Unfortunately, a hole developed in the platinum crucible, and part of the second melt was lost. The remainder of the flux from the second fusion was dissolved and prepared for chemical analysis also. Analysis of the dissolved flux from each fusion resulted in finding 0.047 g of thorium and 0.018 g of uranium (i.e., 2.2 and 4.7%, respectively) of the total recovered heavy-metal content. These values indicate that a small fraction of the SiC-coated particles remained unbroken after blending and burning. The amounts of typical gamma emitters in the leached residue ranged from ~ 0.2 to 36% of the total amounts found in the experiment.

The residue from FTE-4-3-1-8 required three treatments before it appeared to be dissolved (Table 2). In general, this residue yielded less uranium and thorium (1.2 and 0.4%, respectively) than before, and the results may indicate substantially more of the coated particles (TRISO-UO_2) were broken in the blender than the previous sample. However, the total percentages of the fission products in the residue were much higher for FTE-4-3-1-8 and ranged from ~ 1 to 98% for selected fission products. These residue results may be partially in error.

Leaching of the alumina beads. One concludes from the leach results that the alumina beads had $\sim 0.2\%$ of burner ash attached for FTE-4-3-5-7 and $\sim 0.6\%$ for FTE-4-3-1-8. The uranium and thorium results were consistent with these values; however, the fission products were found in substantially greater amounts in FTE-4-3-1-8 than in FTE-4-3-5-7 (Tables 3 and 4). Major fractions of the total ^{110}Ag and ^{155}Eu were found on the beads. These residue results may be partially in error.

Table 3. Radioactivity leached from the alumina beads and sintered-nickel filter for FTE-4-3-5-7 fuel: TRISO-coated (2.75 Th/U)C₂--
TRISO-coated ThC₂

	Alumina bead		Sintered nickel	
	Leach	%	Filter	%
U, g	<0.001	<0.3	<0.001	<0.1
Th, g	0.002	<0.1	0.002	0.1
⁹⁹ Tc, mg	0.001	0.1	-	-
<u>Nuclide, dpm</u>				
⁹⁰ Sr	7.29E08	0.1	1.94E09	0.3
⁹⁵ Zr	≤3.30E08	0.6	≤3.24E08	0.6
⁹⁵ Nb	≤2.70E07	2.0	≤1.08E07	0.8
¹⁰⁶ Ru	≤6.00E08	1.0	≤1.80E08	0.3
¹¹⁰ Ag	≤7.50E07	56.4	-	-
¹²⁵ Sb	≤3.30E08	2.8	≤1.32E08	1.1
¹³⁴ Cs	7.62E09	3.6	2.41E09	1.2
¹³⁷ Cs	1.69E10	3.4	4.57E09	0.9
¹⁴⁴ Ce	3.81E09	0.1	1.82E09	0.1
¹⁵² Eu	≤1.23E08	1.2	≤4.56E07	0.4
¹⁵⁴ Eu	6.90E07	0.5	≤1.56E07	0.1
¹⁵⁵ Eu	≤1.17E09	22.8	≤6.60E07	1.3

Table 4. Radioactivity leached from the alumina beads and sintered-nickel filter for FTE-4-3-1-8 fuel: TRISO-coated UO₂--BISO-coated ThO₂

	Alumina bead		Sintered nickel	
	Leach	%	Filter	%
U, g	<0.001	<0.3	<0.001	<0.1
Th, g	0.015	0.3	0.005	0.1
<u>Nuclide, dpm</u>				
⁹⁰ Sr	4.98E09	0.6	2.74E10	3.5
⁹⁵ Zr	≤9.90E09	14.4	≤1.80E08	0.3
⁹⁵ Nb	≤1.92E07	1.0	≤4.86E06	0.2
¹⁰⁶ Ru	≤2.28E08	<0.1	1.85E09	0.2
¹¹⁰ Ag	≤2.73E08	62	-	-
¹²⁵ Sb	≤1.32E09	7.9	≤1.02E08	0.6
¹³⁴ Cs	2.85E10	10.9	1.01E09	0.4
¹³⁷ Cs	6.60E10	10.0	2.29E09	0.4
¹⁴⁴ Ce	2.16E11	6.0	2.20E09	<0.1
¹⁵² Eu	≤8.70E08	11.0	≤1.98E07	0.3
¹⁵⁴ Eu	≤6.00E08	4.4	≤1.02E07	<0.1
¹⁵⁵ Eu	≤1.47E09	25.6	≤3.36E07	0.6

Leaching of the sintered-nickel filters. The sintered-nickel filters were dissolved in aqua regia. Uranium and thorium concentrations were essentially at the detection limits and represented $\leq 0.1\%$ of the totals. A dilution was prepared for gamma analysis. Of the isotopes listed in Tables 3 and 4, only ^{90}Sr , ^{106}Ru (one case), ^{134}Cs , ^{137}Cs , and ^{144}Ce were above the background detection limits. The amounts were of the order of 0.1 to 1% of the totals for each isotope in the list.

5.4 Off-Gas Studies

The purge and burner gases were collected and analyzed for ^{85}Kr and ^{14}C (or ^3H). The filters and off-gas lines were analyzed by the methods described under Sect. 5.2.1 - Burning Procedure and Disassembly.

5.4.1 Krypton-85

The yields of ^{85}Kr predicted by ORIGEN are 53 mCi for FTE-4-3-5-7 and 57 mCi for FTE-4-3-1-8. The ^{85}Kr distributions found on the two experiments are given in Tables 5 and 6. The total amounts recovered were 58.7 mCi for FTE-4-3-5-7 and 16.5 mCi for FTE-4-3-1-8. This latter total appears low, however, and it is interesting to note that larger amounts were released by the oxide fuel during grinding and dissolving while the carbide fuel released most of its ^{85}Kr during burning. The values for ^{85}Kr found in the blender and dissolver purges and burner off-gases are given in Tables 5 and 6.

More ^{85}Kr was released during the grinding and leaching of FTE-4-3-1-8 than for FTE-4-3-5-7 (Tables 5 and 6), but FTE-4-3-1-8 had a substantially lower total amount of ^{85}Kr (16 mCi vs 59 mCi). One explanation for this discrepancy is that the 16.57 mCi from FTE-4-3-1-8 burner off-gas is low.

Table 5. Distribution of ^{85}Kr in off-gas system for FTE-4-3-5-7 fuel:
TRISO-coated (2.75 Th/U) C_2 --TRISO-coated ThC_2

Operation	Off-gas volume liters (STP)	Total ^{85}Kr		
		(Σ dpm)	(mCi)	(%)
Grind	14.8	6.65E09	3.0	5.1
Burn	27.4	1.23E11	55.4	94.4
Leach	33.8	6.13E08	0.3	0.5
Total	76.0	1.30E11	58.7	100.0

Table 6. Distribution of ^{85}Kr in off-gas system for FTE-4-3-1-8 fuel:
TRISO-coated UO_2 --BISO-coated ThO_2

Operation	Off-gas volume liters (STP)	Total ^{85}Kr		
		(Σ dpm)	(mCi)	(%)
Grind	10.4	1.79E10	8.06	48.7
Burn	31.7	1.42E10	6.40	38.6
Leach	82.0	4.68E09	2.11	12.7
Total	124.1	3.68E10	16.57	100.0

5.4.2 Carbon-14

Carbon-14 analyses were performed on samples of the burner off-gas for both runs and for the leach of the run with carbide fuel (Tables 7 and 8). Alternate samples were analyzed for CO₂ and for ¹⁴C for the first run, and CO₂ and ¹⁴C analyses were done on all samples in the second. The carbon in the off-gas included flush carbon; therefore the results were adjusted accordingly. These ratios correspond to about 1600 and 450 g N₂/MTHM in the fuel, respectively.³

The ¹⁴C and CO₂ in the leacher purge off-gas is assumed to have come from a residual amount of burner off-gases left in the lines and gas meter since special steps to avoid this contingency were not taken. (It could conceivably have come from unburned carbides during leaching.)

5.4.3 Composition of the burner off-gas

The gases used were composited in a gas collection bag. Samples of the composited gas were taken at the end of each phase of operation.

The quantities of oxygen and carbon dioxide measured in the gas samples are listed in Tables 9 and 10. The samples withdrawn for gas analysis from the FTE-4-3-5-7 combustion contained from ~ 1.2 to 10.9% nitrogen and water vapor. Gas samples from FTE-4-3-1-8 contained from ~ 0.4 to 1.6% water vapor (from the wet-test meter). Nitrogen indicates the degree of air inleakage to the samples.

5.4.4 Deposition of semi-volatiles on the off-gas line and absolute filters

The outer surface of the off-gas line was washed to remove external contamination and sectioned into 36 pieces. These were assembled into three groups (12 pieces each) and labeled: (1) near burner, (2) center section, and (3) just before the absolute filters. Only very small

Table 7. Carbon-14 determinations for FLE-4-3-5-7 fuel: TRISO-coated (2.75 Th/U)C₂--TRISO-coated ThC₂

Operation	Volume liters (STP)	Sample code	C-14 total		CO ₂ (%)
			(dpm)	(mCi)	
Burn	27.4	GB-1	6.17E07	0.027	-
		GB-2	-	-	56.3
		GB-3	6.27E07	0.028	-
		GB-4	-	-	42.0
		GB-5	6.54E07	0.029	-
		GB-6	-	-	51.1
		Av.	6.33E07	0.028	49.8
Leach	33.8	GB-7	1.31E06	<0.001	-
		GB-8	1.12E06	<0.001	-
		GB-9	-	-	2.4
		Av.	1.22E06 ^a	<0.001 ^a	

^a Corrected for carbon added as flushes.

Table 8. Carbon-14 determinations for FTE-4-3-1-8 fuel: TRISO-coated
 UO_2 --BISO-coated ThO_2

Operation	Volume liters (STP)	Sample code	C-14 total		CO ₂ (%)
			(dpm)	(mCi)	
Burn	31.7	GB-1	3.91E07	0.018	43.5
		GB-2	Sample bulb leaked		-
		GB-3	3.38E07	0.015	-
		GB-4	3.04E07	0.014	40.6
		GB-5	4.22E07	0.019	-
		GB-6	3.20E07	0.014	-
		Av.	3.55E07	0.016	42.1

Table 9. Composition of "burn" off-gas samples for FTE-4-3-5-7 fuel: TRISO-coated (2.75 Th/U)C₂--
TRISO-coated ThC₂

	O ₂		CO ₂		O ₂ in CO ₂		C in CO ₂		N ₂ + H ₂ O
	(%)	(g)	(%)	(g)	(%)	(g)	(%)	(g)	(%)
GB-2	42.5	16.62	56.3	30.27	72.7	22.01	27.3	8.26	1.2
GB-4	47.1	18.42	42.0	22.58		16.42		6.16	10.9
GB-6	45.4	17.75	51.1	27.48		19.98		7.50	3.5
Av.	45.0	17.60	49.8	26.78		19.47		7.31	5.2

Table 10. Composition of "burn" off-gas samples for FTE-4-3-1-8 fuel: TRISO-coated UO₂--
BISO-coated ThO₂

	O ₂		CO ₂		O ₂ in CO ₂		C in CO ₂		N ₂ + H ₂ O
	(%)	(g)	(%)	(g)	(%)	(g)	(%)	(g)	(%)
GB-1	36.7	16.61	43.5	27.07	72.7	19.69	27.3	7.38	1.6
GB-4	40.5	18.31	40.6	25.26		18.37		6.89	0.4
Av.	38.6	17.46	42.1	26.17		19.03		7.14	1.0

fractions of materials passed through the sintered-metal filter into the downstream off-gas system and deposited on the lines (Table 11).

5.4.5 Deposition of fission products on the absolute filters

Three absolute filters were used in series. They were disposable, polypropylene encased, 100% pretested assemblies. The effective filter area for each assembly is 1 ft². Each assembly was DOP-tested prior to use to verify its filtering integrity. The outer shell was wrapped in a polyethylene sleeve to reduce the external contamination levels. After being taken from the hot cell, the outer wrapping was removed in a laboratory hood, and the outer surfaces were washed to remove any loose contamination. The whole filter was analyzed for fission product inventory (RAL). These results were so similar for the three filters that they were further disassembled to remove the paper element. The paper elements were analyzed in a low-level counting facility (FPS) for extended times.

The results are given in Tables 11 and 12 for FTE-4-3-5-7. It is clear that the casings were contaminated in spite of the precautions. Only ¹³⁷Cs could be detected on the second and third filter papers, but the levels of ¹³⁷Cs were too near background (and statistically inconclusive) to permit the determination of the relative efficiencies of the filters in the series.

6. EVALUATION OF RESULTS

6.1 Material Balances

Material balances based on overall weights and other hot-cell measurements are presented in Tables 13 and 14 for FTE-4-3-5-7 and 4-3-1-8, respectively. The grand total material balance was 105% for

Table 11. Off-gas entrainment for FTE-4-3-5-7 fuel: TRISO-coated (2.75 Th/U)C₂--
TRISO-coated ThC₂

	Off-gas line			Absolute filter assembly			Experiment total
	1	2	3	1	2	3	
⁹⁹ Tc, µg	≤0.83	≤0.52	≤0.73	-	-	-	1.46E03
<u>Nuclide, dpm</u>							
⁹⁰ Sr dpm	4.58E05	8.35E04	≤7.97E03	-	-	-	6.71E11
⁹⁵ Zr	-	-	-	a	a	a	5.15E10
⁹⁵ Nb	-	-	-	a	a	a	≤1.37E09
¹⁰⁶ Ru	2.07E05	a	a	a	a	a	5.97E10
¹²⁵ Sb	7.40E04	a	a	a	a	a	1.19E10
¹³⁴ Cs	3.91E05	4.37E03	2.04E03	4.07E04	4.38E03	8.79E02	2.10E11
¹³⁷ Cs	5.48E05	7.10E03	3.11E03	4.88E04	7.38E03	1.63E03	4.92E11
¹⁴⁴ Ce	2.33E06	4.31E04	≤1.12E04	6.80E04	~1.61E04	4.60E03	3.52E12
¹⁵² Eu	-	-	-	a	a	a	≤1.07E10
¹⁵⁴ Eu	≤3.25E04	a	a	a	a	a	1.30E10
¹⁵⁵ Eu	-	-	-	a	a	a	5.14E09

^aAt lower limits of detection.

Table 12. Radioactivity on absolute filters (corrected to April 1, 1975)

FTE-4-3-5-7 fuel: TRISO-coated (2.75 Th/U)C₂--TRISO-coated ThC₂

	AF-1		AF-2		AF-3	
	RAL ^a	FPS ^b	RAL ^a	FPS ^b	RAL ^a	FPS ^b
¹⁰⁶ Ru	c	1.55E03	c	d	c	-
¹²⁵ Sb	c	1.78E03	c	-	c	-
¹³⁴ Cs	4.07E04	~180	4.38E03	-	8.79E02	-
¹³⁷ Cs	4.88E04	5.33E03	7.38E03	~40	1.63E03	~50
¹⁴⁴ Ce	6.80E04	-	~1.61E04	-	4.60E03	-
¹⁵⁵ Eu	c	-	c	-	c	-

^aFilter paper in holder.

^bFilter paper removed from holder.

^cAt lower limits of detection.

^dUndetected.

Table 13. Grand total material balances for FTE-4-3-5-7 fuel:
 TRISO-coated (2.75 Th/U)C₂--TRISO-coated ThC₂

Item	Input	Output
Fuel rod		
U	0.553	0.379
Th	2.378	2.149
Carbide carbon	0.302	-
Other carbon	5.552 ^a	-
SiC	2.392 ^b	2.392 ^d
Fission products	0 ^c	0.150 ^d
Oxide oxygen	0	0.365
Total	11.177	5.435
Measured wt.	11.177 ^e	5.407 ^e
Alumina beads	2.765	2.773
Blender flush carbon	3.014	-
Oxygen	33.264	17.594
CO ₂	-	26.773
Grand total	50.220	52.575
Material balance	100%	105%

^aBy difference.

^bFrom residues out of leacher.

^cIncluded in initial uranium and thorium values.

^dCalculated by ORIGEN code.

^eNot included in grand total.

Table 14. Grand total material balance for FTE-4-3-1-8
fuel: TRISO-coated UO₂--BISO-coated ThO₂

Item	Input (g)	Output (g)
Fuel rod		
U	0.553	0.451
Th	5.184	4.534
Oxide oxygen	0.790	0.707
Carbon	6.777 ^a	-
SiC	0.621	0.621
Fission products	- ^b	0.150
Total weight	13.925	6.463
Measured weight	13.925 ^c	6.633 ^{a,c}
Carbon flush	2.963	-
Alumina beads	2.938	2.940
Blender product (loss)	(1.091)	-
Oxygen	33.260	17.46
CO ₂	0	26.17
Grand total	52.00	53.03
Material balance	100%	102%

^aBy difference.

^bUnirradiated.

^cNot included in grand total.

FTE-4-3-5-7 and 102% for FTE-4-3-1-8. The overall material balances (100 x output/input) for individual components are given below:

	Material balance composition	
	4-3-5-7	4-3-1-8
U	68.5 ^a	81.6 ^b
Th	90.4 ^a	87.5 ^b
Al ₂ O ₃ beads	100.3	100.0
C	82.3	73.3 ^b
O ₂	112.6	98.9

^aBurnup was not taken into account.

^bBlender loss was not taken into account.

The effects of burnup can best be calculated using the ORIGEN code,³ and are presented in Tables 15 and 16. The relative amounts of uranium and thorium agreed well and were within 15% of the predicted amounts.

6.2 Distribution of Fission Products Throughout the Head-end Process

The distribution of fission products from FTE-4-3-5-7 to the various process steps is shown in Table 17. These results indicate that (1) most of the materials are found in the leach solution or residue, and (2) only very small amounts pass through the sintered-metal filter and are trapped on lines or absolute filters. Similar results were found for FTE-4-3-1-8. Any amounts of materials "plated-out" inside the burner were not determined.

Table 15. Uranium-thorium distributions for FTE-4-3-5-7 fuel:
 TRISO-coated (2.75 Th/U)C₂ -- TRISO-coated ThC₂

	General Atomic ¹		ORIGEN ²		Chemical analysis	
	(g)	(%)	(g)	(%)	(g)	(%)
Uranium	0.553	18.9	0.444	16.0	0.379	15.0
Thorium	2.378	81.1	2.336	84.0	2.149	85.0
Total	2.931	100.0	2.780	100.0	2.528	100.0

Table 16. Uranium-thorium distribution for FTE-4-3-1-8 fuel:
 TRISO-coated UO₂ -- BISO-coated ThO₂

	General Atomic ^a		ORIGEN ^b		Chemical analysis	
	(g)	(%)	(g)	(%)	(g)	(%)
Uranium	0.553	9.6	0.490	8.8	0.450	9.0
Thorium	5.184	90.4	5.094	91.2	4.534	91.0
Total	5.737	100.0	5.584	100.0	4.984	100.0

^aSee ref. 6.

^bORIGEN (564-day cooling), August 4, 1976 for 4-3-1-8 and August 5 for 4-3-5-7.

Table 17. Distribution of fission products to the major head-end steps for FTE-4-3-5-7 fuel: TRISO-coated (2.75 Th/U)C₂---
TRISO-coated ThC₂

Fission product	Grand total (100%)	Percentage found in			
		Leach ^a	Residue	Nickel filter	Off-gas equipment
⁹⁰ Sr	6.71E11	98.5	1.2	0.3	8.2E-05
⁹⁵ Zr	5.15E10	93.0	6.4	0.6	b
¹⁰⁶ Ru	5.97E10	64.5	35.2	0.3	3.5E-04
¹²⁵ Sb	1.19E10	92.7	6.2	1.1	6.2E-04
¹³⁴ Cs	2.10E11	97.9	0.9	1.2	2.1E-04
¹³⁷ Cs	4.92E11	98.0	1.1	0.9	1.3E-04
¹⁴⁴ Ce	3.52E12	99.7	0.3	0.1	<7E-05
¹⁵⁴ Eu	1.30E10	98.9	1.0	0.1	<2E-04

^aIncludes alumina-bead leach.

^bUndetected.

7. COMPARISON OF RESULTS FROM THE ORIGEN CODE, CHEMICAL ANALYSIS, AND NONDESTRUCTIVE GAMMA-SCANNING

The ORNL Isotope Generation and Depletion Code (ORIGEN)² is a computer program that can be used for predicting the quantities of activation products, actinides, and fission products from irradiation data using continuously updated nuclear data libraries. The ORIGEN calculation for FTE-4-3-5-7 and FTE-4-3-1-8 were made using irradiation parameters supplied by GAC.⁵ Details of the calculations for this run are given by W. Davis, Jr., et al.³

ORIGEN is used to provide estimates of final compositions after irradiation and aid in pinpointing (with NDA) discrepancies in the experimental results. Discrepancies can indicate losses of nuclides due to migration in the reactor, plateout on the walls of the experimental equipment, fuel handling losses in the hot cell, cross contamination of isotopes, faulty analytical results, and deficiencies in the computer code or input parameters.

The data, after extensive ORIGEN and chemical analysis (and reanalysis) is presented in Table 18 for FTE-4-3-5-7 and in Table 19 for FTE-4-3-1-8 and compared with the results of nondestructive analysis (NDA). Quantities of materials from ORIGEN, chemical analysis, and gamma-scanning are presented by isotope. (ORIGEN predicts many isotopes that are not listed in the table.)

With the exception of ^{234}U , the results of chemical analysis of heavy metals were within about $\pm 15\%$ of the ORIGEN predictions for both fuel rods. The results of chemical analysis (CA) for the other components, principally gamma-emitters, were not the same for both materials. Four isotopes (^{95}Nb , ^{106}Ru , ^{110}Ag , and ^{152}Eu) deviated widely from the ORIGEN (OC) predictions. Omitting these values, the range of ratios

Table 18. Comparison of ORIGEN with chemical analysis and gamma-scan analysis, FTE-4-3-5-7 fuel: TRISO-coated (2.75 Th/U)C₂--
TRISO-coated ThC₂

	ORIGEN ^a (OC)	Chemical analysis (CA)	CA/OC	Gamma scan ^b (NDA)	NDA/OC
²³³ U, g	0.034	0.032	0.94	-	-
²³⁴ U, g	0.008	0.004	0.50 ^c	-	-
²³⁵ U, g	0.340	0.284	0.84	-	-
²³⁶ U, g	0.037	0.031	0.84	-	-
²³⁸ U, g	<u>0.025</u>	<u>0.028</u>	<u>1.12</u>	-	-
Total U, g	0.444	0.379	0.85		
Av. ratio			0.94		
²³² Th, g	2.336	2.149	0.92	-	-
<u>Nuclide, dpm</u>					
⁸⁵ Kr	1.18E11	1.30E11	1.10	-	-
⁹⁰ Sr	1.00E12	6.71E11	0.67	-	-
⁹⁵ Zr	7.68E10	5.15E10	0.67	9.32E10	1.21
⁹⁵ Nb	1.65E11	<1.37E09	0.01 ^c	-	-
¹⁰⁶ Ru	3.93E11	5.97E10	0.15	4.57E11	1.16
¹¹⁰ Ag	6.96E09	1.33E08	0.02 ^c	-	-
¹²⁵ Sb	3.33E10	1.19E10	0.36	2.66E10	0.80
¹³⁴ Cs	5.82E11	2.10E11	0.36	4.66E11	0.80
¹³⁷ Cs	9.96E11	4.92E11	0.49	1.09E12	1.09
¹⁴⁴ Ce	4.39E12	3.52E12	0.80	4.57E12	1.04
¹⁵² Eu	9.83E07	<1.07E10	109. ^c	8.88E09	90.3 ^c
¹⁵⁴ Eu	3.10E10	1.30E10	0.42	3.11E10	1.00
¹⁵⁵ Eu	6.90E09	5.14E09	0.74	1.89E11	27.4 ^c
Av. ratio			0.62		1.01

^aORIGEN (564-day cooling), August 5, 1976.

^bV. C. A. Vaughen et al., Determination of Fission Product Inventories in FTE-4 Fuel Rods by Gamma Scanning (Scan No. 4, p. 14), GCR: 75-37 (December 1975).

^cOmitted from the average.

Table 19. Comparison of ORIGEN with chemical analysis and gamma-scan analysis of FTE-4-3-1-8 fuel: TRISO-coated UO₂--
BISO-coated ThO₂

	ORIGEN ^a (OC)	Chemical analysis (CA)	CA/OC	Gamma scan ^b (NDA)	NDA/OC
²³³ U, g	0.072	0.057	0.79	-	-
²³⁴ U, g	0.010	0.007	0.70	-	-
²³⁵ U, g	0.348	0.326	0.94	-	-
²³⁶ U, g	0.035	0.033	0.94	-	-
²³⁸ U, g	<u>0.025</u>	<u>0.027</u>	<u>1.08</u>	-	-
Total U, g	0.490	0.450	0.92	-	-
			0.89		
Av. ratio					
²³² Th, g	5.094	4.534	0.89	-	-
<u>Nuclide, dpm</u>					
⁸⁵ Kr	1.26E11	3.68E10	0.29	-	-
⁹⁰ Sr	1.01E12	7.78E11	0.77	-	-
⁹⁵ Zr	8.10E10	<6.88E10	0.85	4.88E10	0.60
⁹⁵ Nb	1.74E11	<2.01E09	0.01 ^c	-	-
¹⁰⁶ Ru	3.91E11	8.12E11	2.08 ^c	1.80E11	0.46
¹¹⁰ Ag	6.62E09	<4.40E08	0.07 ^c	-	-
¹²⁵ Sb	3.66E10	<1.67E10	0.46	2.66E10	0.73
¹³⁴ Cs	5.43E11	2.61E11	0.48	1.67E11	0.31
¹³⁷ Cs	1.00E12	6.61E11	0.66	5.00E11	0.50
¹⁴⁴ Ce	4.47E12	3.63E12	0.81	1.81E12	0.40
¹⁵² Eu	1.03E08	<7.92E09	76.9 ^c	6.66E09	64.6 ^c
¹⁵⁴ Eu	2.84E10	<1.37E10	0.48	2.66E10	0.94
¹⁵⁵ Eu	6.67E09	<5.74E09	0.86	1.73E11	25.9 ^c
Av. ratio			0.67		0.56

^aORIGEN (564-day cooling), August 4, 1976.

^bV. C. A. Vaughen et al., Determination of Fission Product Inventories in FTE-4 Fuel Rods by Gamma Scanning, Scan No. 4, p. 14, GCR: 75-37 (December 1975).

^cOmitted from the average.

CA/OC was ~36 to 109% for FTE-4-3-5-7 and about 29 to 89% for FTE-4-3-1-8.

In like manner, comparing the results of nondestructive gamma-scan analysis (NDA)⁴ to ORIGEN (OC), the range of the ratios NDA/OC was ~ 80 to 121% for FTE-4-3-5-7 and 31 to 94% for FTE-4-3-1-8.

8. DISCUSSION

The main purpose of these experiments was to select a suitable means for establishing a credible total inventory of materials in the incoming fuel for use in material balances and as an indirect measure of plateout losses to surfaces of the equipment in future experiments.

We have shown that ORIGEN results for HTGR fuels have been improved by feedback from this work.^{3,4} Adding better estimates of reactor operating times (and decay periods), as well as focusing on the influence of the ORIGEN inputs, has resulted in calculations agreeing between the two rods within +10% (see Tale 20, ORIGEN column.) The two fuel rods were only separated by inches in the reactor. Further work is indicated³ (1) to obtain more detailed reactor flux-time histories, and (2) to improve our understanding of the effects of resonance (epi-thermal) neutron flux as used by ORIGEN in estimating fission rates of ²³³U and ²³⁵U.

Gamma-scanning has great potential in allowing an independent estimate of certain isotopes, and assists in checking and "tuning" ORIGEN so that all isotopes may be estimated more precisely. The NDA results for FTE-4-3-5-7 were within +20% of the final ORIGEN predictions. Unfortunately, this agreement was not sustained with FTE-4-3-1-8 (Table 20, NDA column). It appears that the inhomogeneity⁴ of the broken

Table 20. Comparison of estimates of inventories of FTE-4-3-1-8 with FTE-4-3-5-7 by several methods (numbers are the ratio 4-3-1-8/4-3-5-7 for the listed estimate)

	ORIGEN ^a calculations	NDA ^b results	Chemical analysis (CA)
²³³ U	2.1 ^c	-	1.78 ^c
²³⁴ U	1.25	-	1.75 ^c
²³⁵ U	1.02	-	1.15
²³⁶ U	0.95	-	1.06
²³⁸ U	<u>1.00</u>	-	<u>0.96</u>
Total U	1.10	-	1.19
Av. ratio	1.06	-	1.09
²³² Th	2.18	-	2.11
⁸⁵ Kr	1.07	-	1.00
⁹⁰ Sr	1.01	-	1.16
⁹⁵ Zr	1.05	0.52	~1.3
⁹⁵ Nb	1.05	-	~1.5
¹⁰⁶ Ru	0.99	0.39	13.6 ^c
¹¹⁰ Ag	0.94	-	3.31 ^c
¹²⁵ Sb	1.10	1.00	~1.4
¹³⁴ Cs	0.93	0.36	1.25
¹³⁷ Cs	1.00	0.46	1.34
¹⁴⁴ Ce	1.02	0.40	1.03
¹⁵² Eu	1.05	0.75	~0.74 ^c
¹⁵⁴ Eu	0.92	0.86	~1.0
¹⁵⁵ Eu	<u>0.97</u>	<u>0.92</u>	<u>~1.1</u>
Av. ratio	1.01	0.63	1.21

^aORIGEN (564-day cooling), August 5, 1976 for 4-3-5-7 and August 4 for 4-3-1-8.

^bV. C. A. Vaughen et al., Determination of Fission Product Inventories in FTE-4 Fuel Rods by Gamma Scanning (Scan No. 4, p. 14), GCR: 75-37 (December 1975).

^cOmitted from the average.

^dDue to larger amount of thorium.

fuel rod was not adequately accounted for by the methods used.

Therefore, to establish the usefulness of gamma-scanning, one should develop better methods to correct for fuel rod breakage and other inhomogeneities and repeat the comparisons using other fuels.

As mentioned above (Tables 18 and 19), the results of the chemical analysis indicate that the inventories derived by summing the results from many liquid and solid samples give apparent yield ratios compared with ORIGEN ranging from about 36 to 80% (excluding ^{85}Kr , ^{95}Nb , ^{106}Ru , ^{110}Ag , and ^{152}Eu). However, when the results of chemical analyses for the two runs are compared with each other, the range of ratios of the inventories (4-3-1-8 to 4-3-5-7) was only about 1.25 ± 0.25 , excluding ^{106}Ru , ^{110}Ag , and ^{152}Eu (Table 20, CA column). These data indicate a type of consistency, even though they are not in agreement with ORIGEN. If additional fuel is processed in this manner, the significance of the radiochemical results may become more obviously "plateout" losses. However, as mentioned above, the results in Tables 18 and 19 are the products of many recheck analyses. A number of values remain faulty or indeterminate (<x) because of decay, low concentrations, gamma peak overlap, handling losses or cross-contamination. Thus at this stage, conclusions cannot be drawn about plateout or loss by other mechanisms. Composites of the results for FTE-4-3-5-7 and FTE-4-3-1-8 are included in the Appendix.

9. CONCLUSIONS

With more detailed reactor operating parameters, better calculation of the ORIGEN input parameters and better gamma-scan (NDA) interpretation, it should be possible to routinely obtain close agreement between ORIGEN

predictions and actual inventories determined by gamma-scanning. The chemical analysis for uranium isotopic composition already agrees within about 15% of the ORIGEN values. For other isotopes, the chemical analyses in general gave low values ($\sim 50\% \pm 20\%$) relative to ORIGEN results. This low yield has brought attention to several problems in the separation and analysis of materials having high radiation levels.

10. FUTURE WORK

The continued improvements in hot-cell techniques and the extensive use of experimental "run sheets" has allowed a sharper focus on those in-cell operations that still need improvement. These areas lie within the hot-cell handling and the analytical chemistry techniques. The hot-cell problem areas are the following: (1) interpretation of the gamma scans of incoming fuel, (2) powder handling and solution sampling techniques, (3) collection of particulates from off-gases, and (4) direct measurement of fission-product buildup in the hot-cell equipment.

Several analytical areas need improvement: (1) preparing storable aliquots of sample solutions, (2) reducing the delay between sample submission and analysis, (3) increasing the capability for analyzing minor components, and (4) improving the capability to analyze undissolved solids and precipitates. Some isotopes, especially ^{106}Ru , ^{110}Ag , and ^{234}U , appear to give continued problems in analysis.

11. ACKNOWLEDGMENT

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13. APPENDIX: COMPOSITE RESULTS FOR FTE-4-3-5-7 AND FTE-4-3-1-8

A-1. Results for FTE-4-3-5-7 fuel: TRISO-coated (2.75 Th/U)C₂--TRISO-coated ThC₂

Nuclide, dpm	Leach	Insolubles		Total insolubles	Total leach + insolubles	% insolubles in burner ash	Alumina (beads) leach	Sintered-nickel filter	Combined off-gases	Chemical analysis total (CA)	Gamma scan (NDA)	ORIGEN ^b (OC)	NDA/OC	CA/OC	CA/NDA
		First fusion	Second fusion												
U, g	0.361	0.008	0.010	0.018	0.379	4.7	Trace	<0.001	-	0.379	-	0.444		0.85	
Th, g	2.100	0.006	0.041	0.047	2.147	2.2	Trace	0.002	-	2.149	-	2.336		0.92	
⁹⁹ Tc, mg	1.338	0.095	0.019	0.114	1.452	7.8	0.001	c	<0.002	1.455	-	3.722		0.39	
¹²⁹ I, mg	0.010	<0.001	<0.001	<0.001	0.010	~10	c	c	-	0.010	-	0.598		0.02	
⁸⁵ Kr	6.13E08	-	-	-	6.13E08		-	-	<1.30E11	1.30E11		1.18E11		1.10	
⁹⁰ Sr	6.60E11	7.50E08	7.62E09	8.37E09	6.68E11	1.3	7.29E08	<1.94E09	<5.49E05	6.71E11	c	1.00E12		0.67	
⁹⁵ Zr	4.75E10	2.79E09	4.94E08	3.28E09	5.08E10	6.5	<3.30E08	<3.24E08	c	5.15E10	9.32E10	7.68E10	1.21	0.67	0.55
⁹⁵ Nb	1.28E09	<5.52E06	<4.50E07	<5.05E09	<1.33E09	3.8	<2.70E07	<1.80E08	c	<1.37E09	c	1.65E11		0.01	
¹⁰⁶ Ru	3.79E10	5.15E07	2.09E10	2.10E10	5.89E10	35.7	<6.00E08	<1.80E08	2.07E05	5.97E10	4.57E11	3.93E11	1.16	0.15	0.13
¹¹⁰ Ag	3.81E07	1.73E07	2.20E06	1.95E07	5.76E07	33.9	<7.50E07	c	c	1.33E08	c	6.96E09		0.0	
¹²⁵ Sb	1.07E10	1.50E07	7.26E08	7.41E08	1.14E10	6.5	<3.30E08	<1.32E08	7.40E04	1.19E10	2.66E10	3.33E10	0.80	0.36	0.45
¹³⁴ Cs	1.98E11	5.80E06	1.83E09	1.84E09	2.00E11	0.9	7.62E09	2.41E09	4.43E05	2.10E11	4.66E11	5.82E11	0.80	0.36	0.45
¹³⁷ Cs	4.65E11	1.86E07	5.53E09	5.55E09	4.71E11	1.2	1.69E10	4.57E09	6.16E05	4.92E11	1.09E12	9.96E11	1.09	0.49	0.45
¹⁴⁴ Ce	3.50E12	1.02E10	1.07E09	1.13E10	3.51E12	0.3	3.81E09	1.82E09	<2.37E06	3.52E12	4.57E12	4.39E12	1.04	0.80	0.77
¹⁵² Eu	<1.05E10	<1.08E07	<8.40E06	<1.92E07	<1.05E10	0.2	<1.23E08	<4.56E07	c	<1.07E10	8.88E09	9.83E07	90.3	109	1.20
¹⁵⁴ Eu	1.28E10	5.13E07	8.52E07	1.37E08	1.29E10	1.1	6.90E07	<1.56E07	<3.25E04	1.30E10	3.11E10	3.10E10	1.00	0.42	0.42
¹⁵⁵ Eu	3.85E09	2.26E07	2.99E07	5.25E07	3.90E09	1.3	<1.17E09	6.60E07	c	5.14E09	1.89E11	6.90E09	27.4	0.74	0.03

^aV. C. A. Vaughen et al., Determination of Fission Product Inventories in FTE-4 Fuel Rods by Gamma-Scanning (Scan No. 4, p. 14), GCR: 75-37 (December 1975).

^bORIGEN (564-day cooling), August 5, 1976.

^cUndetected.

Table A-2. Composite of results for FTE-4-3-1-8 fuel: TRISO-coated UO₂--BISO-coated ThO₂

	Leach	R-1-A	R-1	R-1 NFR	Total insolubles	Total leach + insolubles	% insolubles in burner ash	Aluminum bead leach	Sintered nickel filter	Off-gas line	AF-1	AF-2	AF-3	Combined off-gases	Chemical analysis total (CA)	Gamma scan ^a (NDA)	ORIGEN ^b (OC)
U, g	0.445	0.004	0.001		0.005	0.450	1.2	0.001	<0.001	-	-	-	-	-	0.451 g	-	0.490
Th, g	4.495	0.017	0.002		0.019	4.514	0.4	0.015	0.005	-	-	-	-	-	4.534 g	-	5.094
⁹⁹ Tc, mg	1.15	0.042	0.452	c	0.494	1.644	30.0	c							1.644 mg	-	3.723 mg
¹²⁹ I, mg	0.048	<0.001	c	c	<0.001	0.048	<2.1	-	-	-	-	-	-	-	0.048 mg	-	0.654 mg
Nuclide, dpm																	
⁸⁵ Kr	4.68E09	-	-	-	-	4.68E09		-	-	-	-	-	-	3.68E10	3.68E10		1.26E11
⁹⁰ Sr	7.14E11	3.18E10	1.10E08	c	3.19E10	7.46E11	4.3	4.98E09	2.74E10	<1.84E06	c	c	c	<1.84E06	7.78E11	c	1.01E12
⁹⁵ Zr	5.53E10	9.78E10	2.46E09	c	3.44E09	5.87E10	5.9	<9.90E09	<1.80E08	<7.80E04	<2.85E04	<2.16E04	<2.88E04	<1.57E05	<6.88E10	4.88E10	8.10E10
⁹⁵ Nb	<1.38E09	<8.40E07	<2.65E08	2.61E08	<6.10E08	<1.99E09	30.6	<1.92E07	<4.86E06	<1.50E03	<6.18E02	<6.24E02	<6.24E02	<3.37E03	<2.01E09	c	1.74E11
¹⁰⁶ Ru	1.62E10	6.72E11	1.21E11	7.12E08	7.94E11	8.10E11	98.0	<2.28E08	1.85E09	<4.61E04	3.50E04	<9.60E03	<5.58E03	<9.63E04	8.12E11	1.80E11	3.91E11
¹¹⁰ Ag	1.62E08	4.12E06	1.35E06	c	<5.47E06	<1.67E08	3.3	<2.73E08	c	c	c	c	c	c	<4.40E08	c	6.62E09
¹²⁵ Sb	7.68E09	2.59E09	<4.75E09	3.25E08	<7.67E09	<1.53E10	50.1	<1.32E09	<1.02E08	<2.04E04	<2.88E03	<2.64E03	<2.16E03	<2.81E04	<1.67E10	2.66E10	3.66E10
¹³⁴ Cs	1.94E11	3.52E09	3.41E10	c	3.76E10	2.32E11	16.2	2.85E10	1.01E09	<2.42E04	<1.62E03	4.32E03	<9.00E02	<3.10E04	2.61E11	1.67E11	5.43E11
¹³⁷ Cs	5.05E11	9.00E09	7.88E10	c	8.78E10	5.93E11	14.8	6.60E10	2.29E09	<5.09E04	4.72E03	7.74E03	<1.20E03	<6.46E04	6.61E11	5.00E11	1.00E12
¹⁴⁴ Ce	3.35E12	3.54E10	3.03E10	c	6.57E10	3.42E12	1.9	2.16E11	2.20E09	<8.41E04	1.47E04	<1.02E04	<1.02E04	<1.19E05	3.63E12	1.81E12	4.47E12
¹⁵² Eu	<6.00E09	<1.14E08	<9.15E08	c	<1.03E09	<7.03E09	14.6	<8.70E08	<1.98E07	<4.80E03	<1.74E03	<1.68E03	<3.78E03	<1.20E04	<7.92E09	6.66E09	1.03E08
¹⁵⁴ Eu	1.29E10	8.34E07	8.84E07	c	1.72E08	1.31E10	1.3	<6.00E08	<1.02E07	<5.04E03	<1.44E03	<1.44E03	<1.38E03	<9.30E03	<1.37E10	2.66E10	2.84E10
¹⁵⁵ Eu	4.15E09	4.97E07	3.54E07	c	8.51E07	4.24E09	2.0	<1.47E09	<3.36E07	<1.69E04	<2.58E03	<1.80E03	<3.72E03	<2.50E04	<5.74E09	1.73E11	6.67E09

^aV. C. A. Vaughn et al., Determination of Fission Product Inventories in FTE-4 Fuel Rods by Gamma Scanning

(Scan No. 4, p. 14), GCR: 75-37 (December 1975).

^bORIGEN (564-day cooling), August 4, 1976.

^cUndetected.

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