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**SCOPING STUDY OF FLOWPATH OF SIMULATED FISSION
PRODUCTS DURING SECONDARY BURNING OF CRUSHED
HTGR FUEL IN A QUARTZ FLUIDIZED-BED BURNER**

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SCOPING STUDY OF FLOWPATH OF SIMULATED FISSION PRODUCTS
DURING SECONDARY BURNING OF CRUSHED HTGR FUEL
IN A QUARTZ FLUIDIZED-BED BURNER

by

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ABSTRACT

The results of four experimental runs in which isotopic tracers were used to simulate fission products during fluidized bed secondary burning of HTGR fuel were studied. The experimental tests provided insight relative to the flow path of fission products during fluidized-bed burning of HTGR fuel.

SUMMARY

Four experimental scoping tests were completed in which crushed fertile particles containing radioactive tracers were burned in a 2.3-inch diameter quartz fluidized-bed burner. The purpose of the tests was to provide insight regarding the flowpath (distribution in the burner and off-gas systems) of potentially volatile fission products, particularly Cs, Rb, Ru, and Se, during secondary burning of HTGR fuel.

The test results showed that an in-vessel, sintered-metal filter having a mean pore size of 5 microns removed > 99% of simulated fission products tested, but the burner filter may plug with Ru. Se-75 was the most plentiful isotope found in the off-gas system; Cs-134 was the second most plentiful isotope in the off-gas; no Rb was detected in the off-gas. The amount of radioactive tracers in the various off-gas equipment sections increased with increasing fluidizing velocity. No correlation between tracer activity and temperature of off-gas equipment components was apparent.

The tests supplied insight for designing future tests using irradiated samples.

TABLE OF CONTENTS

ABSTRACT	1
SUMMARY	11
I. INTRODUCTION	1
II. DESCRIPTION OF EQUIPMENT	2
III. EXPERIMENTAL PROCEDURE	4
IV. RESULTS AND DISCUSSION	5
V. ACTIVITY DISTRIBUTION.	26
VI. CONCLUSIONS AND RECOMMENDATIONS.	29

FIGURES

1. Schematic Diagram of Equipment Used to Study Flow Path of Volatile Fission Products,	3
2. Temperature Profile of Each Equipment Component for Run 1.	9
3. Gamma Activity Profile in Off-Gas System for Run 1	12
4. Temperature Profile of Each Equipment Component for Run 2.	13
5. Gamma Activity Profile in Off-Gas System for Run 2	16
6. Temperature Profile of Each Equipment Component for Run 3.	17
7. Gamma Activity Profile in Off-Gas System for Run 3	20
8. Temperature Profile of Each Equipment Components for Run 4	23
9. Gamma Activity Profile in Off-Gas System for Run 4	24
10. Decontamination Factor as a Function of Fluidizing Velocity for In-Vessel Filter	28

TABLES

I. Summary of Operating Conditions and Run Results.	6
II. Summary of Activity and Carrier Distribution for Each Run.	7
III. Gamma Activity in Equipment Components for Run 1	11
IV. Gamma Activity in Equipment Components for Run 2	15
V. Gamma Activity in Equipment Components for Run 3	19
VI. In-Vessel Filter Gamma Activity Before and After Decontamination in Boiling Acid-Thorex Solution	21
VII. Gamma Activity of Equipment Components for Run 4	25
VIII. Distribution of Tracers Throughout System.	27

SCOPING STUDY OF THE FLOWPATH OF SIMULATED FISSION PRODUCTS DURING SECONDARY BURNING OF CRUSHED HTGR FUEL

I. INTRODUCTION

During head-end reprocessing of Ft. St. Vrain fuel, whole fuel elements are crushed to less than 3/16-inch particle size. The matrix graphite is burned in a primary fluidized-bed burner to expose silicon carbide coatings on fissile and fertile particles. The silicon carbide coated particles are separated by gas classification and are broken to expose uranium carbide, thorium carbide, and pyrolytic carbon. The exposed carbon and carbides are burned in a secondary fluidized-bed burner. The resulting secondary burner ash is leached to recover uranium and thorium; the leachate is processed in a solvent extraction system.

It is believed that some volatile fission products will vaporize during the secondary burning step which takes place at 900°C. The possible consequences of volatile fission products are: 1) plugging of in-vessel sintered metal filters, 2) plugging of off-gas lines downstream from the burner, 3) high radiation levels in off-gas lines, complicating maintenance, and 4) interference with other cleanup equipment located downstream such as the catalytic converter or KALC process.

This report describes the results of four experiments in which isotopic tracers were adsorbed onto a bed of crushed HTGR fertile particles and the mixture was burned in a batch secondary fluidized-bed burner at operating conditions recommended for the HTGR secondary burner. The principle objective of the tests was to determine the volatility and deposition locations of actual fission products anticipated in the burner and off-gas system. Only those fission products with oxide boiling points less than 1000°C were studied. The tracers selected for testing were Ru-103, Cs-134, Rb-86, and Se-75. These particular tracers were chosen because of their short half-life.

II. DESCRIPTION OF EQUIPMENT

The secondary burner and off-gas components used for the experiments are shown in Figure 1. The burner system was composed of a burner vessel, an in-bed sintered metal filter, and a heating system consisting of beaded heaters controlled by a variable electric source. The burner was constructed of 2.3-inch I.D. quartz pipe, 26 inches long, having a distributor plate which contained nineteen holes, 0.025 inches in diameter. A 6-inch-long by 1.5-inch-diameter, 5-micron-mean-pore-size sintered metal filter (Metal Filter #1) constructed of 304L stainless steel was located in the upper section of the burner. The filter surface area was 0.20 sq. ft. The filter was used to remove particulates from the off-gas system. A manual blowback system allowed blowback of fines from the filter. Four thermocouples were located in the burner; one in the bottom of the bed, one in the middle of the bed, one an inch below the filter, and one inside the filter approximately one inch from the lower end of the filter.

The off-gas system was composed of five sections of glass located between the burner vessel and a second sintered metal filter chamber; the second filter (Filter 2) was 3 inches long, 1 inch in diameter, and 5-micron in mean pore size. The first four sections of glass each contained a thermocouple to allow measurement of the gas temperature at the particular location and to evaluate plating of the fission products as a function of temperature. Immediately following the metal-filter chamber were two additional sections of glass: the first contained a thermocouple and the second was followed by a filter chamber. The filter chamber contained a fiber filter 1 mm thick and 5 inches in diameter. The fiber filter was used to represent a HEPA filter; it was tested to be capable of a minimum of 99.7% efficiency for particles larger than 0.3 microns and greater than 98% efficient for particles as small as 0.05 microns. Directly following the fiber filter were three sections of glass: the first contained a thermocouple, the second was an outlet tee to the CO, CO₂, and O₂ analyzers, and the third was an adaptor for the gas sample bomb.

Figure 1. Schematic Diagram of Equipment used to Studying Flowpath of Volatile Fission Products (Locations and nomenclature of thermocouples and equipment sections are shown)

The gas sample bomb was 6.5 inches long and 2 inches in diameter, having stopcocks at both ends. Following the glass sample bomb was another section of glass containing a thermocouple and a glass section. The glass section was connected to a tube that penetrated a three inch, flow-through NaI detector. The NaI detector was connected to a single channel analyzer and the gamma count-rate meter to monitor activity in the off-gas. The results were recorded on a strip chart recorder. (The NaI detector was used only in Runs 3 and 4). The off-gas was exhausted to a laboratory hood.

III. EXPERIMENTAL PROCEDURE

The experimental procedure and materials used for the tests were similar to those anticipated for plant burner operations. The bed was composed of fertile particles crushed by a roll crusher at a gap setting of 0.012 inches. The tracers, Cs-134, Se-75, and Rb-86 in 0.5 M HCl and Ru-103 in 0.5 HNO₃ were pipetted onto carbon that contained 5% nonradioactive Ru and dried. (The Ru carbon mixture was used for the Ru carrier). The very low concentration of nitrate and chloride anions should have virtually no affect on the overall chemical reactions taking place in the bed. The fertile particle bed and the ruthenium-carbon-tracer mixture were place in the burner vessel and intimately mixed by fluidization with nitrogen. After mixing, a bed sample was taken for analysis. Other carriers* consisting of non-radioactive CsC₈, RbC₈, and TeO₂ (carrier for Se-75) were added to each batch. The carbides were weighed and handled in an inert atmosphere of nitrogen because of their pyrophoric nature in air. For this reason they were also added to the burner which contained a nitrogen environment. After all materials and the apparatus were in place, the heating system for the burner was activated and the fluidizing-gas flow initiated. The fluidizing gas was air during start-up; nitrogen, oxygen or a mixture of N₂ and O₂ was subsequently required to obtain the desired temperature. The temperature at the bottom of the bed was allowed to increase to 900°C before the beaded heaters were deactivated. CO, CO₂, and O₂ concentrations

* The carriers are used to help increase the concentration of the element to allow driving more of the radioactive tracer into the gaseous stream so that it could be detected.

in the off gas were monitored during the entire run. At different time intervals during the run, the temperatures of the eleven thermocouples were recorded.

The pressures in the off-gas system, including the in-vessel filter, were monitored. When the pressure reached 30 inches of water, the filter was "blown back" to clear the filter of fines.

Near the end of a run, when the oxygen concentration in the off-gas began to increase, the fluidizing velocity was decreased and external heat applied to the burner. The temperature was maintained at a constant level until no CO or CO₂ was detected in the off-gas; the system was then shut down. After the run was completed and the system cooled, a bed sample was taken for analysis and the rest of the system disassembled for individual component analysis by gamma scanning techniques.

During the burner operation, the amount of activity in the final off gas was monitored by the flow-through NaI detector placed at the end of the off-gas train.

IV. RESULTS AND DISCUSSION

Four experimental runs were made in which the principal operating variables were the fluidizing velocity, bed size, and amount of tracer. Fluidizing velocity and bed size especially affect in-vessel filter temperature and off-gas equipment component temperatures. The various operating conditions and results for the four runs are shown in Table I. The amount of tracer added to the bed is shown in Table II. The duration of each run was one to three hours.

TABLE I
SUMMARY OF OPERATING CONDITIONS AND RUN RESULTS

<u>Run</u>	<u>Run-1</u>	<u>Run-2</u>	<u>Run-3</u>	<u>Run-4</u>
Initial(a) Bed wt,g	1362	720	775	865
Burned Bed wt.,g	1133	633	659	707
Ave. Bed Temp, °C	766	871	878	888
Fluidizing Velocity, ft/sec	1.1	0.8	1.5	1.2
Ave. In-Bed Filter Temp, °C	429	448	514	492
Burn Rate(b) kgC/hr-ft ²	9.3	7.4	11.3	8.1
Ave. CO ₂ In Off-Gas, %	74	76	65	59
Fluidizing Gas Mixture, N ₂ :O ₂	1:3	1:3	2:3.5	2:3
Run Duration(c), min.	104	132	74	98

(a) Crushed in roll crusher at gap setting of 0.012 inch.

(b) Based on % CO₂ in off-gas.

(c) From the time heat was applied until CO₂ was less than 1%.

TABLE II

SUMMARY OF ACTIVITY AND CARRIER DISTRIBUTION FOR EACH RUN

	<u>Run-1</u>	<u>Run-2</u>	<u>Run-3</u>	<u>Run-4</u>
<u>Starting Bed</u>				
Cs-134, μci	156	10730	1135	23500
%	57.7	89.7	36.6	83.9
Ru-103, μci	51	1460	1432	1000
%	18.9	7.7	46.2	3.6
Se-75, μci	59	86	351	3243
%	21.8	0.4	11.3	11.6
Rb-86, μci	0	0	178	459
%	0	0	5.7	1.6
Total, μci	2.7×10^2	1.9×10^4	3.1×10^3	2.8×10^4
<u>Carrier in Bed</u>				
CsC ₈ , g	14	8	8	10
5% Ru on Carbon, g	45	30	30	30
TeO ₂ , g	4	2	2	2
RbC ₈ , g	0	0	2	2

Run 1

The bed for Run 1 was composed of 1362 grams of fertile particles crushed in a roll crusher at a gap setting of 0.012 inch. The amount of tracers added to the bed were: 156 microcuries of Cs-135, 51 microcuries of Ru-103, and 59 microcuries of Se-75. All tracers were in the liquid chloride form. No Rb was used in this run.

The tracers were pipetted and dried on 45 grams of carbon containing 5% ruthenium. After mixing the ruthenium bearing carbon with the bed material by fluidization in the burner with nitrogen, 14 grams of CsC_8 and 4g TeO_2 were added to the burner. The amount of carrier used corresponded to the quantity calculated for each individual isotope that would be expected to be in an equivalent amount of FSVR irradiated fertile particles that had been in the reactor for 6 years and cooled 6 months before processing, as shown in Table II.

The operating parameters for Run 1 are given in Table I. The average bed temperature was 766°C with a 200°C spread between the bottom and top thermocouple in the bed as shown in Figure 2. (Curve numbers shown in Figure 2 correspond to equipment component numbers shown in Figure 1.) The fluidizing velocity, 1.1 ft/sec, was probably too low because the top of the bed cooled much more rapidly than the bottom. A higher fluidizing velocity could not be obtained using the larger bed because of in-vessel filter plugging even with filter blowback. The average filter temperature was approximately 429°C for the main combustion period, which lasted for about 45 minutes. The off-gas was cooled to 30°C before it exited the system.

The CO_2 concentration in the off-gas averaged 74%; CO and O_2 concentrations were zero during the main combustion period. A nitrogen to oxygen fluidizing gas ratio of 1:3 was used.

Figure 2. Temperature Profile of Each Equipment Components for Run-1.
(Curve numbers and thermocouple--T.C.--numbers correspond to equipment component nomenclature given in Figure 1.)

After burning was complete and the system was cooled, the equipment was disassembled for gamma analysis. Because of the small amount of tracer activity in the off-gas equipment components, only gross gamma readings were taken for each equipment component in the system downstream from the in-vessel filter. The gamma activity for each off-gas equipment component is given in Table III and shown in Figure 3. The tracer activity of all equipment components appears to be uniform except for that of the second sintered metal filter, which has approximately four times the activity per cm^2 of external surface as the other off-gas components. This indicates the filter may be selectively collecting material that has been released by the burner. Figure 3 also shows a decrease in tracer activity for the individual component parts that are located downstream from the filter. The temperature of the second metal filter was between 60 and 70°C during the main combustion period. The decontamination factor across the in-vessel filter was 2.8×10^4 based on the quantity of activity charged to the burner and the amount found downstream of the burner. The burnrate was 9.3 kgC/hr-ft² for the run, based on CO₂ concentration in the off-gas.

Run 2

The bed for Run 2 was 720 grams of crushed fertile particles. The operating parameters are shown in Table I. The amount of tracers and carriers used are shown in Table II. A larger quantity of tracers was used in this run because of the small amount of activity found in the off-gas system after Run 1. The tracers were pipetted onto 30g of carbon containing 5% ruthenium, dried, and mixed with the bed. The average temperature during the main combustion period was 871°C; good agreement was maintained between the bottom and top bed thermocouples.

The temperature profile of each system component for this run is shown in Figure 4. The off-gas was cooled to 32°C before leaving the system. The average temperature of the in-vessel filter was 448°C.

TABLE III

GAMMA ACTIVITY IN EQUIPMENT COMPONENTS FOR RUN 1
 (Equipment Location and Component Nomenclature Corresponds to that Given in Figure 1)

Position	Gross Gamma, cts/sec	Average Temp., °C	Area of Com- ponent, cm ²	Gross gamma, cts/sec-cm ²
Bed (Final)	8.7×10^7	766	-	-
Filter-1	4.5×10^4	429	28.3	1.6×10^3
Glass-1	2.4×10^4	-	-	-
Glass-2	8.8×10^3	-	176.0	420.0
Glass-3	91	122	16.7	5.4
Glass-4	86	111	13.4	6.4
Glass-5	92	96	16.9	5.4
Glass-6	55	83	13.1	4.2
Glass-7	29	64	16.3	1.8
Glass-8	918	64	431.0	2.1
Filter-2	209	64	9.4	22.0
Glass-9	117	46	15.3	7.6
Glass-10	111	39	17.3	6.4
Fiber Filter	206	39	95.0	2.1
Glass-11	552	39	134.0	4.1
Glass-12	59	33	15.8	3.7
Glass-13	0	32	15.2	0.0
Glass-14	59	32	13.4	4.4
Glass-15	495	32	244.0	2.0
Glass-16	65	30	15.8	4.1
Total	8.7×10^7

Figure 3. Gamma Activity Profile in Off-Gas System from Run 1

Figure 4. Temperature Profile of Each Equipment Component for Run-2
(Curve and thermocouple--T.C.--correspond to equipment component nomenclature given in Figure 1.)

The fluidizing velocity was 0.8 ft/sec; an average N_2/O_2 fluidizing gas ratio of 1:3 was used. The average CO_2 in the off-gas was 76%; the CO and O_2 concentrations were essentially zero during the main combustion period.

When the run was complete and the system cooled, each component was gamma scanned. Because of the small amount of gamma activity found in the off-gas system, only gross gamma readings were taken of the off-gas equipment components downstream of the in-vessel filter. The tracer gamma activity for each off-gas component is given in Table IV and Figure 5. There was a sudden increase in activity on the second sintered metal filter and a general decrease in activity in the components downstream from the filter. The temperature of this filter was between 70 and 80°C for the main combustion period. The decontamination factor across the in-vessel filter was 1.1×10^6 . The burnrate was 7.4 kgC/hr-ft².

Run 3

A starting bed of 775 g of crushed fertile particles was used in Run-3. The operating parameters are shown in Table I. The bed was mixed with 30g of carbon containing 5% ruthenium and the amount of dried tracers and carriers shown in Table II. Rb-86 was used in this run.

The average temperature during the main combustion period was 878°C, and agreement between bed thermocouples was good. The temperature profile for the system components is shown in Figure 6. The off-gas was cooled to 34°C before exiting the system. The average in-vessel filter temperature was 514°C. This temperature is higher than that of the other runs because of a higher fluidizing velocity, 1.5 ft/sec.

The average N_2/O_2 fluidizing gas ratio was 2:3.5. The average CO_2 concentration in the off gas was 65%; the O_2 and CO concentrations were zero during the main combustion period.

TABLE IV

GAMMA ACTIVITY OF EQUIPMENT COMPONENTS FOR RUN 2

(Equipment Location and Component Nomenclature Corresponds to that Given in Figure 1)

Position	Gross Gamma, cts/sec	Gross Gamma, cts/sec-cm ² (a)	Ave. Temp., °C(b)
Bed (Final)	2.0×10^8	-	871
Filter-1	2.7×10^5	9.6×10^3	448
Glass-1	3.8×10^4	-	-
Glass-2	2.8×10^4	160.0	-
Glass-3	7.6	0.46	135
Glass-4	6.9	0.51	129
Glass-5	8.7	0.51	110
Glass-6	8.2	0.63	95
Glass-7	15.9	0.98	72
Glass-8	27.0	0.12	72
Filter-2	12.8	1.36	72
Glass-9	5.6	0.37	49
Glass-10	4.1	0.24	42
Fiber Filter	40.0	0.41	42
Glass-11	16.5	0.12	42
Glass-12	3.2	0.20	34
Glass-13	3.2	0.21	32
Glass-14	3.7	0.28	32
Glass-15	16.0	0.22	32
Glass-16	2.6	0.16	32
Total	2.0×10^8		

(a) Area of each component same as in Table III

(b) Excluding first reading.

Figure 5. Gamma Activity Profile in Off-Gas System for Run-2
(Equipment nomenclature corresponds to that given
in Figure 1.)

Figure 6. Temperature Profile of Each Equipment Component for Run 3
(Curve and thermocouple--T.C.--numbers correspond to
equipment component nomenclature given in Figure 1.)

After the run was complete and the system cooled each equipment component was analyzed for gamma activity. The gamma activity results are tabulated in Table V and shown graphically in Figure 7. Figure 7 shows the approximately 60% of the gamma activity found upstream of the second sintered metal filter was Se-75. Downstream of the second filter the activity is evenly distributed between Ru-103 and Cs-134 on the filter. Downstream from the fiber filter the activity generally decreased. The fiber filter apparently did not collect appreciable quantities of tracers relative to the other components downstream from the second metal filter. This indicated that the tracers passing through the filters were in the gaseous or aerosol state. The temperature of the second filter was between 100 and 110°C during the main combustion period.

The DF across the in-vessel filter was 5.2×10^3 for all isotopes: 1.7×10^3 for Se-75, 6.9×10^3 for Ru-103, 9.3×10^3 for Cs-134 and infinite for Rb-86. The burnrate was 11.3 kgC/hr-ft². The gamma detection system composed of a NaI flowthrough detector and a single channel analyzer with a gamma count-rate meter showed no increase in gamma activity over background during the entire run.

After the run, the in-vessel filter was boiled in an acid thorex solution for 2 hours. The resulting decontamination factor (D.F.) was 18, indicating either plating of isotopes on the filter or entrapment in the pores; neither is desirable. Table VI shows the gamma activity on the filter before and after decontamination and that Ru-103 was not removed by decontamination as well as the other isotopes.

Run 4

The starting bed for Run 4 was 865g of crushed fertile particles. The operating parameters are given in Table I. The tracers were pipetted onto 30g of carbon containing 5% ruthenium, dried, and mixed with the bed. The quantities of tracer and carrier are given in Table II.

TABLE V

GAMMA ACTIVITY IN EQUIPMENT COMPONENTS FOR RUN 3

(Equipment Location and Component Nomenclature Corresponds to that Given in Figure 1)

Position (a)	Cs-134		Ru-103		Se-75		Total (c)		Average Temperature, °C
	D/S	D/S/cm ² (b)	D/S	D/S/cm ²	D/S	D/S/cm ²	D/S	D/S/cm ²	
Bed (Final)	4.2 x 10 ⁷	-	5.3 x 10 ⁷	-	1.3 x 10 ⁷	-	1.1 x 10 ⁸	-	878
Filter-1	3.7 x 10 ⁵	1.3 x 10 ⁴	1.4 x 10 ⁵	4.9 x 10 ³	4.5 x 10 ⁵	1.6 x 10 ⁴	9.7 x 10 ⁵	3.4 x 10 ⁴	514
Glass-1	8.4 x 10 ⁴	-	1.0 x 10 ⁵	-	3.6 x 10 ⁵	-	5.5 x 10 ⁵	-	-
Glass-2	4.6 x 10 ⁴	261	4.3 x 10 ⁴	244.0	1.4 x 10 ⁵	795	2.3 x 10 ⁵	1307	-
Glass-3	61	3.7	56	3.4	594	36	711	43.1	184
Glass-4	80	6.0	208	15.5	507	38	795	59.5	174
Glass-5	126	7.5	145	8.6	542	32.1	813	48.2	155
Glass-6	80	6.1	112	8.5	340	26	532	40.6	139
Glass-7	73	4.5	127	7.8	295	18	495	30.3	104
Glass-8	2410	5.6	5040	11.7	2450	5.7	9900	23.0	104
Filter-2	558	59.4	272	28.9	1340	142	2170	230.3	104
Glass-9	90	5.9	155	10.1	119	7.8	364	23.8	68
Glass-10	76	4.4	90	5.2	93	5.4	259	15.0	54
Fiber Filter	58	0.6	57	0.6	577	6.1	692	7.3	54
Glass-11	389	2.9	631	4.7	699	5.2	1719	12.8	54
Glass-12	54	3.4	66	4.2	66	4.2	186	11.8	39
Glass-13	31	2.0	27	1.8	39	2.6	97	6.4	36
Glass-14	36	2.7	59	4.4	54	4.0	149	11.1	36
Glass-15	460	1.9	650	2.7	561	2.3	1671	6.9	36
Glass-16	32	2.0	40	2.5	32	2.0	104	6.5	34
Total in Off Gas	4614	118.6	7735	120.6	8308	337.4	2.1 x 10 ⁴	576.6	-
Total	4.3 x 10 ⁷	-	5.3 x 10 ⁷	-	1.4 x 10 ⁷	-	1.1 x 10 ⁸	-	-

(a) Position of numbered glass component is shown in Figure 1.

(b) Area of each glass component is the same as that given in Table IV.

(c) Value includes Rb-86.

Figure 7. Gamma Activity Profile in Off-Gas System for Run 3 (Equipment nomenclature corresponds to that given in Figure 1.)

TABLE VI
IN-VESSEL FILTER GAMMA ACTIVITY BEFORE AND AFTER
DECONTAMINATION IN BOILING ACID THOREX SOLUTION^(a)

	<u>Run -3 Before Decontamination</u>	<u>Run -3 After Decontamination</u>	<u>Decontamination Factor</u>
Cs-134, D/S	3.7×10^5	1.5×10^4	24.7
%	38.1	28.3	
Ru-103, D/S	1.4×10^5	1.7×10^4	8.2
%	14.5	32.1	
Se-75, D/S	4.5×10^5	2.1×10^4	21.4
%	46.3	40.6	
Rb-86, D/S	1.1×10^4	0	∞
%	1.1	0	
Total, D/S	9.7×10^5	5.3×10^4	18.3

(a) Acid thorex is 13M HNO₃ - 0.1M Al(NO₃)₃ - 0.05M HF.

The average temperature during the main combustion period was 888⁰C; very good agreement between bed thermocouples was obtained indicating good mixing and fluidization. The temperature profile for the system is shown in Figure 8. The off gas was cooled to 34⁰C before leaving the system. The average in-vessel filter temperature was 492⁰C. The fluidizing velocity was 1.2 ft/sec. The average CO₂ concentration in the off gas was 59% and the CO and O₂ concentrations were again essentially zero. The average N₂ to O₂ ratio in the fluidizing gas was 2:3.

After the run, the system components were analyzed for gamma activity. The gamma activities are listed in Table VII and shown graphically in Figure 9. A drastic decrease in activity was found in the off gas relative to previous runs even though a larger quantity of tracers was used. The only explanation is the low fluidizing velocity. Figure 9 shows that Ru-103 is released from the bed. The main activity is Se-75 and there is a high percentage of Cs-134. Again no Rb-86 was found in the off-gas. There was an increase in gamma activity on the second sintered metal filter and a general decrease in gamma activity in the components downstream from the filter. The fiber filter did retain some of the tracers in this run. The temperature of the second metal filter ranged between 80 to 90⁰C during the main combustion period. The DF across the in-vessel filter was 1.6×10^5 for all isotopes. The DF for Se was 5.0×10^4 for Ru 4.8×10^4 , and for Cs 3.0×10^5 . The burnrate was 8.1 kgC/hr-ft².

The gamma detection system showed no increase in activity during any part of the run.

Figure 8. Temperature Profile of Each Equipment Component of Run 4
(Curve and thermocouple numbers correspond to equipment component nomenclature given in Figure 1.)

Figure 9. Gamma Activity Profile in Off-Gas System for Run-4
(Equipment nomenclature corresponds to that given
in Figure 1.)

TABLE VII

GAMMA ACTIVITY OF EQUIPMENT COMPONENTS FOR KLN 4
(Equipment Location and Component Nomenclature Corresponds to that Given in Figure 1)

Position(a)	Cs-134		Ru-103		Se-75		Total(c)		Average Temperature, °C
	D/S	D/S/cm ² (b)	D/S	D/S/cm ²	D/S	D/S/cm ²	D/S	D/S/cm ²	
Bed (Final)	8.7×10^8	-	3.7×10^7	-	1.2×10^8	-	1.0×10^9	-	888
Filter-1	1.8×10^6	6.9×10^4	6.6×10^4	2.3×10^3	4.5×10^5	1.6×10^4	2.3×10^6	8.1×10^4	492
Glass-1	3.7×10^5	-	2.4×10^4	-	1.1×10^5	-	5.0×10^5	-	-
Glass-2	1.5×10^5	852	1.2×10^4	68	4.7×10^4	267	2.1×10^5	1.2×10^3	-
Glass-3	71	4.3	5	0.3	287	17.2	363	21.8	160
Glass-4	260	19.4	4	0.3	150	11.2	414	30.9	149
Glass-5	82	4.9	10	0.6	150	8.9	242	14.4	136
Glass-6	33	2.5	0	0	89	6.8	122	9.3	118
Glass-7	23	1.4	5	0.3	63	3.9	91	5.6	89
Glass-8	1100	2.6	120	0.3	940	2.2	2160	5.1	89
Filter-2	220	23	44	4.7	34	3.6	298	31.3	89
Glass-9	20	1.3	0	0	0	0	20	1.3	60
Glass-10	23	1.3	3	0.2	10	0.6	36	2.1	48
Fiber Filter	330	3.5	171	1.8	210	2.2	711	7.5	48
Glass-11	520	3.9	330	2.5	400	3.0	1250	9.4	48
Glass-12	4	0.3	3	0.2	0	0	7	0.5	35
Glass-13	9	0.6	0	0	0	0	9	0.6	34
Glass-14	6	0.6	0	0	0	0	6	0.6	34
Glass-15	150	0.6	69	0.3	85	0.3	304	1.2	34
Glass-16	11	0.7	3	0.2	9	0.6	23	1.5	34
Total in Off Gas	2862	70.9	767	11.7	2427	60.5	6056	143.1	-
Total	8.7×10^8	-	3.7×10^7	-	1.2×10^8	-	1.0×10^9	-	-

(a) Position of numbered glass component is shown in Figure 1.

(b) Area of each glass component is the same as that given in Table IV.

(c) Value includes Rb-86.

V. ACTIVITY DISTRIBUTION

Distribution data for the isotopic tracers in the bed and sintered metal in-vessel filter are given in Table VIII. The greatest quantity of isotope collected on the filter was Se-75. The percentage of this isotope on the filter was two to ten fold higher than the starting percentage in the bed. The average total D.F. across the in-vessel filter was approximately 10^5 ; the D.F. appeared to be dependent on the fluidizing velocity. Figure 10, which is a plot of the decontamination factor as a function of fluidizing velocity, shows that, as the fluidizing velocity increases, the amount of activity in the off gas is increased. However, the most activity found in the off gas was 0.88% of that initially in the bed; as low as 0.13% was found at lower fluidizing velocities. The 0.88% level corresponds to 2.8×10^4 curies in the off gas during each burner batch of 6-year-irradiated, 6-month-cooled fertile particles.

Each run showed a collection of tracers on the second sintered metal filter located in the off-gas system; this increases the D.F. of the system from two to four. The fiber filter used in the off-gas showed an increase in tracer concentrations in Runs 2 and 4 when the fluidizing velocities were low. Data for Run 1 seems to deviate somewhat from the others. This is believed to be the result of a much larger bed, resulting in significantly different operating conditions as well as a very low tracer concentration in the initial bed.

TABLE VIII

DISTRIBUTION OF TRACERS THROUGHOUT SYSTEM

	Run-1	Run-2	Run-3	Run-4
<u>Bed</u>				
Cs-134, D/S	5.8×10^6	6.3×10^8	4.2×10^7	8.7×10^8
%	58.6	91.3	38.2	83.3
Ru-103, D/S	1.9×10^6	5.4×10^7	5.3×10^7	3.7×10^7
%	19.2	7.8	48.2	3.5
Se-75, D/S	2.2×10^6	3.2×10^6	1.3×10^7	1.2×10^8
%	22.2	0.46	11.8	11.5
Rb-86, D/S	0	0	6.6×10^6	1.7×10^7
%	0	0	6.0	1.6
Total D/S	9.9×10^6	6.9×10^8	1.1×10^8	1.0×10^9
cts/s	8.7×10^7	2.0×10^8	-	-
<u>In-Vessel Filter</u>				
Cs-134, D/S	8.8×10^3	6.6×10^5	3.7×10^5	1.8×10^6
%	19.5	71.7	38.1	78.3
Ru-103, D/S	8.8×10^3	2.2×10^5	1.4×10^5	6.6×10^4
%	19.6	23.9	14.5	2.9
Se-75, D/S	2.7×10^4	4.3×10^4	4.5×10^5	4.5×10^5
%	60	4.6	46.3	19.6
Rb-86, D/S	0	0	1.1×10^4	0
%	0	0	1.1	0
Total D/S	4.5×10^4	9.2×10^5	9.7×10^5	2.3×10^6
Grand Total On Filter #1, %	0.45	0.13	0.88	0.23
<u>Downstream</u>				
From Filter #1	3.1×10^3 cts	1.8×10^2 cts	2.1×10^4 D/S	6.1×10^3 D/S
DF Across Filter #1	2.8×10^4	1.1×10^6	5.2×10^3	1.6×10^5
DF Across Filter #2	5.2×10^4	2.1×10^6	2.1×10^4	4.2×10^5

Figure 10. Decontamination Factor as a Function of Fluidizing Velocity for In-Vessel Filter (Filter-1).

VI. Conclusions and Recommendations

Conclusions

1. The experimental runs showed that the in-vessel filter removes greater than 99% of the total activity.
2. The in-vessel filters may eventually plug with Ru as shown by the results of the decontamination attempts on the Run-3 filter.
3. Apparently no rubidium entered the off-gas system.
4. The major fission products component on the in-vessel filter and in the off-gas system was Se-75.
5. There was no apparent increase of activity with respect to temperature of any component in the off-gas system between 25 and 400°C.
6. The general gamma activity detected is less downstream of the second filter than upstream of the filter. This indicates that the metal filter is probably filtering particulates from the off-gas, allowing either vapor or particles of less than 0.05 microns to pass through. This is substantiated because very little buildup of tracers was detected on the fiber filter.
7. The higher the fluidizing velocity the more tracers are found in the off-gas system.
8. The material balance is essentially 100% because of the small amount of tracers entering the off-gas system compared to that remaining in the bed.
9. The tracers used in this experiment were dissolved in 0.5 M HCl and 0.5 M HNO₃. The tracers were then dried on carbon before they were mixed with bed material. This procedure does not exactly duplicate irradiated fuel, but the basic chemical reactions should be the same because of the very low relative quantity of nitrate and chloride anions present in the bed. The tests results were very similar to those experienced during other experiments in which very small quantities of irradiated fuel samples were processed.
10. Based on these data, it appears that the tracer activity in the off-gas does not result from volatile fission products but from isotopes in a very fine particulate form, perhaps in the form of an aerosol.

Recommendations

1. Two sintered metal filters and a HEPA or fiber filter should be used for experimental testing.
2. The release of fission products as a function of fluidizing velocity should be examined more extensively.
3. Decontamination of sintered metal filters should be conducted after each run.
4. The entire off-gas system including radon holdup, tritium removal, iodine removal, the catalytic oxidizer, and if possible, the Kr removal system, should be assembled on an integrated basis to determine the effect of the fission products on each system component.
5. If possible, all fission products should be tested to determine which species actually remain in the burner and which can be filtered if they reach the off-gas system.
6. The experimental tests conducted during this study were of a preliminary scoping nature. Additional studies should be conducted to verify the results of the tests, to explore possible filter plugging, and to determine the state of the fission products. The studies could be conducted using simulated fission products (tracers) or actual irradiated fuel.

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