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RADIOCHEMICAL EXAMINATION OF PEACH BOTTOM HTGR COMPONENT SAMPLES

by

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ABSTRACT

This document describes the results of a program conducted at General Atomic (GA) to radiochemically examine primary coolant circuit components of the Peach Bottom HTGR. The purpose of this program was to provide (1) absolute values of plateout activity in the primary coolant circuit, (2) fission gas release data for fuel compacts, (3) sorption data for cesium on fuel element graphite (spine and sleeve), and (4) diffusion coefficient data for cesium in fuel element graphite (spine and sleeve).

The resulting plateout data consist of specific activities for Sr-90, Cs-134, and Cs-137. These were the only reportable activities found. Iodine was below the limits of detection by neutron activation analysis. The Sr-90 activity ranged from about 5 to 10^{-4} to $9 \times 10^{-4} \mu\text{Ci}/\text{cm}^2$ in the cold duct and 2×10^{-3} to $9 \times 10^{-3} \mu\text{Ci}/\text{cm}^2$ in the steam generator. The strontium found was orders of magnitude lower than the Cs-137 values of about 0.8 to $4 \mu\text{Ci}/\text{cm}^2$ in the cold duct and 2 to $12 \mu\text{Ci}/\text{cm}^2$ in the steam generator. Additionally, it was found that the Cs-134 activity was, in general, higher than the Cs-137 activity throughout the primary helium coolant circuit. These plateout data provide (1) absolute calibration for the in-situ gamma scan data obtained previously by Oak Ridge National Laboratory (ORNL) and Intelcom Rad Tech (IRT) Corporation, and (2) verification of the in-situ gamma scan data.

Post irradiation fission gas release tests on driver fuel element compacts gave R/B values for krypton and xenon somewhat higher than measured reactor values. It was determined by metallographic examination that the small failed fraction at end-of-life was appreciably hydrolyzed after the fuel was removed from the core. This explains the relatively high R/B values found in the postirradiation tests. Failure fractions determined by metallography are in good agreement with values determined by hot-chlorine leaching experiments at ORNL.

These plateout and fission gas release results, along with data concerning metallic fission product sorption and diffusion in core graphite component materials, provide a data base for use in the verification of design methods used to predict the release of fission products from HTGR cores.

This radiochemical examination program was a part of the Peach Bottom HTGR End-of-Life Program, which was jointly sponsored by the Energy Research and Development Administration (now Department of Energy) and the Electric Power Research Institute.

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1. INTRODUCTION

A primary objective of the Peach Bottom End-of-Life (PBEOL) Program (Ref. 1), jointly sponsored by the Energy Research and Development Administration (now Department of Energy) and the Electric Power Research Institute, was to provide data concerning the extent and distribution of fission product plateout activity in the primary coolant circuit of the Peach Bottom HTGR. These data are necessary for validating design methods, consisting of computer codes and input data, for use in predicting the transport and distribution of fission products in the primary circuit of HTGR systems under normal and accident conditions (Ref. 2). A description of the Peach Bottom HTGR and a history of reactor operation are given in Ref. 3.

Preliminary evaluation of the Peach Bottom primary circuit plateout distribution was made from in-situ gamma scan measurements performed during the life of the reactor as a part of an operational surveillance program conducted at Oak Ridge National Laboratory (ORNL) (Ref. 4). After final shutdown of the reactor on October 31, 1974, the primary circuit ducting was gamma scanned by Intelcom Rad Tech (IRT) Corporation under subcontract to General Atomic (GA) as part of the PBEOL Program (Ref. 5). In addition, the steam generator tubes were gamma scanned by IRT from the secondary side to complement the duct scanning and complete mapping of the plateout distribution (Ref. 5).

Subsequent to the in-situ gamma scanning work, primary coolant circuit component removal work was initiated (Refs. 6 and 7). The component samples removed included 75-mm-diameter trepan samples from eight locations around primary coolant loop 1 and portions of steam tubes from the inlet and outlet of the superheater, evaporator, and economizer sections of the loop 1 steam generator. Component samples removed were subjected to extensive metallurgical examination (Ref. 8) and tritium permeation tests (Refs. 9 and 10).

Postirradiation examination of several driver fuel elements has been conducted at ORNL (Refs. 11-a,-b,-c,-d, and 12-a and -b). This work provided information needed for use in the verification of design methods.

The present document describes the results of a radiochemical examination program (Ref. 13) conducted at GA to provide (1) absolute values for plateout activity in the Peach Bottom primary coolant circuit, (2) fission gas release data for fuel compacts, (3) sorption data for cesium on fuel element graphite (spine and sleeve), and (4) diffusion coefficient data for cesium in fuel element graphite (spine and sleeve). The resulting plateout data, in the form of specific activities ($\mu\text{Ci}/\text{cm}^2$) provide (1) absolute calibration for the in-situ gamma scan data obtained previously by ORNL and IRT, (2) verification of the in-situ gamma scan data, and (3) a data base for verification of the design methods for predicting the release and distribution of fission products in HTGR. Use of the plateout data for design methods verification is described in Ref. 14. The resulting fission gas release, sorption, and diffusion data, along with the plateout data, provide a data base for verification of design methods used to predict the release of fission products from HTGR cores (Ref. 2).

The following sections of this report describe, in turn, (1) the radiochemical tests conducted on primary circuit component samples (trepan and steam generator tube samples), (2) fission gas release measurements performed on fuel compacts, (3) sorption tests performed on fuel element graphite samples, and (4) diffusion measurements performed on fuel element graphite samples.

2. RADIOCHEMICAL EXAMINATION OF PRIMARY CIRCUIT COMPONENT SAMPLES

Prior to the final decommissioning of the Peach Bottom HTGR, component samples from the primary coolant duct and steam generator were obtained as part of the PBEOL Program. Catalytic, Inc., under subcontract to GA, performed component sample removal operations in accordance with the work scope for the PBEOL Program. The component samples were shipped to GA for examination. Component sample removal operations are described in Refs. 6, 7, and 15.

As a part of the radiochemical examination program, the following measurements were conducted on the primary circuit component samples:

1. Gamma Counting. Gamma counting of the samples was performed to provide backup fission product distribution data for substantiating the in-situ gamma scans performed earlier (Refs. 4 and 5).
2. Leach Tests. Leaching experiments were conducted (1) to determine strontium and iodine specific activity levels, (2) to provide cesium specific activity values for comparison with gamma scan values, (3) to evaluate the effectiveness of leaching agents, and (4) to obtain information on the depth of penetration of the fission product activity. It was necessary to utilize a leach procedure to determine the amounts of strontium and iodine plated out on the components.
3. Iodine Analysis. Direct measurement of iodine by gamma counting was not possible because all the iodine activity had decayed. Determination of iodine, therefore, required radiochemical separation followed by (1) activation of stable I-129 to the gamma-emitting isotope 12.5-hour I-130 and (2) gamma counting of the I-130 activity.

4. Beta counting (strontium analysis). The important strontium nuclides Sr-89 and Sr-90 are beta emitters and can only be determined by means of radiochemical separation followed by beta counting.
5. Decontamination Tests. These tests were conducted to determine the effectiveness of selected chemical agents for decontaminating the surfaces of steam generator tube samples.
6. Carbonaceous Layer Examination. All the primary circuit components were coated with a black carbonaceous layer where exposed to the helium coolant. Analyses were performed to characterize the layer.

Sections 2.1 through 2.4 present, in turn, (1) component sample description and disposition, (2) a description of the six experimental methods, (3) results and discussion of the component sample radiochemical examination, and (4) summary and conclusions of component sample examination work.

2.1. COMPONENT SAMPLE DESCRIPTION AND DISPOSITION

2.1.1. Trepan Samples

Trepan samples (75 mm in diameter) were obtained from eight locations around coolant loop 1. The locations were selected to coincide with the external gamma scans obtained by ORNL during reactor operation (Ref. 4) and by IRT after reactor shutdown (Ref. 5). Seven locations were in the core return duct (cold) and one in the core exit duct (hot).

The original scope of work included ten locations, identified as 1-1 through 1-10 in Fig. 1. However, due to the inaccessibility of locations 1-1 and 1-5, these locations were deleted. In addition, it was originally intended to obtain four trepan samples, spaced 90 deg apart around the duct, at each location. This was not possible at locations 1-2 and 1-6 where only three trepan samples were removed.

Fig. 1. Isometric of primary coolant system

Table 1 lists the cold duct trepan samples and Table 2 lists the hot duct trepan samples as identified per Ref. 6. Location 2 was at the junction of the cold return and concentric ducting, as shown in Fig. 2. The hot gas from the reactor entered the steam generator through an inner duct containing thermal insulation (SOLAMI) on the inner surface. The cold gas returning to the reactor passed down the annulus between the inner and outer ducts. Thus, samples obtained from location 2 were composed of outer duct, inner duct, and SOLAMI insulating material from the inner duct (L2, IL2, and SIL2, respectively, in Table 2). Thirty cold duct, five hot duct, and five SOLAMI trepan samples were removed for examination.

2.1.2. Steam Generator Tube Samples

Portions of steam generator tubes (~46 cm in length) from the economizer, evaporator, and superheater sections of the Peach Bottom loop 1 steam generator were removed for examination. The details concerning removal operations are given in Refs. 6 and 7. Figure 3 shows a cross-sectional view of the steam generator and identifies the general area of steam tube removal. The actual tube samples removed are identified on the tube field shown in Fig. 4. Tables 3, 4, and 5 list the tube samples removed from the superheater, economizer, and evaporator sections, respectively. As noted below, tube samples were packaged in protective containers for preservation of surfaces or in cardboard sleeves. The tubes packaged in cardboard were useful for metallurgical studies only (Ref. 8). Samples packaged in inert gas protective containers were used in the radiochemical examination studies.

Thirty-one superheater, 20 economizer, and 16 evaporator tubes were removed for postirradiation radiochemical examination. An additional 17 superheater, 16 economizer, and 2 evaporator tubes were removed for metallurgical studies.

TABLE 1
TREPAN SAMPLES (COLD DUCT)

L3-1	L6-1	L8-1	L10-1
L3-2	L6-3	L8-2	L10-2
L3-3	L6-4	L8-3	L10-3
L3-4		L8-4	L10-4
L4-1	L7-1	L9-1	
L4-2	L7-2	L9-2	
L4-3	L7-3	L9-3	
L4-4	L7-4	L9-4	

TABLE 2
TREPAN SAMPLES (HOT DUCT)

L2-1(a)	IL2-1	SIL2-1
	IL2-2	SIL2-2
L2-3(a)	IL2-3	SIL2-3
L2-4(a)	IL2-4	SIL2-4
	IL2-5	SIL2-5

(a) Sample is in cold duct
but part of composite sample.

Fig. 2. Concentric ducting cross section

Fig. 3. Cross section of steam generator

Fig. 4. Steam generator tube sample locations

TABLE 3
SUPERHEATER TUBE SAMPLES

SUI-106 ^(a)	SUI-7 ^(a)	SUI-89	SUO-20
SUI-56 ^(a)	SUI-104	SUI-75	SUO-25 ^(a)
SUI-100 ^(a)	SUI-88	SUI-38	SUO-7
SUI-74 ^(a)	SUI-33	SUI-23	SUO-15
SUI-55 ^(a)	SUI-15	SUI-83	SUO-23
SUI-8 ^(a)	SUI-93	SUI-52	SUO-36
SUI-102 ^(a)	SUI-37	SUI-36	SUO-6
SUI-85 ^(a)	SUI-24	SUI-5	SUO-22
SUI-57 ^(a)	SUI-6	SUI-96 ^(a)	SUO-5
SUI-16 ^(a)	SUI-103	SUI-87 ^(a)	
SUI-101	SUI-63	SUI-40 ^(a)	
SUI-54 ^(a)	SUI-25	SUO-8	
SUI-26 ^(a)	SUI-20	SUO-16	

^(a) Packaged in cardboard container.

TABLE 4
ECONOMIZER TUBE SAMPLES

ECI-116	ECI-115	ECI-78	ECO-49
ECI-117	ECI-95	ECI-60	ECO-68(a)
ECI-118	ECI-101(a)	ECI-87	ECO-87
ECI-119	ECI-102(a)	ECI-68(a)	ECO-104(a)
ECI-120	ECI-103(a)	ECI-49	ECO-115
ECI-121	ECI-104(a)	ECI-28(a)	ECO-7(a)
ECI-122	ECI-105(a)	ECI-9	ECO-8(a)
ECI-113(a)	ECI-106	ECO-9	ECO-10(a)
ECI-114(a)	ECI-96	ECO-28(a)	ECO-11(a)

(a) Packaged in cardboard containers.

TABLE 5
EVAPORATOR TUBE SAMPLES

EVI-167	EVI-103	EVI-78
EVI-168	EVI-104	EVI-50(a)
EVI-169	EVI-105	EVI-51
EVI-170	EVI-75	EVI-52
EVI-128	EVI-76	EVI-25(a)
EVI-102	EVI-77	EVI-26

(a) Packaged in cardboard containers.

Prior to steam tube sample removal operations, a section of the steam generator shroud material was removed for examination. The shroud consisted of a 1-cm-thick outer stainless steel plate and 2.5-cm-thick SOLAMI insulating material composed of corrugated stainless steel sheets approximately 1 mm thick. The shroud samples were shipped in a protective container for radiochemical examination.

2.1.3. Tube and Trepan Sample Shipping and Handling

Catalytic, Inc., was responsible for removing, packaging, and shipping the tube and trepan samples to San Diego in accordance with Ref. 6. The samples, shipped in 200-liter steel drums, arrived via common carrier on February 28, 1976. The drums were all in satisfactory condition with no visible evidence of physical damage. All samples received were cross checked against the shipment bill of lading and verified through documentation provided by Catalytic, Inc., in accordance with GA Quality Control standards (Ref. 16).

The individual samples were shipped sealed in inert-environment containers pressurized to 0.2 MPa (2 atm) with dry nitrogen. Upon receipt, all containers were pressure checked, examined for damage, and gamma scanned. The containers were gamma scanned before and after sample removal to provide a gross indication of fission product activity losses from the samples during shipping.

After removal from the shipping containers, each sample identification was verified and the physical condition of each sample was recorded. Table 6 gives the as-received condition of the containers and samples. Examination of the before and after gamma scan data in Table 6 shows that shipping did not appreciably affect the as-packaged condition of most of the samples. However, activity losses from some samples (e.g., trepan location 2 and superheater outlet tube SUO-6) were appreciable. [Activity losses were taken into account in estimating the uncertainty of specific activity data (see Section 2.3.3).]

TABLE 6
DESCRIPTION AND APPEARANCE OF AS-RECEIVED SAMPLES

Drum No.	Sample Container No.	Container Internal Gas Pressure [kPa (psig)]	Sample No.	Gross Gamma Activity (a) (mR/h)						Sample Appearance
				Before			After			
				T	M	B	T	M	B	
1	7	0	L6	2	9	7	1	1	1	Good, no surface damage
	9	78 (11.3)	L2	--(b)	--	--	--	--	--	Good, no surface damage
	10	73 (10.7)	L2I	12.5	8.5	5	9	1	1	Good, no surface damage
	12	0	L2	1	13	8	0	2	1	Good, no surface damage
2	5	0	L9	4	7	5	1	1	1	Good, no surface damage
	6	0	L8	5	11	8	--	--	--	Good, no surface damage
	8	83.5 (12.1)	L2	9	14	17	9	12	15	Good, no surface damage
	11	0	L2I	9	15	17	2	5	7	Good, no surface damage
3	1	0	L4	6	9	7	--	--	--	Good, no surface damage
	2	0	L10	3	10	5	--	--	--	Good, no surface damage
	3	0	L3	7	15	9	2	2	2	Good, no surface damage
	4	0	L7	5	11	7	--	--	--	Good, no surface damage
4	22	0	SUI-75	14	90	60	6	4	7	--
	7	0	SUI-101	--	--	--	--	--	--	Smudges and fingerprints
	27	0	SUI-88	30	85	65	5	5	10	Scratches and fingerprints
	13	0	SUI-93	20	30	40	6	6	7	--
	12	0	SUI-33	14	85	21	5	8	6	--
	4	76 (11.1)	SUI-104	25	130	85	--	--	--	--
	59	72 (10.5)	SUI-37	30	95	70	--	--	--	--
5	5	0	SUO-6	21	130	82	10	25	60	--
	49	76 (11.0)	SUI-24	25	70	60	5	7	10	--
	58	0	SUI-15	30	70	60	6	8	12	--
	55	0	SUI-89	--	--	--	--	--	--	--
	11	100 (14.5)	SUI-25	14	90	75	--	--	--	--
	14	48 (7.0)	SUI-63	--	--	--	--	--	--	--
	28	0	SUI-103	--	--	--	--	--	--	Scratches along length of tube, no rust
6	61	86 (12.5)	SUI-36	22	90	47	--	--	--	--
	45	0	ECI-122	11	23	16	1	1	1	--
	36	80 (11.6)	SUO-15	40	85	70	--	--	--	--
	21	0	SUO-22	30	90	70	--	--	--	--
	2	55 (8.1)	ECI-117	11	30	18	1	2	2	Scratches, rust, no identifying marks
	53	0	SUO-5	20	100	20	--	--	--	--
	10	0	SUO-36	50	100	80	--	--	--	--
7	34	10 (1.5)	ECI-115	--	--	--	--	--	--	Scratches and rust
	23	0	ECI-119	15	25	21	1	1	1	Smudges and scratches
	16	0	ECI-120	14	30	20	1	1	1	--
	20	0	ECI-121	11	22	29	1	1	2	--
	37	6.9 (1.0)	ECI-78	25	45	60	3	5	4	Scratches on one side
	50	0	ECI-96	20	40	30	4	3	5	Smudges and scratches, bottom one-third cleaned of deposit
	42	0	ECI-60	--	--	--	--	--	--	Scratches and smudges along length

TABLE 6 (Continued)

Drum No.	Sample Container No.	Container Internal Gas Pressure [kPa (psig)]	Sample No.	Gross Gamma Activity(a) (mR/h)						Sample Appearance
				Before			After			
				T	M	B	T	M	B	
8	62	80 (11.6)	SUI-20	13	85	50	3	3	3	--
	51	79 (11.5)	SUI-38	16	85	60	3	5	4	--
	38	0	SUO-7	50	130	95	6	6	9	Smudges and scratches
	25	82 (12.0)	SUO-23	55	90	65	5	7	9	--
	43	0	SUO-20	10	110	40	1	7	10	--
	31	0	SUO-8	50	120	110	1	1	1	Scratches and fingerprints
	3	82 (12.0)	SUI-5	22	100	41	1	1	1	--
9	19	27 (4.0)	ECI-106	20	30	25	2	1	3	Smudges, scratches, and rust
	9	0	ECI-116	11	25	17	1	1	1	Rust all over, scratches
	29	0	ECI-95	20	35	30	1	1	3	Smudges, scratches, and rust
	30	0	ECI-118	15	30	25	1	1	2	Smudges and scratches
	18	86 (12.5)	ECI-49	12	30	17	1	1	2	Scratches along length of tube, rust
	15	82 (12.0)	ECI-87	7	32	23	2	2	4	--
	47	83 (12.1)	ECI-9	9	18	12	2	1	4	--
10	64	0	EVI-104	25	70	60	1	2	2	Smudges and fingerprints, no rust
	40	0	EVI-103	30	60	40	2	2	2	--
	39	31 (4.5)	EVI-167	30	60	55	2	2	2	Smudges and scratches, poor condition
	60	89 (13.0)	EVI-170	25	70	55	3	3	3	Scratches, tube in poor condition
	68	0	EVI-105	35	70	55	2	2	2	Scratches and fingerprints
	26	87 (12.6)	EVI-168	30	50	30	2	2	2	Scratches and fingerprints
	1	0	EVI-128	7	55	40	1	2	2	Short tube, multiple scratches
11	6	34 (5.0)	EVI-26	24	70	35	--	--	--	--
	17	0	EVI-75	45	70	40	2	2	3	Scratches and fingerprints along length of tube
	57	0	EVI-78	--	--	--	--	--	--	--
	35	0	EVI-51	40	75	40	2	2	3	--
	44	0	EVI-169	40	60	40	3	3	4	Smudges and scratches on one side
	63	0	EVI-52	19	60	50	2	3	3	--
	33	0	EVI-77	35	60	50	3	4	4	--
12	54	24 (3.5)	SUI-6	20	100	55	1	1	1	--
	8	79 (11.5)	SUI-83	--	--	--	--	--	--	--
	56	0	SUI-52	35	100	70	--	--	--	--
	52	0	SUI-23	28	85	65	--	--	--	--
	48	0	SUO-16	17	80	70	3	3	4	--
	46	0	ECO-49	--	--	--	--	--	--	Scratches and fingerprints
	41	0	ECO-9	10	25	22	--	--	--	Scratch down length of tube

TABLE 6 (Continued)

Drum No.	Sample Container No.	Container Internal Gas Pressure [kPa (psig)]	Sample No.	Gross Gamma Activity(a) (mR/h)						Sample Appearance
				Before			After			
				T	M	B	T	M	B	
13	69	0	EVI-25	--	--	--	--	--	--	Cardboard container Scratches and rust Cardboard container -- -- Short tube, scratches
	24	0	ECO-115	17	30	12	2	2	3	
	66	--	EVI-50	--	--	--	--	--	--	
	32	91 (13.3)	ECO-87	8	35	23	2	2	2	
	65	0	EVI-102	20	60	45	--	--	--	
	70	--	EVI-76	--	--	--	--	--	--	
			SUI-40							
			SUI-7							
			SUI-54							
			SUI-26							
14			SUI-100							These samples were shipped in cardboard containers. Gamma activities were not measured. The samples were taken for use in metallurgy studies.
			SUI-96							
			SUI-57							
			SUI-74							
			SUI-85							
			SUI-55							
			SUI-56							
			SUI-87							
			SUI-16							
			SUI-8							
			SUI-106							
			ECI-114							
			SUO-25							
			ECI-101							
			ECI-113							
			ECI-68							
			ECI-102							
			ECI-104							
			ECI-105							
			ECI-103							
			ECI-28							
			ECO-7							
			ECO-8							
			ECO-11							
			ECO-28							
		ECO-10								
		ECO-68								
		ECO-104								
15	13		Shroud							Good, no surface damage

(a) Gross activity measurement made before and after sample removal from shipping container at top (T), middle (M), and bottom (B) of container.

(b) -- means not measured.

Examination of the tube samples after removal from the shipping containers revealed substantial disturbance of the surfaces of several tubes. This was assumed to have resulted from handling in very restricted conditions prior to loading into the containers. As noted in the last column of Table 6, a major portion of the tubes were scraped, smudged, or displayed fingerprints on the outer surface. In addition, some carbon steel tube samples received in containers with low internal gas pressure were found to be rusted, indicating air ingress during shipping. As indicated in Table 6, about 60% of the containers showed no gas pressure, indicating leakage of cover gas. It should be noted, however, that all containers were leak tested and accepted by Quality Assurance prior to shipment. It is therefore assumed that the leaks developed during transit.

After completion of gamma scanning, the samples were sectioned and distributed for additional radiochemical studies, metallurgy examination, tritium permeation tests, and archive storage. A number of samples were also prepared for shipment to organizations outside GA. Table 7 lists the sample destination, description, and quantity of material shipped. The samples are being studied by the various organizations to provide additional information on metallic fission product plateout, interaction with the base metal, and liftoff. These tests were not funded under PBEOL Program and will be reported separately by the recipient organizations.

Portions of tube and trepan samples were stored in the inert gas pressurized shipping containers to provide as-received samples for additional tests should they become necessary.

2.2. EXPERIMENTAL METHODS FOR RADIOCHEMICAL EXAMINATION OF PRIMARY CIRCUIT COMPONENT SAMPLES

This section describes the various methods and procedures used for radiochemical examination of primary circuit component samples.

TABLE 7
DISTRIBUTION OF COMPONENT SAMPLES TO OUTSIDE ORGANIZATIONS

Destination	Shipping Container	Sample Container No.	Sample Identification	Sample Length (cm)	Number of Samples
Sulzer Bros., Switzerland	Certified wooden crate	22	SUI-75	16.5	2
Harwell, England	Certified wooden crate	--	SUI-5	1.3	1
			SUO-5	1.3	1
			EVI-26	1.3	1
			ECI-87	1.3	1
			ECO-87	1.3	1
ORNL	Drum No. 13	62	SUI-20	16.5	2
		14	SUI-64	16.5	2
		28	SUI-103	16.5	2
		31	SUO-8	16.5	2
		45	ECI-120	16.5	2
		16	ECI-122	16.5	2
	Drum No. 2	4	L3-4	7.6	1
			L7-4	7.6	1
	Drum No. 5	10	SUO-36	10.2	2
		4	EVI-52	10.2	2
			SUI-104	10.2	2
		12	SUI-52	10.2	2
			SUI-15	10.2	2
		26	SUI-33	10.2	2
			EVI-168	10.2	2
		36	SUI-24	10.2	1
			SUO-15	10.2	2
		47	EVI-77	10.2	2
			ECI-49	10.2	2
		18	ECI-9	10.2	1
			EVI-51	10.2	1
			EVI-103	10.2	2
CEA, France	Drum No. 8	46	ECO-9	10.2	2
		65	ECO-49	10.2	1
			EVI-102	10.2	2
		24	ECO-115	10.2	2
		27	SUI-101	10.2	2
		30	SUI-88	10.2	2
			ECI-118	10.2	1
			ECI-119	10.2	1
			ECI-95	10.2	1
			ECI-78	10.2	1

2.2.1. Method for Sample Gamma Scanning

The trepan and tube samples, as received from the reactor, were subjected initially to nondestructive gamma scanning. A block diagram describing the necessary equipment for gamma scanning is shown in Fig. 5. The Ge(Li) detector had a resolution of 2.5 keV full width half maximum (FWHM) for the reference Co-60 1330 keV photopeak. The detector was calibrated with a National Bureau of Standards (NBS) traceable standard. The various geometries (tube, trepan, and leach solution) were individually calibrated. An NBS Cs-137 source was periodically counted to assure that the detector performance did not change during the period of sample examination.

The equipment, as shown in Fig. 5, consisted of a detector, preamplifier, an amplifier, a baseline restorer for detector signal shaping, a line driver for transmitting the detector signal to the multichannel analyzer with minimal distortion, and the SIGMA II computer system with appropriate programs for analysis of the recorded spectra (Ref. 17). All spectra obtained were stored on magnetic tape for future reference. The spectra were analyzed for all identifiable photopeaks. Integration of photopeaks and calculation of specific activities of observed radioisotopes were performed as described in Ref. 17.

Steam generator tube samples were gamma scanned axially and circumferentially using the geometry and equipment shown in Fig. 6. A Unislide positioning device allowed axial gamma scanning of tube samples automatically every 2.5 mm over the length of the tube. In addition, at each axial location, four scans were obtained 90 deg apart circumferentially around the tube with the 0 deg spectrum referenced to the tube identification and marking procedure (Ref. 7). The axial and circumferential scans were obtained to determine plateout activity gradients within the steam generator tube bundle.

Fig. 5. Block diagram of equipment used for gamma counting

Fig. 6. Diagram of equipment and geometry for gamma scanning of tube samples

The Ge(Li) detector was protected from stray gamma rays by 100 mm of lead shielding with an adjustable front surface collimator as shown in Fig. 6. The collimator reduced the gross activity to a level suitable for appropriate detector response (i.e., a dead time indication of $\leq 10\%$).

Trepan samples were gamma scanned at three locations across the face of the disk in the direction of coolant flow. Each trepan sample was marked prior to removal from the coolant loop ducting for location and coolant flow direction. Each scan represented $\sim 0.5 \text{ cm}^2$ of surface or an average of $\sim 2\%$ of the total trepan surface area.

2.2.2. Method for Leach Experiments

Preliminary leach experiments were carried out to determine (1) the method best suited to the leaching of cesium and strontium activities from tube and trepan samples, and (2) whether I-129 could be detected in the leach solutions.

Three steam generator tube samples (ECI-60, EVI-78, and ECI-87) and one trepan sample (L-10) were selected for this initial testing. The samples were cut in half, and all eight halves were weighed. One-half of each sample was warmed for 30 minutes in 1N NaOH to which was added 1 ml NaOCl plus iodine and strontium carriers. Following removal of these half samples from the basic leach solution, each was rinsed, dried, and reweighed. This set of four half samples plus the other set of four, as yet untreated, half samples were warmed for 10 minutes in 4N HNO_3 with hydrazine hydrate and strontium and iodine carriers. All samples were rinsed and dried and final weights were obtained. The 12 component samples were gamma counted after leaching. The 12 solutions were gamma counted and submitted to Analytical Chemistry (Radiochemical Section) for I-129 and Sr-90 analyses (Ref. 18) (see Sections 2.2.3 and 2.2.4).

Results of these preliminary analyses (see Section 2.2.4) showed that rather severe leaching conditions, in terms of time, temperature, and acid concentration, were required in order to remove all the activity from the samples. In many cases, it was necessary to remove appreciable amounts of the base metal in order to remove all the activity.

In view of these results, the following acid leach method was adopted. The samples to be leached were placed in a test tube or beaker, which was mounted in an ultrasonic cleaner. Acid solutions used were 4N HNO_3 and 3N HCl . The acid solution was added, and after a period of 7 to 8 hours, the leach solution was decanted into a clean 0.1 liter (4-oz) polyethylene bottle. The sample was rinsed three times with distilled water, and the washings were added to the leach solution in the polyethylene bottle. The sample was then checked for gamma activity with a G-M counter. If appreciable activity remained, the acid leach was repeated. All the leach solutions contained a strontium carrier. A procedure for the acid-leach work was written and approved (Ref. 19).

This acid-leach procedure was performed on 12 steam generator tube samples and 3 trepan samples. The tube samples, which were about 25 mm in length, were cut from the middle of the original (as-received) tube samples. The trepan samples were as-received.

In accordance with the leach procedure (Ref. 19), one sample was leached in a stepwise manner to determine leach rates and to provide information concerning the depth of penetration of activity into the material. A SOLAMI sample (SIL2-2, from the inner hot duct) was selected for this stepwise leach experiment. The first part of the stepwise leach experiment was conducted utilizing 4N HNO_3 solution. The leach times for each treatment are included in Table 17 which is presented with the results in Section 2.3.2. After five leach treatments with the 4N HNO_3 solution and a total leach time of 410 minutes, most of the cesium activity still remained. The cesium activity was completely removed by leaching with 3N HCl solution for approximately 48 hours.

All leach solutions contained flakes of the carbonaceous material deposited on the helium side of all samples. This residual undissolved material was filtered away from the leach solutions. The residue and leach solutions were analyzed for all detectable activity using gamma counting equipment in combination with the Sigma II computer-analyzer system (see Section 2.2.1).

Finally, a leach test was performed in which a 25-mm tube sample, after weighing, was fired in air at 900°C for 15 minutes in a furnace before leaching in order to burn off the layer of carbonaceous material. After the firing the residue was collected, dissolved, and analyzed for strontium. The sample after burnoff was acid-leached to remove the remaining activity. The resulting leach solutions and residues were analyzed for strontium (see Section 2.2.4).

2.2.3. Method for Iodine Analysis

An activation analysis technique was used in an attempt to determine the amount of iodine plated out in the primary circuit. Direct measurement of iodine by gamma counting was not possible due to the long delay between reactor shutdown and component examination.

Activation analysis is a nuclear method of elemental analysis in which radioisotopes are created from the elements in a sample by bombardment with suitable radiation. The radioisotopes, produced in direct proportion to the quantity of respective elements, are then identified and quantitatively measured. The procedure consists generally of two steps: (1) activation of the sample, and (2) measurement of the produced radioisotopes by gamma counting. The method as used in this work involves activation of I-129, a long-lived fission product ($t_{1/2} = 1.6 \times 10^7$ years) which activates in a thermal neutron flux to I-130 with a half-life of 12.5 hours.

Twelve solution samples (four basic and eight acidic) resulting from preliminary leach experiments (see Section 2.2.2) and representing four plateout locations were delivered to Analytical Chemistry for I-129 analysis (Ref. 18).

Before activation, radiochemical separations for the isolation and purification of iodine (iodine carrier plus I-129) were carried out. Steps of this separation procedure included extraction of iodine into carbon tetrachloride, extraction of iodide into H_2SO_3 , extraction of iodine into xylene, and re-extraction of iodide into H_2SO_3 . These iodide solutions and carrier iodide standards were irradiated in the TRIGA Mark I rotating specimen rack for 5 minutes at a reactor power level of 10 kW. Carrier iodide is monoisotopic I-127 which activates in a thermal neutron flux to 25.0-minute I-128, whose principal gamma ray is 0.44 MeV. The direct comparison of standard and sample photopeak count rates corrected for decay gave the chemical yield for each separation. The samples, along with I-129 comparator standards, were then irradiated at 250 kW for 30 minutes. Following decay of the 25-minute I-128 from the carrier iodide, the samples and standards were counted for the 12.5-hour I-130. Comparison of standard and sample photopeak count rates, corrected for decay and yield, would give the I-129 concentration of each sample, if it were detectable in the leach solutions.

2.2.4. Method for Beta Counting (Strontium Analysis)

Samples of the leach solutions, which contained a strontium carrier, were analyzed for fission product strontium. The method used is based upon a separation technique whereby strontium is separated from other fission products by precipitation as the carbonate (Ref. 20).

The strontium procedure includes basic precipitation with carbonate for volume reduction, two fuming nitric acid precipitations, an iron hydroxide scavenge for decontamination, and a final basic carbonate precipitation for filtering, yield determination, and beta counting of the 28.9-year Sr-90 isotope plus the 64.0-hour Y-90 isotope as it grows in.

After separation, the strontium activity was assayed by counting in a 2π gas proportional beta detector. The beta counting apparatus was calibrated using NBS traceable standards. The carbonate precipitate was counted several times during in-growth of the daughter product yttrium,

and solution of simultaneous equations based on these counting data yielded Sr-90 activities. Specific activity for Sr-90 plateout was then determined for the specific sample studied.

The first solutions to be submitted for strontium analyses were the 12 solutions from the preliminary leach experiments (see Section 2.2.2). The results, given in Table 8, consist of (1) the amount of strontium activity removed by the base treatment, (2) the amount of activity removed by a subsequent acid treatment of the same sample, and (3) the amount of activity removed by acid treatment of the sister sample. The results indicate that the acid solutions were more effective than the base solutions in removing strontium activity. It was concluded that rather severe leach conditions were required. (See Section 2.2.2 for description of leach procedure adopted.)

The second phase leach solutions submitted to Analytical Chemistry for strontium analyses consisted of 12 solutions resulting from the acid-leach treatment of 12 steam generator tubes and 3 solutions resulting from the acid-leach treatment of 3 trepan samples.

As described in Ref. 18, in all cases the solutions contained residual, undissolved material. The residues were filtered out, and the residues and solutions were treated as separate samples. Residues were dried in platinum dishes with strontium carrier, ashed, and then fused with sodium carbonate. The carbonate melt was acid dissolved. From this point, the regular strontium radiochemistry procedure was followed. In several cases, gram quantities of iron and/or nickel in the residue were soluble following the fusion. For both residue and solution samples, the first step in the procedure is, of necessity, a basic carbonate precipitation of the strontium, which allows it to be concentrated to a small volume for subsequent steps.

As indicated above (Section 2.2.2), severe sample leaching was required on many of the samples in order to remove the gamma activity. This resulted in relatively large quantities of iron and/or nickel in solution. For the basic carbonate precipitation, the solution must first

TABLE 8
STRONTIUM-90 RESULTS OF PRELIMINARY LEACH EXPERIMENTS

Solution	Sr-90 Activity (dpm)	Solution	Sr-90 Activity (dpm)
ECI-60 base 1	3.00×10^3	ECI-87 base-1	1.43×10^3
ECI-60 acid-1	5.98×10^3	ECI-87 acid-1	3.53×10^3
ECI-60 acid-2	4.30×10^3	ECI-87 acid 2	7.50×10^3
EVI-78 base-1	6.00×10^3	L-10 base-1	1.29×10^3
EVI-78 acid-1	1.03×10^4	L-10 acid-1	3.19×10^3
EVI-78 acid-2	3.64×10^4	L-10 acid-2	2.27×10^3

be made basic, bringing down great quantities of iron and/or nickel hydroxides which either largely or completely carry with them the strontium carrier. No strontium yield was obtained for several of the samples and a rather low yield was, with much effort, obtained on several of the others. The low yield is important in terms of the uncertainty which must be attached to the final strontium activity.

As described in Section 2.2.2, a trepan sample (SIL2-2) was given five timed-leach treatments. This resulted in ten strontium analyses, five on the solutions and five on the residues.

Also, as described in Section 2.2.2, an untreated superheater tube sample (SU0-23) was fired before leaching in order to eliminate the carbonaceous layer. The loose residue was treated as one sample for strontium analysis; the cold-leaching solution and the hot-leaching solution were treated as two separate samples; and the combined residue from both leachings was considered a fourth sample.

2.2.5. Method for Decontamination Tests

Decontamination tests were performed on four steam-tube samples in accordance with an approved procedure (Ref. 13). The decontamination agents used and the treatment conditions are given in Table 9. Two of the agents, TURCO 4501-A and TURCO 4502, are proprietary formulations provided by the TURCO Products Division of Purex Corporation. TURCO 4501-A is a highly alkaline amber liquid, which contains potassium hydroxide. TURCO 4502 is a highly alkaline powder material, which contains potassium permanganate as well as a stable wetting agent.

Each of the four steam-tube samples was 25 mm long. They were cut from large as-received tube samples after the samples were gamma scanned (see Section 2.2.1). Each of the four samples was placed in a clean test tube and treated in turn with each of the agents under the conditions given in Table 9. After the specified time at temperature for each treatment, the solutions were decanted into four polyethylene bottles, the

TABLE 9
OPERATION SEQUENCE AND CONDITIONS FOR DECONTAMINATION TESTS

Sequence of Operation(a)	Reaction Temperature	Time of Treatment (h)
1. Degrease ^(b)	Room temp	0.5
2. Alkaline immersion ^(c)	Room temp	1
3. TURCO 4501-A immersion	120°C ^(d)	1
4. TURCO 4502 immersion	100°C ^(d)	1
5. Acid immersion ^(e)	Room temp	0.5
6. Detergent	Room temp	1

(a) Distilled water rinse after each step.

(b) 1,1,1-trichloroethane treatment.

(c) 1N NaOH

(d) Treatment temperature recommended by manufacturer
of agent.

(e) 4N HNO₃.

tube samples and test tubes were washed with distilled water, and the washings were added to the polyethylene bottles. The polyethylene bottles were then sealed, wipe tested, and transferred to the counting laboratory.

Each solution was gamma counted, and activities of Cs-134 and Cs-137, which were the only activities detected, were calculated (by the SIGMA II computer) from the count data. From these activities and from the sample dimensions, specific activities removed ($\mu\text{Ci}/\text{cm}^2$) were calculated.

2.2.6. Method for Carbonaceous Layer Examination

The primary circuit components all contained a black carbonaceous-appearing deposit on the side exposed to the helium coolant. An effort was made to characterize this deposit.

The deposit on several tube samples was examined by visual and mechanical techniques. A section of superheater tube (SUI-16) was submitted for microprobe analysis to determine the composition and thickness of the surface deposit. An attempt was made to determine fission products, such as cesium, barium, and cerium nuclides, within the deposit using the microprobe. A sample of the deposit was analyzed by emission spectroscopy.

2.3. RESULTS AND DISCUSSION OF PRIMARY CIRCUIT COMPONENT RADIOCHEMICAL EXAMINATION

The results of the radiochemical examination of the primary circuit component samples, obtained using the methods described in Section 2.2, are presented and discussed in the following sections.

2.3.1. Results of Component Sample Gamma Scanning (Including Cesium Data)

Gamma scans were performed on (1) selected trepan samples, (2) almost all of the steam generator tube samples, and (3) a hot duct trepan sample, including the thermal insulation. Examination of the gamma spectra showed that only the cesium isotopes, Cs-134 and Cs-137, were present at

detectable levels. The Cs-134 activity was found to be consistently higher than the Cs-137 activity. Trace amounts of Co-60 were evident in some spectra. ORNL external gamma scans showed several metallic radioisotopes in addition to Cs-134, Cs-137, and Co-60. The difference between ORNL and GA observations is probably due to the higher detector efficiency of ORNL equipment.

As shown in Fig. 1, the trepan locations were chosen to coincide with the in-situ gamma scans obtained by ORNL (Ref. 4) and IRT (Ref. 5). The trepan samples from each location, listed in Tables 1 and 2, were scanned at three points across the face of the disk in the direction of coolant flow. There was little apparent difference between scans or between samples at each location; therefore, all scans from each trepan location were averaged. The results in the form of specific activities are given in Table 10. Data are included for both the hot (reactor exit) and cold (reactor return) ducts.

The hot duct scan, location 1-2 (inner), includes the thermal insulation (SOLAMI) material and inner surface of the inner duct trepan. Figure 7 shows the sample geometry, which includes a hot gas face plate (identified as A), corrugated stainless steel sheets (B), an outer cold face plate (C), and the inner duct trepan (D). The specific activities of the various sections of the hot duct sample are given in Table 11. The corrugated sheets and cold face plate were gamma scanned as one piece, resulting in an activity approximately twice the activity of the hot face plate. This was not unexpected since the surface area of the corrugated sheets is substantially greater than that of the hot face plate. The sum of sections A, B, C, and D is given in Table 10 as position 1-2 (inner). The data in Table 10 show a decrease in cesium activity in the direction of coolant flow. The Cs-137/Cs-134 ratio appears to be increasing in the direction of flow, although there is scatter in the data.

TABLE 10
CESIUM PLATEOUT VALUES DETERMINED FROM
GAMMA SCANS OF TREPAN SAMPLES

Position ^(a)	Specific Activity ($\mu\text{Ci}/\text{cm}^2$) ^(b)		Ratio Cs-137/Cs-134
	Cs-134	Cs-137	
1-2 (inner)	5.06 ^(c)	3.66 ^(c)	0.72
1-3	2.66	2.21	0.83
1-4	1.99	1.69	0.85
1-6	1.86	1.52	0.83
1-7	1.55	1.23	0.79
1-8	1.88	1.51	0.80
1-10	1.03	0.98	0.95
1-2 (outer)	1.03	0.82	0.80

(a) Refer to Fig. 1 for position locations. Position 1-2 (inner) is in the hot duct. Other positions are in the cold duct (downstream of the steam generator).

(b) Each specific activity value is the average of all values obtained from gamma scans for trepan samples at a given location (see text). All activities ($1 \mu\text{Ci}/\text{cm}^2 = 370 \text{ Bq}/\text{mm}^2$) were decayed back to final shutdown on October 31, 1974.

(c) The activities shown for position 1-2 (inner) were assumed to all be on the hot gas face plate surface; therefore, the surface area used was the detector collimator area of 1.4 cm^2 .

Fig. 7. Typical thermal barrier (SOLAMI) cross section

TABLE 11
CESIUM ACTIVITY ASSOCIATED WITH
HOT DUCT TREPAN SAMPLE SIL2-2

Section Counted	Specific Activity ($\mu\text{Ci}/\text{cm}^2$) (a,b)	
	Cs-134	Cs-137
A,B,C, and D	5.06	3.66
A	2.33	1.29
B,C, and D	2.95	2.42
D	0.65	0.99

(a) All activities shown were decayed back to final shutdown on October 31, 1974 ($1 \mu\text{Ci}/\text{cm}^2 = 370 \text{ Bq}/\text{mm}^2$).

(b) Surface area used for all sections counted was the detector collimator area of 1.4 cm^2 .

The results of the steam generator tube gamma scans are shown in Table 12. The data for the superheater tubes show a general increase in activity down the length of the tube. The increase found was small ($\sim 20\%$) but consistent throughout the tube samples studied. A similar trend was observed for the in-situ gamma scans reported in Ref. 5. The reported axial profile, Fig. 19 of Ref. 5, shows specific activities highest near the impingement baffle at the entrance to the steam generator and lowest at the ends of the bundle. The tube samples examined after removal were taken from a location some 2 m below the bottom of the tube sheet, just above the impingement baffle. This area of the tube was found by in-situ gamma scans to have the steepest activity gradient.

The axial concentration gradient appears to be highest in the superheater tubes and nearly flat in the economizer section of the steam generator. This again is consistent with the data reported in Ref. 5.

The specific activities given in Table 12 are the average of four scans circumferentially around the tube at the top, middle, and bottom of each sample. The four circumferential counts were found to vary by a factor of two between the front (helium inlet side) and rear (helium outlet side) surface of the tube. However, for comparison with the in-situ gamma scanning data, the four counts were averaged.

Figure 8 shows the cesium deposition profile in the primary coolant duct downstream of the steam generator. The data points were taken from Table 10. The curve in Fig. 8 is a least-squares fit of in-situ gamma scan data for Cs-137 obtained by IRT (Ref. 5). It can be seen that the present data and the IRT data for Cs-137 agree.

This agreement in the cold duct is in contrast to the steam generator data. Cesium-134 activity was reported by IRT (Ref. 5) to be lower than Cs-137 throughout the steam generator. The data reported here indicate the Cs-134 activity to be substantially higher than the Cs-137 activity (see Fig. 9). Some of the data points in Fig. 9 represent an average of data for several tubes at a given radial depth into the steam generator bundle. Thus, the first Cs-137 data point in the superheater inlet section

TABLE 12
SPECIFIC ACTIVITIES OF Cs-134 AND Cs-137 DERIVED FROM STEAM GENERATOR TUBE GAMMA SCANS

Sample No.	Location (a)	Specific Activity(b)		Ratio Cs-137/ Cs-134	Sample No.	Location (a)	Specific Activity(b)		Ratio Cs-137/ Cs-134
		(μCi/cm ²) (c)					(μCi/cm ²) (c)		
		Cs-134 (d)	Cs-137				Cs-134 (d)	Cs-137	
SUI-5	Top	10.80	7.47	0.69	SUI-52	Top	10.20	7.13	0.70
	Middle	14.50	9.84	0.68		Middle	13.70	9.57	0.70
	Bottom	15.60	10.40	0.67		Bottom	13.80	9.15	0.66
SUI-6	Top	10.80	7.38	0.68	SUI-63	Top	11.50	8.21	0.72
	Middle	14.40	9.73	0.67		Middle	15.40	10.30	0.67
	Bottom	13.20	8.96	0.68		Bottom	16.00	10.60	0.66
SUI-15	Top	9.03	6.09	0.67	SUI-75	Top	6.93	4.98	0.72
	Middle	11.10	7.36	0.66		Middle	15.20	10.70	0.70
	Bottom	13.80	9.23	0.67		Bottom	14.40	9.60	0.69
SUI-20	Top	11.30	7.91	0.70	SUI-83	Top	9.87	6.78	0.69
	Middle	14.10	9.78	0.69		Middle	12.10	8.27	0.69
	Bottom	15.00	10.10	0.67		Bottom	12.10	7.87	0.65
SUI-23	Top	8.81	6.10	0.69	SUI-88	Top	10.40	7.17	0.69
	Middle	12.00	8.42	0.70		Middle	12.40	8.34	0.67
	Bottom	13.80	9.09	0.66		Bottom	14.70	9.69	0.66
SUI-24	Top	8.29	5.43	0.65	SUI-89	Top	9.05	6.04	0.67
	Middle	10.30	6.66	0.65		Middle	15.50	10.60	0.68
	Bottom	12.80	8.44	0.66		Bottom	17.20	11.50	0.67
SUI-25	Top	8.21	5.95	0.72	SUI-93	Top	10.60	7.36	0.69
	Middle	11.40	7.67	0.67		Middle	16.00	11.00	0.69
	Bottom	13.90	9.44	0.68		Bottom	18.50	12.60	0.68
SUI-33	Top	8.67	5.91	0.68	SUI-101	Top	11.00	7.71	0.69
	Middle	11.10	7.63	0.69		Middle	16.60	11.30	0.68
	Bottom	12.00	7.82	0.65		Bottom	18.10	12.50	0.69
SUI-36	Top	6.14	4.41	0.72	SUI-103	Top	9.67	6.61	0.68
	Middle	8.22	5.65	0.69		Middle	14.10	9.54	0.68
	Bottom	10.70	7.23	0.68		Bottom	17.00	11.50	0.68
SUI-37	Top	9.38	6.58	0.70	SUI-104	Top	13.70	9.39	0.69
	Middle	12.00	8.25	0.69		Middle	17.30	11.70	0.68
	Bottom	12.80	8.52	0.66		Bottom	17.20	11.30	0.65
SUI-38	Top	7.35	5.02	0.68	SUO-5	Top	7.51	4.90	0.65
	Middle	11.70	8.12	0.69		Middle	11.40	7.57	0.67
	Bottom	14.60	9.87	0.68		Bottom	17.30	7.75	0.66

TABLE 12 (Continued)

Sample No.	Location ^(a)	Specific Activity ^(b) ($\mu\text{Ci}/\text{cm}^2$) ^(c)		Ratio Cs-137/ Cs-134	Sample No.	Location ^(a)	Specific Activity ^(b) ($\mu\text{Ci}/\text{cm}^2$) ^(c)		Ratio Cs-137/ Cs-134
		Cs-134 ^(d)	Cs-137				Cs-134 ^(d)	Cs-137	
SUO-6	Top	10.70	6.87	0.64	EVI-52	Top	5.01	3.80	0.76
	Middle	10.30	6.84	0.66		Middle	4.94	3.84	0.78
	Bottom	10.20	6.57	0.64		Bottom	5.17	4.03	0.78
SUO-7	Top	13.50	9.00	0.67	EVI-76	Top	4.57	3.55	0.78
	Middle	14.60	9.72	0.66		Middle	5.05	3.80	0.75
	Bottom	15.50	10.20	0.66		Bottom	4.41	3.43	0.78
SUO-8	Top	12.70	8.37	0.66	EVI-77	Top	--	--	--
	Middle	15.70	10.20	0.65		Middle	4.98	3.59	0.72
	Bottom	18.10	11.70	0.65		Bottom	4.68	3.68	0.79
SUO-15	Top	15.70	10.20	0.65	EVI-78	Top	--	--	--
	Middle	16.50	11.10	0.67		Middle	4.74	3.68	0.78
	Bottom	14.30	9.49	0.66		Bottom	4.61	3.59	0.78
SUO-16	Top	9.79	6.35	0.65	EVI-102	Top	4.75	3.73	0.78
	Middle	12.20	7.95	0.65		Middle	4.75	3.73	0.78
	Bottom	12.50	7.94	0.63		Bottom	4.72	3.63	0.77
SUO-20	Top	18.80	12.80	0.68	EVI-103	Top	--	--	--
	Middle	20.90	13.70	0.65		Middle	4.98	3.59	0.72
	Bottom	19.20	12.60	0.65		Bottom	4.68	3.68	0.79
SUO-22	Top	12.20	7.79	0.63	EVI-104	Top	4.38	3.50	0.80
	Middle	13.00	8.53	0.66		Middle	5.08	3.73	0.74
	Bottom	13.30	8.55	0.64		Bottom	4.87	3.61	0.74
SUO-23	Top	14.30	9.45	0.66	EVI-105	Top	4.59	3.59	0.78
	Middle	14.60	9.79	0.67		Middle	5.01	3.84	0.77
	Bottom	11.90	7.82	0.66		Bottom	4.95	3.72	0.75
SUO-36	Top	16.40	10.60	0.65	EVI-128	Top	3.51	2.68	0.76
	Middle	16.40	10.70	0.65		Middle	3.76	2.86	0.76
	Bottom	16.80	10.80	0.64		Bottom	3.88	2.98	0.77
EVI-26	Top	8.14	6.11	0.75	EVI-167	Top	4.57	3.43	0.75
	Middle	9.67	7.23	0.75		Middle	4.78	3.59	0.75
	Bottom	9.00	6.73	0.75		Bottom	4.48	3.78	0.75
EVI-51	Top	5.48	4.08	0.74	EVI-168	Top	4.56	3.31	0.74
	Middle	5.62	4.21	0.75		Middle	4.60	3.35	0.73
	Bottom	5.14	3.90	0.76		Bottom	4.42	3.27	0.74

TABLE 12 (Continued)

Sample No.	Location (a)	Specific Activity ^(b)		Ratio Cs-137/ Cs-134	Sample No.	Location (a)	Specific Activity ^(b)		Ratio Cs-137/ Cs-134
		(μCi/cm ²) (c)					(μCi/cm ²) (c)		
		Cs-134 (d)	Cs-137				Cs-134 (d)	Cs-137	
EVI-169	Top	4.54	3.29	0.73	ECI-116	Top	1.76	1.29	0.73
	Middle	4.62	3.40	0.73		Middle	2.45	1.91	0.78
	Bottom	4.36	3.17	0.73		Bottom	3.03	2.32	0.76
EVI-170	Top	4.03	3.05	0.76	ECI-117	Top	1.95	1.55	0.79
	Middle	4.02	3.08	0.77		Middle	2.91	2.42	0.77
	Bottom	3.82	2.85	0.75		Bottom	3.13	2.50	0.88
ECI-9	Top	1.63	1.61	0.99	ECI-118	Top	3.13	2.57	0.82
	Middle	2.38	1.50	0.63		Middle	2.75	2.79	0.83
	Bottom	1.66	1.34	0.81		Bottom	2.24	1.68	0.75
ECI-49	Top	--	--	--	ECI-119	Top	3.21	2.54	0.79
	Middle	3.13	2.19	0.70		Middle	2.99	2.39	0.80
	Bottom	2.73	2.16	0.79		Bottom	2.11	1.65	0.78
ECI-60	Top	3.13	2.48	0.79	ECI-120	Top	2.87	2.11	0.77
	Middle	3.63	2.84	0.78		Middle	2.67	2.03	0.76
	Bottom	5.50	4.27	0.78		Bottom	1.97	--	--
ECI-78	Top	4.57	3.58	0.78	ECI-121	Top	2.85	2.32	0.81
	Middle	4.30	3.33	0.77		Middle	2.74	2.14	0.78
	Bottom	3.54	2.66	0.75		Bottom	2.00	1.47	0.73
ECI-87	Top	--	--	--	ECI-122	Top	2.57	2.08	0.81
	Middle	3.14	2.53	0.81		Middle	2.57	2.08	0.81
	Bottom	2.69	2.19	0.81		Bottom	1.97	1.57	0.80
ECI-95	Top	3.28	2.51	0.76	ECO-9	Top	2.22	1.66	0.74
	Middle	3.15	2.45	0.78		Middle	2.57	1.87	0.73
	Bottom	2.60	1.93	0.74		Bottom	2.59	1.98	0.76
ECI-96	Top	3.83	3.06	0.80	ECO-49	Top	2.24	1.74	0.77
	Middle	3.65	2.88	0.79		Middle	2.73	2.01	0.73
	Bottom	--	--	--		Bottom	3.09	2.27	0.73
ECI-106	Top	3.52	2.77	0.79	ECO-115	Top	--	--	--
	Middle	2.97	2.35	0.79		Middle	--	--	--
	Bottom	--	--	--		Bottom	2.05	1.69	0.83
ECI-115	Top	1.85	1.55	0.84					
	Middle	2.56	2.11	0.82					
	Bottom	3.04	2.50	0.82					

(a) Top, middle, and bottom indicate an average of four counts circumferentially around the tube at the top, middle, and bottom of the tube sample. Top location corresponds to the top of the steam generator bundle.

(b) Decayed back to reactor shutdown on October 31, 1974.

(c) $1 \mu\text{Ci}/\text{cm}^2 = 370 \text{ Bq}/\text{mm}^2$.

(d) Average of 604 and 795 keV photopeaks.

Fig. 8. Cesium deposition profiles in the cold duct

Fig. 9. Cesium plateout distribution in primary circuit derived from gamma scanning data

shown in Fig. 9 is the average of tubes SUI-83, -89, -93, -101, -103, and -104 activities as given in Table 12. These tubes are, in effect, the first to see the hot core exit gas. Other data points given in Fig. 9 were determined in a like manner. (See Section 2.3.3 for further data comparison.)

Figure 9 shows excellent agreement with the Cs-137 data in Fig. 25 of Ref. 5 at an axial depth of 3.4 m into the steam generator. The only variation was the higher Cs-134 data found in the present study. The discrepancy observed for Cs-134 is not presently understood.

2.3.2. Results of Leach Experiments (Including Cesium Data)

As described in Section 2.2.2, the leach experiments were carried out in two phases. The first phase involved preliminary experiments performed to determine (1) the best method for the leach experiments, and (2) whether I-129 could be detected. Three steam generator tube samples and one trepan sample were utilized.

In these preliminary experiments, two different leach procedures were tried: (1) a warm 30-minute leach in 1N NaOH with NaOCl and iodine and strontium carriers, and (2) a warm 10-minute leach in 4N HNO₃ with hydrazine hydrate and strontium and iodine carriers. Results of these preliminary experiments showed that rather severe acid leaching conditions were required to remove all the activity from the samples. In many cases, it was necessary to remove appreciable amounts of the base metal in order to remove all the activity. Accordingly, as described in Section 2.2.2, a leach method was adopted that utilized 7-to-8 hours leaches with 4N HNO₃ and 3N HCl solutions with strontium carrier.

The second phase of the work involved use of the adopted leach method on the following samples: six superheater tube samples, two evaporator tube samples, four economizer tube samples, three trepan (duct) samples, and one trepan (SOLAMI) sample. The tube samples were cut from the as-received tube samples and were about 25 mm in length. The trepan and SOLAMI samples were used as-received.

It was found that the HNO_3 solution effectively removed the activity from the superheater tube samples, but use of the HCl solution was required to remove all the activity from the economizer and evaporator tube samples. This suggests that the activity is more easily removed from the superheater tubes than from the economizer and evaporator tubes. On the other hand, in the decontamination studies (Section 2.3.6), it was found that the activity was more easily removed from the economizer and evaporator tubes than from the superheater tubes. It appears, therefore, that the activity can be readily removed from any of the tubes with selection of the right agent.

As described in Section 2.2.2, all the leach solutions contained flakes of carbonaceous material, which were removed from the samples during the leach procedure. The leach solutions and carbonaceous residues were gamma counted separately. Cesium-134 and Cs-137 were the only gamma-emitting nuclides detected. In general, most of the cesium activity was associated with the carbonaceous layer (see Section 2.3.7).

Specific activities of Cs-134 and Cs-137 derived from gamma analyses of the leach solutions and residues are given in Table 13. Uncertainties in the data are estimated in Section 2.3.3.

Figure 10 is a plot of the cesium data derived from the leach solutions. Some of the data points represent an average of data for several tubes at a given radial depth into the steam generator bundle. The cesium deposition profile in Fig. 10 is similar to that in Fig. 9, which is a profile plot of the cesium gamma scan data.

Results of the stepwise leach experiment are given in Table 14. As indicated by the table and described in Section 2.2.2, the first part of the stepwise leach experiment was conducted utilizing 4N HNO_3 solution. After five leach treatments with the 4N HNO_3 solution and a total leach time of 410 minutes, most of the cesium activity still remained, indicating that the cesium nuclides had penetrated appreciably into the SOLAMI material. The cesium activity was completely removed by leaching with 3N HCl solution for approximately 48 hours.

TABLE 13
SPECIFIC ACTIVITIES OF Cs-134 AND Cs-137 DERIVED FROM
GAMMA ANALYSES OF LEACH SOLUTIONS^(a)

Sample ^(b)	Specific Activity ^(c) ($\mu\text{Ci}/\text{cm}^2$) ^(d)	
	Cs-134 ^(c)	Cs-137
SUI-5	5.12	3.80
SUI-36	4.07	2.97
SUI-83	8.38	6.21
SUI-101	10.50	7.85
SUO-5	4.58	3.17
SUO-36	8.70	6.23
EVI-51	4.58	3.96
EVI-105	4.46	3.77
ECI-9	2.05	1.91
ECI-119	3.29	2.98
ECO-9	2.93	2.54
ECO-115	3.30	2.92
L3-1	3.07	2.86
L6-4	2.09	2.06
L10-2	0.69	0.71
SIL2-2	0.36	0.26

(a) Including Cs activities in carbonaceous layer.

(b) The tube samples used for the leach experiments were 25 mm sections cut from the middle of the as-received tube samples. The trepan and SOLAMI samples were used as-received.

(c) Decayed back to reactor shutdown on October 31, 1974.

(d) $1 \mu\text{Ci}/\text{cm}^2 = 370 \text{ Bq}/\text{mm}^2$.

(e) Average of specific activity values derived from 604 and 795 keV peaks.

Fig. 10. Cesium plateout distribution in primary circuit derived from leach solution data

TABLE 14
RESULTS OF STEPWISE LEACH EXPERIMENT ON
SOLAMI SAMPLE SIL2-2(a)

Leach Solution	Leach Time (min)	Specific Activity Removed ($\mu\text{Ci}/\text{cm}^2$)	
		Cs-134	Cs-137
4N HNO_3	5	0.055	0.047
4N HNO_3	15	0.021	0.019
4N HNO_3	30	0.021	0.019
4N HNO_3	60	0.017	0.015
4N HNO_3	300	0.015	0.013
3N HCl	2900(b)	<u>0.23</u>	<u>0.15</u>
Total Removed		0.36	0.26

(a) Conducted at room temperature.

(b) Approximately 48 hours.

An experiment was conducted in which a 25-mm steam generator tube sample (from tube SUO-23) was fired prior to leaching. Strontium analyses were performed on the solution and residue; the results are given in Section 2.3.5.

The leach and decontamination test results did not yield convincing information about depth of fission product penetration into the surface of the metal components. The presence of the carbonaceous deposition layer was a complication. Nevertheless, the fact that strong chemical agents were required to remove activity from the steam generator tubes indicates that cesium penetrated to some depth into the tube surfaces.

2.3.3. Discussion and Evaluation of Cesium Plateout Data

As described above, cesium plateout data, in the form of specific activities ($\mu\text{Ci}/\text{cm}^2$), were obtained from (1) gamma scans of trepan and steam generator tube samples and (2) gamma analysis of leach solutions. In an earlier program, IRT obtained cesium plateout data from (1) gamma scans of the external surface of the primary coolant circuit ducting shortly after the final shutdown of the reactor, (2) gamma scans of the steam generator tubes from the secondary side, and (3) internal scans of two cold vertical ducts.

Representative cesium plateout data from these sources are compared in Table 15. The GA gamma scan data for the tube samples were taken from Table 12. Middle-of-tube specific activity values were selected from Table 12 because the 25-mm tube samples used in the leach experiments were cut from the middle of the as-received tube samples. The GA gamma scan data for the trepan samples were taken from Table 10. The GA leach data were taken from Table 13. The IRT gamma scan data were taken from Ref. 5.

The comparison in Table 15 shows that the cesium specific activity data and the Cs-137/Cs-134 ratio data from the three sources (GA gamma scan,

TABLE 15
COMPARISON OF CESIUM SPECIFIC ACTIVITY DATA

Sample No.	Cs-134 Specific Activity ^(a) ($\mu\text{Ci}/\text{cm}^2$) ^(b)			Cs-137 Specific Activity ^(a) ($\mu\text{Ci}/\text{cm}^2$) ^(b)			Ratio Cs-137/Cs-134		
	GA Gamma Scan ^(c)	GA Leach ^(d)	IRT Gamma Scan ^(e)	GA Gamma Scan	GA Leach	IRT Gamma Scan	GA Gamma Scan	GA Leach	IRT Gamma Scan
SUI-5	14.50	5.12	5.25	9.84	3.80	8.77	0.68	0.74	1.67
SUI-36	8.22	4.07	6.45	5.65	2.97	6.63	0.69	0.73	1.03
SUI-83	12.10	8.38	4.32	8.27	6.21	6.54	0.68	0.74	1.51
SUO-5	11.40	4.58	4.83	7.57	3.17	7.48	0.66	0.69	1.55
SUO-36	16.40	8.70	4.38	10.70	6.23	7.48	0.65	0.72	1.71
EVI-51	5.62	4.58	3.45	4.21	3.96	4.34	0.75	0.86	1.26
ECI-9	2.38	2.05	3.55	1.50	1.91	3.65	0.63	0.93	1.03
L3	2.66	3.07	2.62	2.21	2.86	2.22	0.83	0.93	0.85
L6	1.86	2.09	2.17	1.52	2.06	1.82	0.82	0.98	0.84

(a) Decayed back to reactor shutdown on October 31, 1974.

(b) $1 \mu\text{Ci}/\text{cm}^2 = 370 \text{ Bq}/\text{mm}^2$.

(c) The GA gamma scan data were taken from Tables 10 and 12 (see text).

(d) The GA leach data were taken from Table 13.

(e) The IRT gamma scan data were taken from Ref. 5.

GA leach, and IRT gamma scan) for the trepan locations (L3 and L6) are in good agreement. Also, as noted in Section 2.3.1 and shown in Fig. 8, good agreement was found for the GA gamma scan and IRT data for all the trepan locations in the cold duct.

In contrast to the good agreement found for the cold duct data, the steam generator tube data from the three sources show some discord. The three sets of data (in Table 15) for the evaporator and economizer (EVI and ECI) tube samples are in reasonable agreement; however, the Cs-137/Cs-134 ratio values for the IRT data seem out-of-line (see below). Considering the Cs-134 data for the superheater (SUI and SUO) tube samples, both the GA leach and the IRT data are low relative to the GA gamma scan data. Considering the Cs-137 data for the superheater tube samples, the GA gamma scan data and the IRT data are in reasonable agreement but the GA leach data are relatively low.

The Cs-137/Cs-134 ratios for the GA gamma scan and the GA leach data are in reasonable agreement. The ratios for both sets of data increase with downstream distance in the primary circuit. This increase is consistent with expected behavior. The downstream cesium is older, and the shorter-lived Cs-134 located downstream would have died out relative to the Cs-137.

The IRT Cs-137/Cs-134 ratios seem unreasonable for two reasons: (1) the values for the tube samples are much higher than those for the other two sets of data, and (2) the values decrease with downstream distance in the primary circuit. The method used by IRT to gamma scan the steam generator tubes involved inserting a detector into the secondary side of selected in-place steam generator tubes. Calibration was difficult, and the resulting gamma spectra were difficult to interpret, especially for Cs-137 because the Cs-134 and Cs-137 peaks overlapped.

The cesium specific activity data in Tables 10, 11, 12, and 13 show considerable scatter, indicating that the uncertainty of the data is rather large. It is clear that the systematic errors are larger than the random errors. The predominant systematic errors are believed to be loss of activity during cutting and shipping operations. It is known that all the tube samples had a carbonaceous layer on the helium side and that this layer was heavy, flaky, and subject to loss. Examination of the data in Table 6 shows that appreciable activity was lost from some of the samples to the container during shipping.

In view of these considerations, systematic and random errors were estimated at the 68% (1σ) confidence level. Of the estimated systematic errors, that for the cutting operation at the plant was conservatively taken to be the highest because there was little basis for this estimate. The estimated systematic errors for the GA gamma scan data for the steam generator tubes are +0.20 for the cutting operation at the plant and +0.15 for the shipping operation. The random error is estimated to be ± 0.15 for the counting operation. Combining these in accord with Ref. 21, one obtains -0.15 and $+\sqrt{(0.20)^2 + (0.15)^2 + (0.15)^2} = +0.29$. Thus, the error associated with a single point can have the range -0.15 to $+0.29$ at the 68% confidence level. This means, for example, that the real value of a data point of 10 would be in the range of $+8.5$ to $+12.9$ with 68% confidence. These estimated uncertainties apply to the GA specific activity data in Tables 12 and 15 for the steam generator tube samples.

The above estimated error of -0.15 to $+0.29$ seems reasonable on the basis of the following treatment of gamma scan data given in Table 12. The data of Table 12 can be divided into sets according to radial depth from the standpoint of helium flow into the steam generator bundle. For example, tubes SUI-5, -6, -15, -20, and -23 were at the same approximate radial depth. The middle Cs-134 specific activity values for these tube samples are 14.50, 14.40, 11.10, 14.10, and 12.00, respectively. The standard deviation for this set of data is 1.56 or 12% of the average value of 13.22. Standard deviations were calculated for several sets of data,

including data for ECI tubes; the average standard deviation was 13.2%, which is close to the above estimated error. The standard deviation value of 13.2% represents the random error for the gamma scan data and does not take into account systematic or constant errors in one direction.

Estimated systematic errors for the GA leach data for the steam generator tubes are +0.20 for the cutting operation at the plant, +0.15 for the shipping operation, +0.10 for the cutting operation at the laboratory to obtain the 25-mm tube samples, and +0.10 for losses during the leach operation. The random error is estimated to ± 0.20 . The combination estimated errors for a single data point, calculated as above, are +0.35 and -0.20 at the 68% confidence level. These uncertainties apply to the GA leach data given in Tables 13 and 15 for the steam generator tubes.

Estimated systematic errors at 68% confidence for the GA gamma scan data for the trepan samples are +0.20 for the cutting operation at the plant and +0.15 for the shipping operation. Random errors are estimated to be ± 0.15 . The combination estimated errors, calculated as above, are +0.29 and -0.15 at 68% confidence. These apply to the data in Tables 10 and 15.

Estimated systematic errors at 68% confidence for the GA leach data for the trepan samples are +0.20 for the cutting operation at the plant, +0.15 for the shipping operation, and +0.10 for losses during the leach operation. The estimated random error is ± 0.20 . The combination estimated errors, calculated as above, are +0.34 and -0.20 at 68% confidence. These apply to the data in Tables 13 and 15.

On the basis of the uncertainty estimates and all the above considerations relative to the three sets of cesium specific activity data, it is concluded that (1) for Cs-134 plateout in the steam generator tubes, the GA gamma scan data are the most reliable and accurate for use in code validation work, (2) for Cs-137 plateout in the steam generator tubes, the IRT and GA gamma scan data are equally reliable and accurate, and

(3) for Cs-134 and Cs-137 plateout in the duct, the three sets of data for the trepan samples are equally reliable and accurate.

2.3.4. Results of Iodine Analysis

Efforts to determine the amount of plated out iodine were confined to an activation analysis technique. Direct measurement of iodine by gamma counting was not possible due to the long delay between reactor shutdown and component examination.

As described in Section 2.2.3, 12 leach solutions from evaporator and economizer inlet tube samples and trepan location L-10 were analyzed for iodine. No I-129 was detectable in any of the 12 solutions analyzed. Upper limits to its possible concentration were established for each sample. These limits largely reflect the background activity present in each sample following irradiation and its chemical yield. Upper limits for the samples ranged from 2.2×10^{-5} μCi down to 2.1×10^{-6} μCi of I-129.

It appears that essentially no iodine was released from the core to the primary coolant circuit. The fuel element internal gas purge system was apparently very effective in controlling iodine release.

2.3.5. Results of Component Sample Beta Counting (Strontium Data)

As described in Sections 2.2.2 and 2.2.4, strontium analyses were performed on solutions and residues from leach experiments on 12 steam generator tube samples and 3 trepan samples. The resulting Sr-90 specific activity data are given in Table 16.

The Sr-90 data in Table 16 have a relatively large uncertainty because of (1) possible losses of activity during cutting, shipping, and leaching operations and (2) difficulties during the analysis caused by iron leached from the samples. Estimated errors at the 68% confidence level (1 σ) are included in Table 16.

TABLE 16
SPECIFIC ACTIVITY DATA FOR Sr-90 DERIVED FROM
RADIOCHEMICAL ANALYSIS OF LEACH SOLUTIONS

Sample No.	Specific Activity (a) ($\mu\text{Ci}/\text{cm}^2$) (b)	Estimated Error (c) (%)		
		Random Error (d)	Systematic Error (e)	Combined Error (f)
SUI-5	3.94×10^{-3}	± 4.9	+31	+31 -5
SUI-36	2.61×10^{-3}	+6.4	+31	+32 -6
SUI-83	4.60×10^{-3}	± 10.0	+31	+33 -10
SUI-101	7.56×10^{-3}	± 14.0	+31	+34 -14
SUO-23(g)	7.10×10^{-3}	± 7.0	+31	+32 -7
SUO-36	3.39×10^{-3}	± 66.0	+31	+73 -66
EVI-51	6.21×10^{-3}	± 9.0	+31	+32 -9
EVI-105	3.40×10^{-3}	± 27.0	+31	+41 -27
ECI-9	2.67×10^{-3}	± 8.0	+31	+32 -8
ECI-119	8.53×10^{-3}	± 13.0	+31	+34 -13
ECO-9	7.32×10^{-4}	± 24.0	+31	+39 -24
ECO-115	2.00×10^{-3}	± 73.0	+31	+79 -73
L3-1	6.15×10^{-4}	± 6.6	+29	+30 -7
L6-4	4.93×10^{-4}	± 7.3	+29	+30 -7
L10-2	8.36×10^{-4}	± 6.8	+29	+30 -7

(a) Decayed to reactor shutdown on October 31, 1974.

(b) $1 \mu\text{Ci}/\text{cm}^2 = 370 \text{ Bq}/\text{mm}^2$.

(c) Estimated at 68% confidence.

(d) Derived from standard deviations for beta counting and chemical yield (Ref. 18).

(e) Based on estimated errors for losses in cutting, shipping, and leach operations (see text).

(f) Square root of sum of squares of random and systematic errors.

(g) Derived from data in Table 15 for tube sample fired in air at 900°C prior to leaching.

The estimated random errors in Table 16 were derived from standard deviations for beta counting and chemical yield given in Ref. 18. Estimated systematic errors at the 68% confidence level for the steam generator tube samples are +0.20 for the cutting operation at the plant, +0.15 for the shipping operation, +0.10 for the cutting operation at the laboratory, and +0.15 for losses during the leaching operation. Combining these by taking the square root of the sum of the squares in accord with Ref. 21, one obtains the value +0.31. Estimated systematic errors at the 68% confidence level for the trepan samples are +0.20 for the cutting operation at the plant, +0.15 for the shipping operation, and +0.15 for losses during the leach operation. Combining these gives +0.29.

The combined error in Table 16 indicates the range associated with a single point. For example, the range of the Sr-90 specific activity for sample SUI-5 is 3.74×10^{-3} to 5.16×10^{-3} at the 68% confidence level.

Figure 11 is a plot of the Sr-90 specific activity data (from Table 16) as a function of swept surface area within the primary coolant loop. Some of the points in Fig. 11 represent averages of the data in Table 16 according to depth in the steam generator. For example, the first data point in Fig. 11 is the average of the values for tubes SUI-83 and -101.

The profile of the Sr-90 data in Fig. 11 is similar to that of the cesium data in Fig. 9. The activity level of Sr-90 is, however, orders of magnitude lower than that of cesium. There appears to be a general decrease in strontium activity across the steam generator. However, the strontium levels within the cold duct (samples L3-1, L6-4, and L10-2) were found to be relatively constant with no apparent concentration gradient (within the scatter in the data).

A section of the hot duct insulating material (SOLAMI) was sequentially leached in nitric acid for up to 7 hours, and the leach solutions were analyzed for strontium. The results, given in Table 17, indicate that

Fig. 11. Strontium plateout distribution in primary circuit

TABLE 17
STRONTIUM ACTIVITY REMOVED BY
SEQUENTIAL LEACHING OF SOLAMI SAMPLE

Sample	Sr-90 Specific Activity Removed (a) ($\mu\text{Ci}/\text{cm}^2$) (b)	Percent Removed
SIL2-2 (5 min)	9.58×10^{-4}	40
SIL2-2 (15 min)	3.25×10^{-4}	14
SIL2-2 (30 min)	3.82×10^{-4}	16
SIL2-2 (1 h)	3.72×10^{-4}	16
SIL2-2 (5 h)	<u>3.30×10^{-4}</u>	<u>14</u>
Total removed	2.37×10^{-3}	100

(a) Decayed back to reactor shutdown on October 31, 1974.

(b) $1 \mu\text{Ci}/\text{cm}^2 = 370 \text{ Bq}/\text{mm}^2$.

approximately 40% of the strontium activity was removed in the first 5 minutes and nearly 70% in the first 50 minutes. It took an additional 6 hours to remove the remaining 30% of activity.

Table 18 gives the results of an experiment in which an untreated tube section was fired before leaching in order to remove the carbonaceous layer. The tube sample was fired in air at 900°C for 15 minutes. The tube appearance changed from carbonaceous black to a dull rusty color as a result of the firing. As described in Ref. 18, the loose residue from the firing was treated as one sample for strontium analysis, the cold leaching solution and the hot leaching solution were treated as two separate samples, and the combined residue from both leachings was considered to be a fourth sample.

The results in Table 18 indicate that about 80% of the strontium activity was associated with the carbonaceous layer that burned off during the firing and that 6N HNO₃ slowly removed the remainder of the strontium activity.

2.3.6. Results of Decontamination Studies

Decontamination tests were conducted to determine the effectiveness of selected chemical agents for decontaminating the surfaces of the steam generator tube samples. As described in Section 2.2.5, the tests were performed on four samples.

Table 19 summarizes the data from the decontamination tests. This table gives (1) the amounts of Cs-134 and Cs-137 activity removed by each agent and (2) the percent removed by each agent. Table 20 compares the initial and removed specific activities for Cs-134 and Cs-137 and gives the percent remaining at the end of the decontamination procedure. Each initial specific activity value in Table 20 is the average of the specific activity values given in Table 12 for the as-received tube sample from which the decontamination sample was cut.

TABLE 18
RESULTS OF EXPERIMENT IN WHICH A TUBE SAMPLE (SUO-23) WAS
FIRED BEFORE LEACHING

Sample	Solution or Residue	Sr-90 Activity Removed (μCi)
Loose material after firing	Residue	1.06×10^{-1}
Cold 6N HNO_3 (16 h)	Solution	1.16×10^{-2}
Warm 6N HNO_3 (8 h)	Solution	1.05×10^{-2}
Combined cold and warm residues	Residue (a)	3.77×10^{-3}
Total Sr-90 activity removed		$1.32 \times 10^{-1} \text{ (b)}$

(a) Combined residues from leach solutions.

(b) The estimated error at 68% confidence for this value is +60%, -6%.

TABLE 19
RESULTS OF DECONTAMINATION TESTS

Sample No.	Solution	Specific Activity Removed (a)		Percent of Total Removed (d)
		(μCi/cm ²) (b)		
		Cs-134 (c)	Cs-137	
SUI-93	1,1,1-trichloroethane	1.02	0.80	32.3
	1N NaOH	0.25	0.19	7.8
	TURCO 4501-A	0.77	0.62	24.7
	TURCO 4502	1.03	0.79	32.4
	4N HNO ₃	0.04	0.03	1.2
	Detergent	<u>0.05</u>	<u>0.04</u>	<u>1.6</u>
	Total removed	3.16	2.47	100.0
SUO-7	1,1,1-trichloroethane	1.67	1.27	30.0
	1N NaOH	0.64	0.50	11.6
	TURCO 4501-A	1.43	1.09	25.7
	TURCO 4502	1.74	1.37	31.8
	4N HNO ₃	0.04	0.03	0.7
	Detergent	<u>0.01</u>	<u>0.01</u>	<u>0.2</u>
	Total removed	5.54	4.27	100.0
EVI-104	1,1,1-trichloroethane	0.63	0.52	13.2
	1N NaOH	1.44	1.22	30.6
	TURCO 4501-A	2.45	2.23	53.7
	TURCO 4502	0.06	0.05	1.3
	4H HNO ₃	0.03	0.03	0.7
	Detergent	<u>0.02</u>	<u>0.02</u>	<u>0.5</u>
	Total removed	4.63	4.07	100.0
ECI-96	1,1,1-trichloroethane	0.14	0.13	3.8
	1N NaOH	2.00	1.85	53.6
	TURCO 4501-A	1.55	1.40	41.1
	TURCO 4502	0.02	0.02	0.6
	4N HNO ₃	0.02	0.02	0.6
	Detergent	<u>0.01</u>	<u>0.01</u>	<u>0.3</u>
	Total removed	3.74	3.43	100.0

(a) Decayed back to reactor shutdown on October 31, 1974.

(b) 1 μCi/cm² = 370 Bq/mm².

(c) Average of 604 and 795 keV photopeaks.

(d) Average of Cs-134 and Cs-137 values.

TABLE 20
EFFECTIVENESS OF DECONTAMINATION PROCEDURE IN
REMOVING CESIUM ACTIVITY

Sample No.	Initial Specific Activity (a) ($\mu\text{Ci}/\text{cm}^2$)		Total Specific Activity Removed (b) ($\mu\text{Ci}/\text{cm}^2$)		Percent Remaining	
	Cs-134	Cs-137	Cs-134	Cs-137	Cs-134	Cs-137
SUI-93	15.00	10.30	3.16	2.47	79	76
SUO-7	14.50	9.64	5.54	4.27	62	56
EVI-104	4.77	3.61	4.63	4.07	None	None
ECI-96	3.74	2.97	3.74	3.43	None	None

(a) Derived from gamma scan data in Table 12.

(b) From Table 19.

The data in Table 20 indicate that the decontamination procedure was much more effective in removing all the cesium activity from the evaporator and economizer tube samples (EVI-104 and ECI-96) than from the superheater tube samples (SUI-93 and SUO-7). More than half the cesium activity remained on the superheater samples after the procedure, whereas essentially none of the activity remained on the evaporator and economizer samples. It is apparent, therefore, that the decontamination agents and conditions selected were much more effective on mild steel (the material of the evaporator and economizer samples) than on Alloy 800 (the material of the superheater tube samples).

For the superheater samples, the results in Table 19 suggest that 1,1,1-trichloroethane, TURCO 4501-A, and TURCO 4502 were the most effective in removing the cesium activity. However, the agent 1,1,1-trichloroethane was probably effective only because it was the first agent used; its action was probably of a mechanical nature in removing loose material.

For the evaporator and economizer samples (EVI-104 and ECI-96), the results in Table 19 indicate that TURCO 4501-A was effective in removing the cesium contamination. The agents 1,1,1-trichloroethane and 1N NaOH appear to have been somewhat effective, but these were the first agents used and their action was probably more mechanical than chemical. TURCO 4502 would probably have been effective; however, by the time this agent was applied, all the cesium activity was gone.

It should be noted that the carbon-rich surface films which were present on the outer (helium-side) surfaces of the samples may have reduced the effectivity of the decontamination agents.

As indicated above, the decontamination procedure was more effective on the economizer and evaporator tubes than on the superheater tubes. Superheater tubes were successfully decontaminated by use of an electrochemical procedure (see Ref. 8). In this procedure, between 0.076 and

0.152 mm (0.003 and 0.006 in.) of metal was removed. This procedure was less effective in decontaminating the carbon steel economizer and evaporator tubes. In leach studies (Section 2.3.2), it was found that a HNO_3 solution removed the activity from superheater tube samples and a HCl solution removed the activity from economizer and evaporator samples. It appears, as stated earlier in Section 2.3.2, that the activity can be readily removed from any of the tubes with selection of the right agent.

It is concluded that (1) the decontamination procedure selected was more effective on carbon steel of the economizer and evaporator tubes than on Alloy 800 of the superheater tubes, (2) the TURCO agents were the most effective of the agents tested, and (3) the activity can be readily removed from either the carbon steel or the Alloy 800 by selection of the right agent.

2.3.7. Results of Deposition (Carbonaceous) Layer Examination

The primary circuit component samples all contained a black carbonaceous-appearing deposit on the surface exposed to the helium coolant. This deposit was examined utilizing visual, microprobe, and chemical analysis techniques as described in Section 2.2.6.

A section of superheater tube SUI-16 was submitted for microprobe analysis to determine the composition and thickness of the surface deposit. The interface region between the deposit and base metal (Alloy 800) was clean and sharp with very little apparent interaction. The thickness of those deposits observed varied from 6 to 12 μm . Large regions of the deposited material were lost during polishing of the specimen due to the friable nature of the deposit. Several inclusions were observed within the base metal. The largest found was 30 to 35 μm deep and 12 to 15 μm wide. This particular inclusion was found to be composed of carbon (60% to 80%), iron (5% to 6%), chromium (3% to 10%), nickel (1% to 3%), and a trace of sulfur. A concentration gradient was detected for all constituents. The carbon was found to be highest in concentration near the

outer diameter of the tube, while the other elements were observed to increase with increasing penetration into the inclusion. The chromium increased to the base metal concentration of 23% (observed). The values reported above are relative due to the lack of appropriate standards for quantitative results.

The deposit contained carbon (80% to 100%), iron (2% to 3%), and traces of chromium and sulfur. None of the deposits or inclusions examined contained oxygen. An attempt was made, by use of the microprobe, to determine fission products (such as cesium, barium, and cerium) within the deposit. None were detectable. (Cesium activity as determined by gamma counting was below the limits of detection for the microprobe.) A piece of the deposit was analyzed by emission spectroscopy, which also showed iron (a few percent), nickel, and sulfur, thus verifying the microprobe results.

The deposit was easily removed in some areas as large flakes. Gamma analysis of the flakes consistently showed approximately 80% of the total gamma (cesium) activity associated with the tube specimen to be contained in the deposit.

2.4. SUMMARY AND CONCLUSIONS OF PRIMARY CIRCUIT COMPONENT RADIOCHEMICAL EXAMINATION

2.4.1. Cesium Plateout Data

Gamma scans of trepan and steam generator tube samples and gamma analyses of leach solutions for selected trepan and tube samples provided plateout data for Cs-134 and Cs-137 in the form of specific activities. Specific activities ranged from <1 to $\sim 12 \mu\text{Ci}/\text{cm}^2$ for Cs-137 and ~ 1 to $\sim 17 \mu\text{Ci}/\text{cm}^2$ for Cs-134. At a given location, Cs-134 activities were generally higher than Cs-137 activities. The two cesium isotopes were the only gamma emitters detectable in quantities sufficient to report.

A deposition layer of carbonaceous material was present on the helium side of all samples. This layer contained most of the cesium activity. The deposition layer was somewhat flaky and subject to loss during cutting and shipping operations. Consequently, the uncertainty of the cesium specific activity data is relatively high.

Plots of cesium activity as a function of swept surface area in the primary circuit show, as expected, a decrease in cesium activity with distance from the core outlet. The Cs-137/Cs-134 ratios increase with downstream distance in the primary circuit. This increase is as expected since the downstream cesium is older.

In an earlier program, IRT obtained cesium plateout data from (1) gamma scans of the external surface of the primary coolant circuit ducting shortly after the final shutdown of the reactor and (2) gamma scans of the steam generator tubes from the secondary side. Thus, there are three sets of cesium activity data: (1) GA gamma scan, (2) GA leach, and (3) IRT gamma scan.

The three sets of data for the trepan sample locations (L3 and L6) are in good agreement. The three sets of data for the evaporator and economizer tube samples are in reasonable agreement. On the other hand, the three sets of data for the superheater tube samples differ appreciably. Both the GA leach and the IRT gamma scan data for Cs-134 on the superheater tubes are low relative to the GA gamma scan data. The GA leach data for Cs-137 on the superheater tubes are low relative to the GA and IRT gamma scan data.

It is concluded that (1) for Cs-134 plateout in the steam generator tubes, the GA gamma scan data are the most reliable and accurate for use in code validation work, (2) for Cs-137 plateout in the steam generator tubes, the IRT and GA gamma scan data are equally reliable and accurate, and (3) for Cs-134 and Cs-137 plateout in the duct, the three sets of data for the trepan samples are equally reliable and accurate.

2.4.2. Iodine Analysis

In an attempt to determine iodine plateout levels, 12 leach solutions from evaporator and economizer tube samples were analyzed for iodine, utilizing an activation analysis technique. No iodine was detected, indicating that the core was effective in retaining iodine.

2.4.3. Strontium-90 Plateout Data

Strontium-90 specific activity data were obtained from radiochemical analyses of solutions and residues from 12 steam generator tube samples and 3 trepan samples. Strontium-90 specific activity levels were in the range of 5×10^{-4} to 9×10^{-3} $\mu\text{Ci}/\text{cm}^2$, several orders of magnitude lower than cesium activities. The Sr-90 data have a large uncertainty because of (1) possible losses of strontium activity during cutting, shipping, and leaching operations and (2) difficulties during the analyses caused by iron leached from the samples.

An experiment in which the carbonaceous layer was burned off in air prior to leaching showed that about 80% of the strontium activity was associated with the carbonaceous layer.

The plateout profile of Sr-90 in the primary circuit is similar to that of cesium. There appears to be a general decrease in strontium activity across the steam generator. However, the strontium levels within the cold duct show no apparent concentration gradient.

2.4.4. Decontamination Studies

Two superheater tube (Alloy 800) specimens, an evaporator tube (carbon steel) specimen, and an economizer tube (carbon steel) specimen were treated with various chemical agents including a base, an acid, and two proprietary formulations (TURCO 4501-A and TURCO 4502). The agents, in the form of solutions, were applied in turn to each specimen, and the

resulting solutions were analyzed to determine the amounts of Cs-134 and Cs-137 removed. Results indicate that (1) the agents tested are more effective in removing radioactive contamination from carbon steel than from Alloy 800 and (2) of the agents tested, the proprietary formulations were the most effective.

2.4.5. Deposition Layer Examination

The deposition (carbonaceous) layer on a superheater tube sample was examined using visual, microprobe, and chemical analysis techniques. The observed deposits contained carbon (80% to 100%), iron (2% to 3%), and traces of chromium and sulfur. The deposit had a friable nature and varied in thickness from 6 to 12 μm . Several inclusions were observed within the base metal (Alloy 800). The largest found was 30 to 35 μm deep and 12 to 15 μm wide and was composed of carbon (60% to 80%), iron (5% to 6%), chromium (3% to 10%), nickel (1% to 3%), and a trace of sulfur. The deposit was easily removed in some areas as large flakes. Gamma analysis of the flakes showed that approximately 80% of the total gamma (cesium) activity was associated with the deposit.

3. FISSION GAS RELEASE DATA FOR PEACH BOTTOM FUEL ELEMENT COMPACTS

This section provides fission gas release (R/B) data for Peach Bottom fuel element compacts measured utilizing the TRIGA King furnace facility at GA. The purpose of the measurements was to provide data for use in conjunction with reactor operation data for validating design methods for predicting the activity circulating in the primary circuit of HTGR systems.

3.1. METHOD FOR FISSION GAS RELEASE MEASUREMENTS

The fission gas release measurements were made using a standard method which involved (1) irradiation of the fuel compacts in a King furnace facility mounted in a TRIGA reactor (Refs. 22 and 23), (2) trapping the released fission gases, (3) gamma counting the collection trap, and (4) calculating the R/B data. The release value (R/B) is defined as the ratio of release rate to birth rate at steady state.

In the standard method, the specimen to be tested is placed inside a special crucible designed for insertion and retrieval from a TRIGA King furnace located in the pool of the GA TRIGA MK I reactor. The furnace, a resistance-heated graphite tube furnace capable of operating at temperatures up to 2000°C, is heated to a reference temperature (normally 1100°C) and the sample is irradiated for 30 minutes at a reactor power level sufficient to produce 10^{14} fissions in the specimen. Several furnaces are available in the facility; the largest can accommodate whole Peach Bottom fuel element compacts.

During irradiation and for 15 minutes afterward, helium purge gas is passed over the sample sweeping out all released fission gases into a liquid-nitrogen-cooled charcoal collection trap. The collection trap is

gamma counted to determine the total activity of the released fission gases, and R/B values are calculated from the gamma count data. Gamma counting and calculation of the R/B values are performed using a counting system in combination with a SIGMA II computer-analyzer system. The method provides R/B data for the noble gas isotopes Kr-85m, Kr-87, Kr-88, Kr-89, Xe-138, and Xe-139.

The fractional release (R/B) values are calculated by comparing the release of gaseous radionuclides from the fuel with the release from a powdered sample of uranium stearate, irradiated and gamma counted under the same conditions as the fuel sample. Uranium stearate has been shown to be 100% releasing for gaseous nuclides with half-lives >30 seconds (Ref. 24). This procedure eliminates the complex calculations and geometry calibrations normally encountered in R/B measurements. However, an absolute counting procedure using the latest values of branching ratios, decay schemes, half lives, neutron cross sections, and fission yields has shown excellent agreement with the stearate comparison method (Ref. 25).

Nuclides with half-lives >5 minutes do not attain their steady-state production values during the routine 30-minute TRIGA irradiation. Equilibrium R/B values can be determined using equations developed to correct the short-term irradiation data (Ref. 26). Recently, a more direct approach has been to subject samples to long-term (>100 hours) irradiation in the TRIGA reactor to obtain equilibrium conditions, thus resulting in a series of steady-state correction factors for the noble gas isotopes of interest (a factor of 1.9 for Kr-85m at 1100°C). The data reported herein for archive fuel compacts subjected to 30-minute irradiations have been corrected to equilibrium conditions using the experimentally derived factors. Analogous tests on irradiated fuel samples have shown virtually no change in activity with length of irradiation. Therefore, the R/B values reported for irradiated fuel compacts were not corrected for steady state (see Ref. 27).

3.2. RESULTS OF FISSION GAS RELEASE TESTS

Nine irradiated compacts and two archive compacts were tested at 1100°C in the TRIGA King furnace facility. The resulting data, given in Table 21, consist of R/B values for the noble gas isotopes Kr-85m, Kr-87, Kr-88, Kr-89, Xe-138, and Xe-139. Temperature dependence data were obtained for compact 22 from element F03-01, compact 14 from element F05-05, and one archive compact, as shown in Table 22. The temperature dependence data for Kr-85m are plotted in Fig. 12 for compact 22 from F03-01 and for the archive compact.

The average Kr-85m R/B value of 2.5×10^{-4} at 1100°C for the two archive compacts (Table 21) is in good accord with the beginning-of-life Kr-85m value of approximately 3×10^{-4} (Ref. 28) observed during initial operation of the Peach Bottom reactor. The Kr-85m R/B values for the irradiated compacts in Table 21 vary from 4.5×10^{-4} to 4.9×10^{-3} at the test temperature of 1100°C. The average value is 2.3×10^{-3} .

The end-of-life Kr-85m value for the fuel in the reactor was 2.1×10^{-4} at the calculated end-of-life effective temperature of 990°C (Ref. 29). This R/B value is the average of three R/B values (2.6×10^{-4} , 1.9×10^{-4} , and 1.7×10^{-4} at 61%, 61%, and 60% power, respectively) measured during final operation of the reactor.

Figure 12 shows that the end-of-life R/B value of 2.1×10^{-4} at 990°C is consistent with the temperature dependence data for the irradiated compact. A comparison of the temperature data for the two compacts with data for other HTGR fuel material (Ref. 27) shows that the data for the two compacts are typical.

The end-of-life R/B value at the reference temperature of 1100°C is 4×10^{-4} . This value was derived from the value 2.1×10^{-4} at 990°C, using the temperature dependence data of Fig. 12. The average Kr-85m value of 2.3×10^{-3} at 1100°C (from Table 21) for the irradiated compacts tested is nearly an order of magnitude higher than the end-of-life reactor value of

TABLE 21
RESULTS OF TRIGA R/B TESTS ON PEACH BOTTOM FUEL COMPACTS

Element No.	Compact No.	R/B at 1100°C(a)					
		Kr-85m	Kr-87	Kr-88	Kr-89	Xe-138	Xe-139
E01-01	3	8.8×10^{-4}	8.5×10^{-4}	6.9×10^{-4}	5.3×10^{-4}	--	--
	14	2.9×10^{-3}	3.2×10^{-3}	2.5×10^{-3}	2.1×10^{-3}	--	--
	23	2.0×10^{-3}	2.2×10^{-3}	1.8×10^{-3}	2.1×10^{-3}	--	--
F03-01	3	1.5×10^{-3}	1.8×10^{-3}	1.6×10^{-3}	1.9×10^{-3}	--	--
	12	4.8×10^{-3}	5.2×10^{-3}	3.9×10^{-3}	4.7×10^{-3}	--	--
	22	1.7×10^{-3}	1.8×10^{-3}	1.5×10^{-3}	1.8×10^{-3}	--	--
F05-05	3	4.0×10^{-3}	5.0×10^{-3}	4.5×10^{-3}	2.8×10^{-3}	--	--
	14	9.7×10^{-4}	7.9×10^{-4}	7.0×10^{-4}	6.9×10^{-4}	3.1×10^{-4}	4.7×10^{-4}
	23	1.3×10^{-3}	1.5×10^{-3}	1.5×10^{-3}	8.7×10^{-4}	--	--
Archive	1	2.5×10^{-4}	1.7×10^{-4}	1.8×10^{-4}	9.0×10^{-5}	--	--
	2	2.5×10^{-4}	1.9×10^{-4}	1.9×10^{-4}	1.1×10^{-4}	--	--

(a) The R/B values for xenon isotopes relative to the values for krypton isotopes are higher than expected based upon in-pile release behavior. The Kr-85m-to-Xe-138 ratio is therefore considerably smaller than that assumed in reference design calculations. The reason is presently unknown, but it is speculated that the hydrolysis of failed fuel particles might be responsible.

TABLE 22
EFFECT OF TEMPERATURE ON FISSION GAS RELEASE (R/B) FROM PEACH BOTTOM FUEL COMPACTS

Element No.	Compact No.	Temp (°C)	R/B					
			Kr-85m	Kr-87	Kr-88	Kr-89	Xe-138	Xe-139
F03-01	22	Room temp ^(a)	1.4×10^{-4}	1.4×10^{-4}	1.2×10^{-4}	9.1×10^{-5}	--	--
		400	1.9×10^{-4}	1.9×10^{-4}	1.6×10^{-4}	1.9×10^{-4}	--	--
		800	4.0×10^{-4}	4.1×10^{-4}	3.0×10^{-4}	3.9×10^{-4}	--	--
		1000	9.2×10^{-4}	8.8×10^{-4}	7.0×10^{-4}	7.3×10^{-4}	--	--
		1100	1.7×10^{-3}	1.8×10^{-3}	1.5×10^{-3}	1.8×10^{-3}	--	--
F05-05	14	Room temp	4.7×10^{-5}	4.4×10^{-5}	3.7×10^{-5}	2.1×10^{-5}	2.8×10^{-5}	3.1×10^{-5}
		800	1.5×10^{-4}	1.5×10^{-4}	1.3×10^{-4}	1.5×10^{-4}	8.0×10^{-5}	1.1×10^{-4}
		1000	6.1×10^{-4}	5.5×10^{-4}	4.7×10^{-4}	5.3×10^{-4}	2.7×10^{-4}	4.3×10^{-4}
		1100	9.7×10^{-4}	7.9×10^{-4}	7.0×10^{-4}	6.9×10^{-4}	3.1×10^{-4}	4.7×10^{-4}
Archive	1	Room temp	6.3×10^{-6}	4.9×10^{-6}	5.6×10^{-6}	1.8×10^{-6}	--	--
		400	1.9×10^{-5}	1.5×10^{-5}	1.5×10^{-5}	7.8×10^{-6}	--	--
		800	4.8×10^{-5}	3.5×10^{-5}	3.4×10^{-5}	2.0×10^{-5}	--	--
		1000	1.5×10^{-4}	9.7×10^{-5}	1.0×10^{-4}	5.5×10^{-5}	--	--
		1100	2.5×10^{-4}	1.7×10^{-4}	1.8×10^{-4}	9.0×10^{-5}	--	--

(a) Approximately 50°C.

Fig. 12. Temperature dependence of fission gas release for fuel compacts

4×10^{-4} . Thus, the R/B values for the irradiated compacts are much higher than one would expect on the basis of reactor data.

After completion of the R/B tests, compact 14 element E01-01 and compact 3 from element F03-01 were returned to ORNL for determination of failed fractions via the hot-chlorine-leach method (Ref. 30). In addition, compact 12 from element F03-01 was examined metallographically at GA. The resulting failed fraction values are given in Table 23. Included in Table 23 are fission gas release values for failed fuel $[(R/B)_f]$ calculated by dividing the Kr-85m R/B values in Table 21 by the failed fraction values in Table 23. The resulting $(R/B)_f$ values (Table 23) are considerably higher than one would expect on the basis of the $(R/B)_f$ value of 0.005, which is the reference value for constrained failed coated carbide particles (Ref. 27).

The metallographic examination of compact 12 from element F03-01 at GA revealed extensive hydrolysis of the failed particle fraction, as shown by the photomicrographs in Fig. 13. Observed failures are attributed to a pressure vessel type failure. This type of coating failure results in wide cracks, which would allow ready access of available moisture to the kernel (see Fig. 13). The extent of hydrolysis of individual compacts is difficult to assess; however, the fact that the failed particles were indeed hydrolyzed would account for the higher-than-expected R/B values for the irradiated compacts.

Let us now assume that the fuel in the Peach Bottom reactor at end-of-life was not hydrolyzed and use the following equation to calculate the average fuel failure:

$$(R/B)_{EOL} = F(R/B)_f + (R/B)_{am} \quad , \quad (3-1)$$

where $(R/B)_{EOL}$ = end-of-life R/B,

F = failed fraction,

$(R/B)_f$ = R/B of failed fuel,

$(R/B)_{am}$ = R/B of as-manufactured fuel.

TABLE 23
DETERMINATION OF FAILED PARTICLE FRACTION IN FUEL COMPACTS

Element No.	Compact No.	Particle Failure Fraction	$(R/B)_f^{(a)}$
E01-01	14	$\sim 0.015^{(b)}$	0.19
F03-01	3	0.055 ^(b)	0.027
F03-01	12	0.033 ^(c)	0.14
Average		0.034	0.12

(a) Fission gas release for failed fuel.

(b) Determined at ORNL by hot chlorine leach (Ref. 30).

(c) Determined at GA by metallographic examination.

Fig. 13. Photomicrographs of compact 12 from F03-01 showing the extent of hydrolysis of failed fuel particles

Using an $(R/B)_f$ of 5×10^{-3} for unhydrolyzed fuel (Ref. 27), an $(R/B)_{EOL}$ of 4×10^{-4} (see above), and an $(R/B)_{am}$ of 2.5×10^{-4} as determined for the two archive compacts (see Table 21), one calculates a failed fraction of 0.030. This value is good in accord with the average observed failed fraction value of 0.034 given in Table 23. This accord indicates that the fuel was not hydrolyzed in-core during reactor operation, but subsequent to reactor shutdown.

The $(R/B)_f$ values given in Table 23 are well within the range of R/B values (up to ~0.40) obtained in fuel hydrolysis tests, including a test on a Peach Bottom Core 1 fuel sample (Ref. 27). It is concluded that the irradiated compacts tested were partially but not necessarily fully hydrolyzed.

3.3. SUMMARY AND CONCLUSIONS OF FISSION GAS RELEASE DATA

Fission gas release (R/B) data at 1100°C for Kr-85m, Kr-87, Kr-88, Kr-89, Xe-138, and Xe-139 were obtained for nine irradiated fuel compacts and two archive compacts. Temperature dependence data were obtained for two irradiated compacts and one archive compact.

The R/B data for the irradiated compacts were higher than one would expect on the basis of the fraction of failed particles. The higher-than-expected R/B values were due to fuel hydrolysis that occurred after the fuel was removed from the reactor.

The average Kr-85m value of 2.5×10^{-4} at 1100°C for the two archive compacts agrees with the beginning-of-life Kr-85m R/B value of approximately 3×10^{-4} observed during initial operation of the Peach Bottom reactor. The effective fraction of failed fuel in the reactor at end-of-life was about 0.03, as estimated from the R/B data. This estimated failure fraction agrees with the average of failed fraction values determined at ORNL by use of a hot chlorine leach method and at GA by metallographic examination.

The temperature dependence data for the compacts are typical of data for HTGR fuel material in general.

4. SORPTION OF CESIUM ON GRAPHITE AND FUEL COMPACT MATRIX

A main objective of the PBEOL Program is to provide information for use in validating fission product transport codes, such as FIPER (Ref. 31) and TRAFIC (Ref. 32), by comparison of calculated and observed behavior of metallic fission products, particularly cesium, in the Peach Bottom HTGR (Ref. 2).

Input data for these codes includes sorption data for metallic fission products on fuel element graphite and fuel rod matrix. Empirical correlations of the sorption data in the form of equations are given in the fuel design data manual (Ref. 33). These data were derived primarily from sorption measurements on H-327 graphite material and fuel rod matrix material utilized in the FSV reactor and on reference graphite (H-451) material and fuel rod matrix material for the large HTGR.

To provide guidance for the application of these input data to calculations for the Peach Bottom HTGR, experiments have been conducted to compare the cesium sorptivity of Peach Bottom fuel element graphite (spine and sleeve) and fuel compact matrix with that of H-327 graphite, taken to be the reference graphite for the experiments. These experiments were conducted by H. G. Staley at GA.

4.1. METHOD FOR SORPTION STUDIES

A relative sorptivity method (described in Refs. 34 and 35) was utilized. In this method, samples of sorbent material are annealed for several hours at 1200°C (or other desired temperature) in sealed tantalum cells containing a reference cesium source. The source usually consists of an H-327 graphite sample impregnated with cesium tagged with Cs-137. The relative sorptivity of each sample with respect to the H-327 source

is deduced from the weights and cesium concentrations of the samples. The result is a sorption ratio, which is the ratio of cesium concentration in the sample to that in the graphite source. Graphite samples are normally in the form of powder with a particle size of 44 to 74 μm .

The samples and source (separated in the tantalum crucible) are annealed for 8 hours at a constant temperature (usually 1200°C). Following the anneal, the samples and source are gamma counted along with standards to determine the relative cesium loadings in each sample.

The source preparation entails adding a known weight of cesium nitrate (containing Cs-137) to distilled water and H-327 graphite powder to form a slurry which is heated to dryness. The source is gamma counted to determine the cesium loading.

4.2. RESULTS OF SORPTION STUDIES

The relative sorptivity data are given in Table 24. The graphite samples were pulverized and sieved in the usual manner to obtain the particle range of 44 to 74 μm . The cesium source graphite was 44 to 74 μm H-327. The fuel compact samples were in the form of chunks from unirradiated spare Peach Bottom fuel compacts. The Peach Bottom sleeve graphite is Great Lakes Carbon Company grade HLM-85. The spine graphite is of nuclear purity and low permeability. The matrix content of the fuel compacts was 70.2% (by weight). It should be noted that the cesium sorption behavior of H-327 graphite does not differ significantly from that of H-451 graphite (Ref. 35). The cesium sorptivity on 44 to 74 μm graphite powder is about 2.2 times greater than that on bulk graphite, and this factor appears to be independent of graphite type (Ref. 35). There is no significant difference between the sorptivity of powder and bulk matrix materials (Ref. 35).

TABLE 24
RELATIVE SORPTIVITY OF CESIUM ON H-327 GRAPHITE AND
PEACH BOTTOM FUEL ELEMENT MATERIALS AT 1200°C

Run	Sample(a)	Final Concentrations (mg Cs/g C)		Sorption Ratio $\left(\frac{\text{Sample Conc.}}{\text{Source Conc.}}\right)$
		H-327 Graphite Source	Sample	
SR-1	Spine graphite sample No. 1	0.68	0.51	0.75
SR-1	Spine graphite sample No. 2	0.68	0.50	0.74
SR-1	Sleeve graphite sample No. 1	0.68	0.88	1.29
SR-1	Sleeve graphite sample No. 2	0.68	0.89	1.31
SR-2	Sleeve graphite sample No. 3	1.81	3.40	1.90
SR-3	Peach Bottom type A fuel compact, chunk No. 1	0.58	3.19	5.50
SR-3	Peach Bottom type A fuel compact, chunk No. 2	0.58	3.25	5.60

(a) Samples were annealed with the source for 8 hours at 1200°C. All the graphite samples were in the form of 44 to 74 μm powder.

The results in Table 24 indicate that, compared with H-327 graphite, the spine graphite is slightly less sorptive and the sleeve graphite is slightly more sorptive. Average sorption ratios are 0.74 for the spine graphite and 1.5 for the sleeve graphite.

The results show that the fuel compact material, compared with H-327 graphite, is appreciably more sorptive of cesium. Taking the factor 2.2 (from above) into account, Peach Bottom fuel compact matrix material is about 12 times more sorptive than bulk H-327 graphite and about 8 times more sorptive than Peach Bottom sleeve graphite. All the cesium sorbed in the fuel compacts is assumed to be in the matrix.

It is of interest to compare the cesium sorptivity of Peach Bottom fuel compact material with fuel rod matrix material for FSV and the large HTGR. As given in Ref. 35, the sorption ratio for FSV production matrix compared with bulk H-327 graphite is 13.2 (at a cesium concentration of 0.48 mg cesium per gram graphite). Considering the uncertainties involved, this value of 13.2 is the same as the above value of 12 for the Peach Bottom matrix material. Reference fuel rod matrix material for the large HTGR is essentially the same as FSV matrix material (Ref. 35). It is concluded that the cesium sorptivity of the three matrix materials (Peach Bottom, FSV, and the large HTGR) are essentially the same.

4.3. SUMMARY AND CONCLUSIONS OF CESIUM SORPTION STUDIES

Experiments were conducted to compare the cesium sorptivity of Peach Bottom fuel element graphite (spine and sleeve) and fuel compact matrix with that of H-327 graphite. The latter was taken to be the reference graphite.

It is concluded from the results that at 1200°C, compared with H-327 graphite, spine graphite is slightly less sorptive and sleeve graphite is slightly more sorptive of cesium. Peach Bottom fuel compact matrix material is about 12 times more sorptive than bulk H-327 graphite and about 8 times

more sorptive than bulk Peach Bottom sleeve graphite. The cesium sorptivity of Peach Bottom fuel compact matrix material is about the same as fuel rod matrix material for FSV and the large HTGR.

5. DIFFUSION OF CESIUM IN FUEL ELEMENT GRAPHITE

This section provides diffusion coefficient data for cesium in Peach Bottom fuel element (spine and sleeve) graphite. These data are the results of experiments performed at GA by K. J. Mysels utilizing a permeation method.

Diffusion coefficient data for metallic fission products in fuel element graphite are important input data for fission product codes. A main objective of the PBEOL Program was to provide information for use in validating fission product transport codes by comparison of calculated and observed behavior of metallic fission products, particularly cesium, in the Peach Bottom HTGR. Diffusion coefficient data for cesium in Peach Bottom fuel element graphite were needed because the available diffusion coefficient data were for other graphites (primarily H-451 and H-327), and there was concern that the diffusion behavior of cesium in Peach Bottom fuel element graphite may be significantly different.

5.1. METHOD FOR CESIUM DIFFUSION STUDIES

In the permeation method, developed by Mysels and Birkett and described in Ref. 36, a source of tracer cesium and a good sink are separated by a leak-tight barrier of the graphite to be studied. This assembly is isolated against outside contamination. After annealing for a known time at a known temperature, the activity of the sink is a measure of the amount of cesium that has permeated through the barrier under these conditions. In addition, the activity of the barrier is a measure of its overall takeup of the cesium, and if desired, a profile can be obtained to show the distribution within the barrier. Typically, the source and sink are made of char-loaded (i.e., highly sorptive) graphite.

Figure 14 shows the parts of the assembly used in the permeation method. The rod is the source, the sleeve is the graphite diffusion barrier, and the skirt is the sink. Plugs and caps are used to prevent bypass of cesium around the diffusion barrier and to isolate the assembly. The furnace used is large enough to accommodate several assemblies.

After annealing under the desired conditions, the cesium activity of all parts was determined by gamma counting to verify the material balance. The activity of the sink measures the flux through the graphite and the activity of the sleeve and plugs measures the cesium take-up.

The permeation method yields a permeation coefficient, π , defined as follows:

$$J = \pi \frac{C_2 - C_1}{L} \quad , \quad (5-1)$$

where J is the steady-state flux per unit surface area and unit time, C_2 is the cesium concentration in the graphite at the graphite-source interface, C_1 is the cesium concentration in the graphite at the graphite-sink interface, and L is the thickness of the graphite. The permeation coefficient is analogous to a diffusion coefficient for steady-state transport.

C_1 is taken as equivalent to zero; C_2 is derived from the concentration of cesium in the source. Since the source is a different, highly sorptive graphite, its concentration is divided by an estimate of its higher sorptivity relative to the graphite, namely, a factor of 10, in order to obtain values for C_2 . The factor of 10 is estimated to be the equilibrium partition coefficient for cesium between the source and graphite materials. This partition coefficient may be uncertain by a factor of at most 2; the absolute permeation coefficients are correspondingly uncertain (see Ref. 37). In testing the Peach Bottom sample, this factor of 10 in sorptivity was assumed. Relative permeation coefficients are unaffected by this assumption.

Fig. 14. Parts of assembly for diffusion studies

The assumption that the sorptivity of the graphite is 10 times less than that of the source (char-loaded) graphite is necessary, because with respect to sorption, steady state takes a very long time (hundreds of hours) to establish, whereas with respect to permeation, steady state is reached very rapidly (within a couple of hours).

Mysels and Birkett found that an important source of variation in the transport behavior of cesium in graphite is the nature of the gaseous atmosphere present during annealing (Refs. 36 and 37). In the presence of carbon monoxide or impure helium, i.e., a slightly oxidizing atmosphere, transport was much higher than in the presence of hydrogen or under continuous evacuation. Hence, a sterile technique was adopted in which samples are handled and experiments are done in a reducing atmosphere of hydrogen in helium (see Ref. 37). The Peach Bottom samples were tested using this sterile technique.

In the experiments on the Peach Bottom samples, it was assumed that the initial rapid transport of cesium, the burst effect (see Ref. 38), is completely eliminated by the sterile technique. This may be over-optimistic as in some experiments a residual burst effect corresponding to some 20 minutes of steady-state transport was detected. This could make the results about 16% too high for a 2-hour anneal.

The standard deviation of a series of experiments using the same source and sample was around 6%, but this was based on a sufficiently long series to make possible the elimination of results which were completely out of line due to radioactive contamination.

Duplicate samples of spine and sleeve graphite were annealed at 865° and 1050°C for various time intervals to determine the cesium flux through the sample and the uptake of the sink material. The source concentration was in the range of 250 to 400 µg cesium per gram graphite and the radioactive tracer was Cs-134. Source, sleeve, and sink were gamma counted at the end of each anneal period. The same source and sleeve were used with

fresh sinks in several consecutive anneals to verify steady-state permeation rates.

5.2. RESULTS OF DIFFUSION STUDIES

The results are given in Table 25 for the spine graphite and in Table 26 for the sleeve graphite. Two spine samples and three sleeve samples were tested.

One of the two Peach Bottom spine samples gave very consistent results, leading to a permeability of $14 \times 10^{-12} \text{ m}^2/\text{s}$ and a time lag of slightly over 1 hour for cesium to penetrate the graphite (from source to sink). The other sample gave less consistent results, the permeabilities ranging between 8.0×10^{-12} and $16 \times 10^{-12} \text{ m}^2/\text{s}$ with an average of about $11 \times 10^{-12} \text{ m}^2/\text{s}$ and a time lag of about 1 hour. These results are all for 865°C . The average of all the permeation coefficient values for the spine samples is $12.4 \times 10^{-12} \text{ m}^2/\text{s}$.

The sleeve samples gave much lower permeabilities. Average values are $0.18 \times 10^{-12} \text{ m}^2/\text{s}$ at 865°C and $2.05 \times 10^{-12} \text{ m}^2/\text{s}$ at 1050°C . The change with temperature is about the same as for H-451 graphite, corresponding to an activation energy of 140 kJ/mol (33 kcal/mole) (Ref. 37). At 865°C , a time lag of about 1 hour was apparent.

Samples of H-451 graphite studied by the permeation method gave permeabilities ranging from 1.5×10^{-12} to $4.2 \times 10^{-12} \text{ m}^2/\text{s}$ (average of $3.0 \times 10^{-12} \text{ m}^2/\text{s}$) at 870°C and showed short time lags of between 1 and 2 hours (Ref. 37).

5.3. SUMMARY AND CONCLUSIONS OF DIFFUSION STUDIES

Diffusion coefficient data for cesium in Peach Bottom fuel element (spine and sleeve) graphite resulted from experiments performed by use of a permeation method. The results show that the spine graphite is about

TABLE 25
RESULTS OF CESIUM DIFFUSION EXPERIMENTS ON SPINE GRAPHITE

Anneal				Component Activity			Permeation Coefficient (10^{-12} m ² /s)
No.	Temp (°C)	Time (h)	Spine Sample No.	Source	Spine	Sink	
PB4	865	2	1	72.2	0.183	0.385	8.0
PB5	865	2	1	(70.0) (a)	ND (b)	0.747	16.0
PB6	865	2	1	68.2	0.387	0.573	12.6
PB8	865	8	1	(67.0)	0.594	1.720	9.6
PB3	865	2	2	(96.0)	ND	0.782	12.2
PB4	865	2	2	94.4	0.260	0.918	14.6
PB5	865	2	2	(94.5)	0.390	0.864	13.7

(a) Values in parentheses are interpolated from component activity data including data from earlier and later experiments.

(b) ND = not determined.

TABLE 26
RESULTS OF CESIUM DIFFUSION EXPERIMENTS ON SLEEVE GRAPHITE

Anneal				Component Activity			Permeation Coefficient (10^{-12} m ² /s)
No.	Temp (°C)	Time (h)	Sleeve Sample No.	Source	Sleeve	Sink	
PB5	865	2	1	(83.0) (a)	ND (b)	0.0103	0.18
PB7	865	6	1	(82.0)	ND	0.0314	0.19
PB8	865	8	1	(81.0)	ND	0.0573	(c)
PB9	1050	6	1	(80.0)	ND	0.359	2.25
PB10	1050	6	1	(78.5)	1.48	0.323	2.06
PB6	865	2	2	94.0	0.204	0.0091	0.145
PB8	865	8	2	(93.5)	ND	0.0507	0.203
PB9	1050	6	2	(92.5)	ND	0.371	2.01
PB10	1050	6	2	92.8	1.22	0.337	1.82
PB9	1050	6	3	(66.0)	0.525	0.275	2.10

(a) Values in parentheses are interpolated from component activity data including data from earlier and later experiments.

(b) ND = not determined.

(c) Omitted as too high due to assumed contamination.

three or four times more permeable to cesium than H-451 graphite, whereas the sleeve graphite is 10 to 20 times less permeable. Time lags for cesium to penetrate the spine and sleeve graphite are very short, on the order of 1 hour. Time lags for cesium in H-451 graphite are similar. The temperature coefficient for the diffusion of cesium in the sleeve graphite is very close to that for H-451 graphite.

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