

Los Alamos National Laboratory is operated by the University of California for the United States Department of Energy under contract W-7405-ENG-36

TITLE THE BEHAVIOR OF FISSION PRODUCTS DURING NUCLEAR ROCKET REACTOR TESTS

AUTHOR(S) Peter C. Bokor, William L. Kirk, and Richard J. Bohl

SUBMITTED TO 8th SYMPOSIUM ON SPACE NUCLEAR POWER SYSTEMS

NOV 13 1994

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

By acceptance of this article the publisher recognizes that the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution or to allow others to do so for U.S. Government purposes.

The Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy.

Los Alamos Los Alamos National Laboratory Los Alamos, New Mexico 87545



**LOG NUMBER: 000**

**"THE BEHAVIOR OF FISSION PRODUCTS DURING NUCLEAR ROCKET  
REACTOR TESTS"**

**AUTHORS: Peter C. Bokor, William L. Kirk, and Richard J. Bohl  
Los Alamos National Laboratory  
MS E550  
Los Alamos, NM 87545  
(505) 667-6394  
FTS 855-0492**

**CAMERA READY MANUSCRIPT prepared for:  
EIGHTH SYMPOSIUM ON SPACE NUCLEAR POWER SYSTEMS  
Albuquerque, New Mexico  
6-10 January 1991**

**SEND CORRESPONDENCE TO: William L. Kirk**

## THE BEHAVIOR OF FISSION PRODUCTS DURING NUCLEAR ROCKET REACTOR TESTS

Peter C. Bokor, William L. Kirk, and Richard J. Bohl  
Los Alamos National Laboratory  
MS E550  
Los Alamos, NM 87545

Fission product release from the fuel of a nuclear rocket reactor during testing and flight, either during normal operation or potential accidents, will be an important consideration for safety analysis, test facility design, and possibly mission architecture. One view is that the fission products (and other radioactive materials) must be contained in the reactor for all normal and most, if not all, accident conditions. Another, perhaps more realistic view is that complete fission product retention is impossible at the extremely high fuel temperature that is essential for attractive engine performance and that the upper range of potential nuclear rocket engine specific impulse can be achieved only with fuel in which there are no geometry or composition constraints imposed by fission product retention considerations.

In any case, the experience base regarding fission product behavior developed during the Rover program, the nuclear rocket development program of 1955-1972, will be useful in planning a renewed nuclear rocket program. During the Rover program, 20 reactors were tested at the Nuclear Rocket Development Station in Nevada. Nineteen of these discharged effluent directly into the atmosphere; the last reactor tested, a non-flight-prototypic, fuel-element-testing reactor called the Nuclear Furnace (NF-1) was connected to an effluent cleanup system that removed fission products before the hydrogen coolant (propellant) was discharged to the atmosphere. In general, we were able to increase both test duration and fuel temperature during the test series. Therefore fission product data from the later part of the program are more interesting and more applicable to future reactors. We have collected fission product retention (and release) data reported in both formal and informal publications for six of the later reactor tests; five of these were Los Alamos reactors that were firsts of a kind in configuration or operating conditions. We have also, with the cooperation of Westinghouse, included fission product data from the NRX-A6 reactor, the final member of series of developmental reactors with the same basic geometry, but with significant design and fabrication improvements as the series continued. Table 1 lists the six selected reactors and the test parameters for each.

**TABLE 1. Test Parameters for Selected Reactors.**

<u>REACTOR</u>	<u>TOTAL TIME AT FULL POWER (MIN)</u>	<u>CHARACTERISTIC FUEL EXIT GAS TEMPERATURE K</u>
Phoebus 1A	11	2400
Phoebus 1B	30	2350
Phoebus 2A	13	2200
Pewee 1	40	2550
NRX-A6	60	2450
NF-1	109	2450

Fuel element geometry was basically identical for all six reactors, slender hexagons containing 19 axial coolant passages. Each element was 19.05 mm across flats and 1.32 m long. The coolant passage diameter was approximately 2.54 mm. All surfaces were coated with a refractory carbide, at first NbC and later in the program ZrC, to reduce corrosion of the fuel matrix by hydrogen (see

Fig 1). The fuel element matrix material in all of these reactors except NF-1 was bead-loaded graphite; that is, 200- $\mu$ m-diam beads consisting of a  $UC_2$  core with pyrocarbon coating that were homogeneously dispersed in graphite. The beads were used to isolate the  $UC_2$  so that it would neither interfere with the fuel element surface coating chemistry nor interact with atmospheric water vapor during storage. However, the data show that the pyrocarbon coating of the beads also acted as a fission product diffusion barrier during reactor tests.

In an effort to improve fuel element lifetime and temperature capability, we developed a different fuel matrix consisting of a mixture of graphite with a solid solution of uranium carbide and zirconium carbide ( $UC-ZrC$ ). These "composite" elements were tested in NF-1 after several years of development. The uranium carbide is dispersed more uniformly in the matrix with no barrier to fission product migration other than fuel element surface coating. The two fuel element matrix types are illustrated in Fig. 2. A discussion of the fission product data for each selected reactor follows. Most of the information for Los Alamos reactors was obtained with the assistance of Ernest A. Bryant and A. Edward Norris, who were principal investigators of the radiochemistry of nuclear rocket reactor fuel. They graciously searched their personal and organizational files to provide reference material. The NRX-A6 material was furnished by Robert Holman of Westinghouse. Tables 2-14 are reproduced directly from materials furnished by these individuals.

### **PHOEBUS 1A**

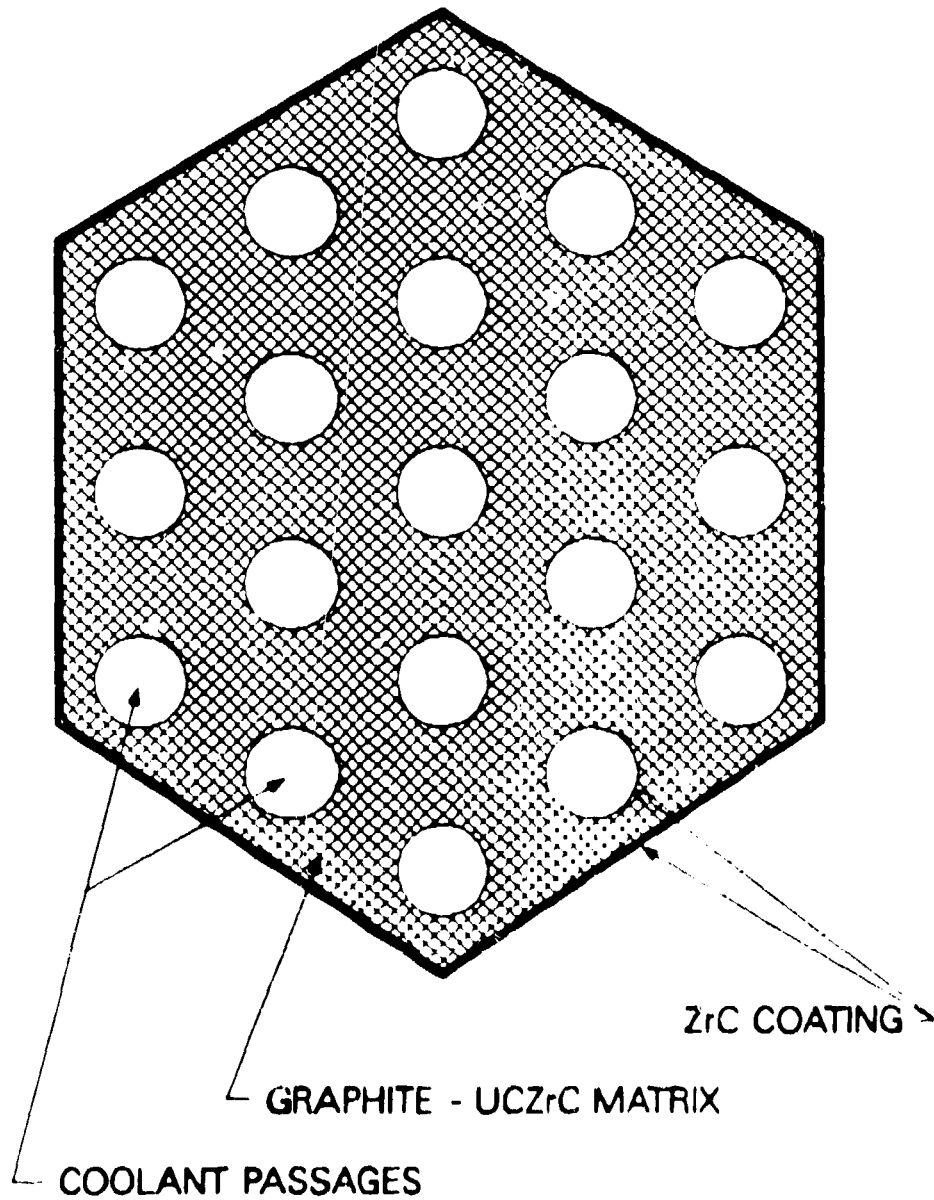
Although there was a loss-of-coolant accident which caused severe damage to the Phoebus-1A core at the end of the full-power test, the accident did not result in a large release of fission products from the fuel elements that remained in the reactor. Fission product retention data were obtained by post-test radiochemical analysis of fuel elements taken from the core of the reactor. Those fuel elements which were sampled lost an average of about 1.4% of the mass-140 fission product chain (measured as Ba-La-140) during the entire test and cool-down period. The loss of gross fission products, as measured with an ion chamber, was about 1% or less.<sup>2</sup>

Release data for the Phoebus 1A full power test were obtained from the analysis of air samples collected by two RB57C aircraft. These samples, obtained during and immediately after the reactor test, indicated that about 1.0% of the gross-fission product inventory was released to the atmosphere during the test. The fractions of individual fission products released varied widely. For example  $^{95}Zr$ ,  $^{97}Zr$ ,  $^{99}Mo$ ,  $^{147}Nd$  measured releases were less than 0.01% and  $^{111}Ag$ ,  $^{121}Sn$ , and  $^{125}Sn$  releases were greater than 10%.<sup>3,4</sup> The results are summarized in Table 2.

**TABLE 2. Phoebus 1A Fission-Product Losses by Analyses of Cloud Samples**

Fission Product	Amount Lost (%)	Fission Product	Amount Lost (%)
Sr89	3.5	I131	1.76
Y91	3.8	Te132	1.93
Y93	2.6	I133	1.65
Zr95	<0.01	I135	0.55
Zr97	<0.01	Ba140	0.25
Mu99	<0.01	Ce143	0.17
Ag111	31.6	Ce144	0.17
Cd115	30.0	Nd147	<0.01
Sn121	26.8	Eu156	1.6
Sn125	11.9	I1235	<0.01
Sb127	8.5		

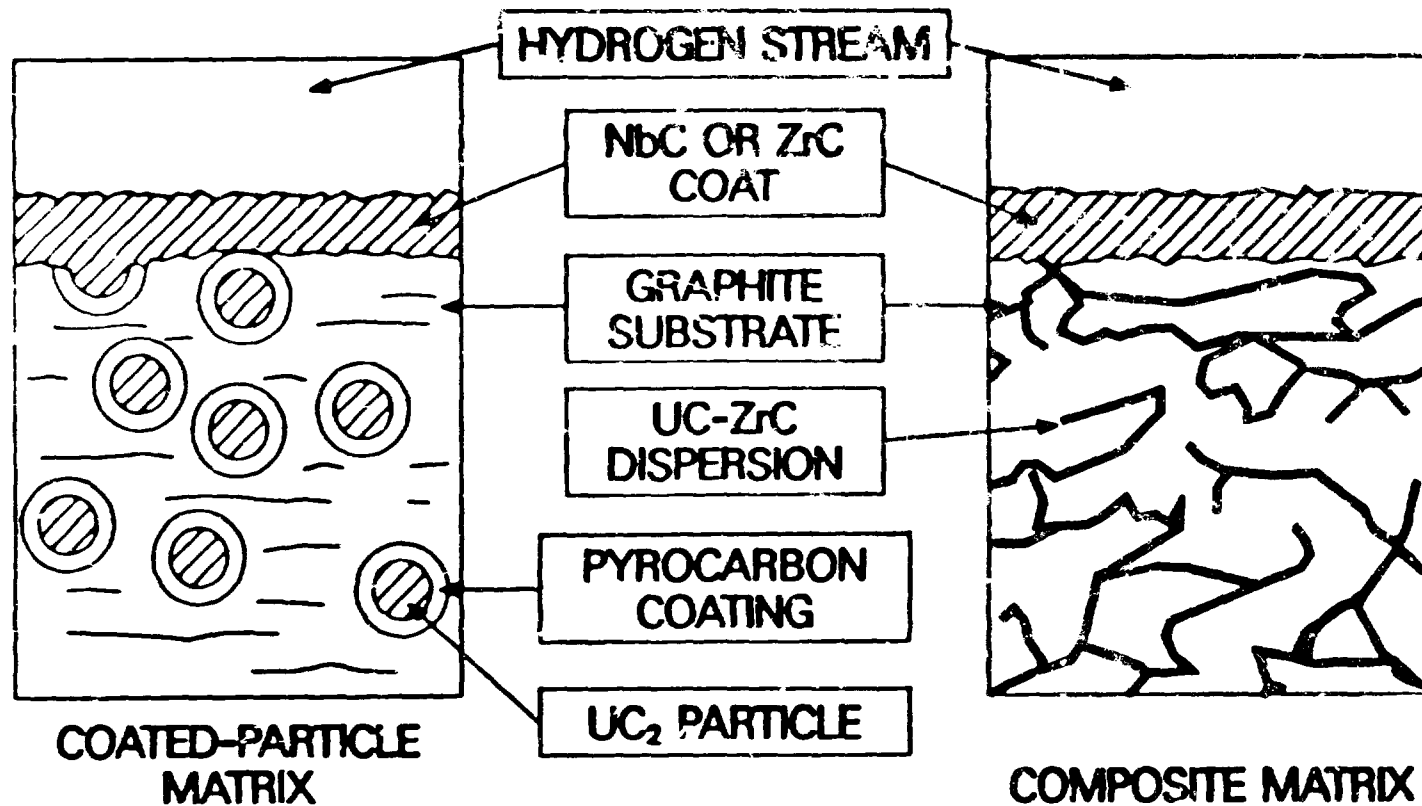
# 19-HOLE FUEL ELEMENT END VIEW



**Los Alamos**

Figure 1.

# FUEL STRUCTURE COMPARISONS



Comparison of the fuel structure in the standard, coated-particle, graphite matrix with the composite matrix fuel. The continuous, webbed UC-ZrC dispersion prevents hydrogen, entering through cracks in the top coating, from eating deeply into the graphite matrix.

Figure 2.

## PHOEBUS 1B

Phoebus 1B testing included a program of radiochemical measurements of the effluent gas during intermediate and full power experiments, and of the residual core material following reactor disassembly. Both the fuel element and the effluent sampling data showed that the release of fission products from the uranium fuel to the atmosphere was limited to about 0.5%. The fission product retention data for Phoebus 1B fuel was obtained by gamma-ray spectrum and radiochemical analysis of fuel elements from the core after the reactor test. The fission product retention values for  $^{140}\text{Ba}$  and  $^{95}\text{Zr}$  shown in Table 3 were derived from gamma-ray spectrum analysis. These retention values are relative to non-diffusing  $^{103}\text{Ru}$  and are average values taken from Stations 44.75 and 50.0. (stations are defined as the length in inches measured from the coolant inlet end of the fuel element. For example the point 25 inches from the coolant inlet end is called "Station 25.") The uncertainties of individual values are about one percent. Average values for the integral retentions of entire fuel elements are 99.8% for  $^{140}\text{Ba}$  and 99.9% for  $^{95}\text{Zr}$  for all nineteen fuel elements listed in Table 3.<sup>5,6</sup>

**TABLE 3. Data from Analysis of Phoebus-1B Fuel Elements by Gamma-ray Spectroscopy**

Reactor(a) Location	Radius in Core(in)	Fission Product Retention	
		$^{140}\text{Ba}$ at Sta. 45-50	$^{95}\text{Zr}$ at Sta. 45-50
30-A1-G	1.31	0.976	0.993
30-C3-F	5.69	0.982	0.999
270-C1-F	5.89	0.999	1.001(b)
330-D3-F	7.27	0.986	0.992
210-D1-B	7.43	0.981	0.988
210-E5-F	9.63	0.987	1.003
150-E1-F	9.86	1.003	1.007
90-F3-E	11.26	0.979	0.993
330-F5-E	11.26	1.013(a)	1.011
150-G4-E	12.86	0.992	1.001
330-G6-E	13.21	0.988	0.997
30-H5-D	14.33	0.997	1.007
150-H3-M	15.30	0.986	0.997
270-H3-M	15.30	1.031(a)	1.011
90-H2-W	16.31	0.980	0.996
210-H2-W	16.31	0.997	0.998
330-H2-W	16.31	1.000	1.002
150-H1-H	16.43	1.014	1.018
330-H1-H	16.43	0.980	0.995

(a) The fuel element location code includes a sector and radius description. This column is included here only to show the variety of fuel elements sampled.

(b) Changes in the gamma-ray spectra due to loss of mass by corrosion cause apparent retention values of greater than 1.00.

Fission product retention values for a number of other nuclides were obtained by performing radiochemical analyses on a few fuel elements from the reactor core. In each case, the amount of the nuclide was compared to the amount of  $^{103}\text{Ru}$  in the sample in order to obtain the fraction retained. Table 4 lists the retention values of various isotopes obtained by radiochemical analysis.

The uncertainties associated with the individual values are about 5%. The losses of  $^{89}\text{Sr}$ ,  $^{91}\text{Y}$ , and  $^{140}\text{Ba}$  are too small to be detectable. The overall loss of  $^{115\text{m}}\text{Cd}$  and also  $^{115}\text{Cd}$  (which does not appear in Table 4) are about 40%, which is in good agreement with the results obtained by the analyses of the exhaust gas.<sup>6</sup>

**TABLE 4. Fission Product Retention: Radiochemical Analysis of Phoebus-1B Fuel**

Reactor(a) Location	Station	Fission Product Retention*				
		$^{89}\text{Sr}$	$^{91}\text{Y}$	$^{140}\text{Ba}$	$^{115\text{m}}\text{Cd}$	$^{111}\text{Ag}$
30-A1-G	2.75	1.00	.99	1.02	1.02	.99
	8.0	1.00	.99	1.01	1.01	.98
	18.50	1.00	.97	1.01	1.03	.99
	23.75	.99	.95	1.01	.74	.87
	29.0	.99	.96	1.01	.41	.74
	34.25	.94	.98	1.01	.24	.67
	39.5	.98	.97	1.02	.20	.63
	44.75	.97	.93	1.01	.19	.62
	50.0	.99	.96	.99	.20	.62
90-F3-E	18.5	1.01	.99	---	1.07	---
	23.75	.99	.98	1.00	.79	---
	29.0	.98	.96	.98	.50	---
330-H1-H	8.0	1.02	1.02	1.02	1.02	---
	13.25	1.05	1.00	1.03	1.05	---
	18.5	.98	.97	.98	.98	---
	23.75	1.00	1.01	1.03	.81	---
	29.0	1.00	1.00	1.00	.52	---
	34.25	.99	.99	1.01	.26	---
50.0	.98	.99	.98	.20	---	

(a) See footnote a, Table 3.

\*Retention values are based on the ratio of the activity of the measured nuclide to the activity of  $^{103}\text{Ru}$ . See footnote b, Table 3.

Although measurement of fission product retention in the fuel elements was a more accurate and reliable way to measure the release of radioactive isotopes, aircraft sampling of the atmosphere in the vicinity of the test site and sampling of the nozzle exhaust gas was also performed. The aircraft sampling of airborne radioactive materials was conducted using two RB57C aircraft flying at altitudes of 1000 to 8000 feet.<sup>7</sup> The nozzle sampling system used a test probe that was introduced into the effluent stream for brief intervals to collect effluent samples in canisters for later analysis.<sup>8</sup>

Based on all three methods of measurement, it can be concluded that of the eighteen fission products measured, there were only five whose loss was clearly greater than one percent. These five were  $^{115}\text{Cd}$ ,  $^{115\text{m}}\text{Cd}$ ,  $^{111}\text{Ag}$ ,  $^{125}\text{Sn}$ , and  $^{127}\text{Sb}$ , all of which were expected to be lost at relatively low temperatures.<sup>9</sup> The results of the measurements from the aircraft sampling of the atmosphere and the sampling of the reactor exhaust gases in the nozzle are shown in Table 5. There is generally good agreement between the two sets of data.<sup>6</sup>



**TABLE 5. Values of fission-product release obtained from nozzle sample data and aircraft-collected samples of Phoebus-1B effluent.<sup>7,8</sup>**

Isotope	Nozzle Percent Release	Apparent Percent Release		Aircraft Average Value
		Sample from Aircraft 851	Sample from Aircraft 832	
<sup>89</sup> Sr	0.7	0.91	1.47	1.2
<sup>91</sup> Y	0.3	0.37	0.88	0.63
<sup>93</sup> Y	---	0.19	0.74	0.47
<sup>95</sup> Zr	0.08	0.16	0.73	0.45
<sup>97</sup> Zr	---	0.15	0.67	0.41
<sup>99</sup> Mo	0.10	0.11	0.35	0.23
<sup>111</sup> Ag	11.0	21.0	21.0	21.0
<sup>115</sup> Cd	38.0	( 40% by assumption )		
<sup>115m</sup> Cd	43.0	45.0	42.0	43.0
<sup>125</sup> Sn	9.0	9.9	8.1	9.0
<sup>127</sup> Sb	---	5.4	4.7	5.1
<sup>131</sup> I	>0.8	0.86*	1.05*	0.96*
<sup>132</sup> Te	0.4	0.74	1.02	0.88
<sup>133</sup> I*	---	0.41	0.57	0.49
<sup>135</sup> I*	0.2	0.8	0.09	0.09
<sup>140</sup> Ba	0.25	0.26	0.74	0.50
<sup>144</sup> Ce	0.09	0.07	0.51	0.29
<sup>147</sup> Nd	---	0.08	0.57	0.33
<sup>156</sup> Eu	---	0.20	0.87	0.54
<sup>235</sup> U	0.09	0.14	0.47	0.31

\* By gamma-spectrum analysis.

### **PHOEBUS 2A**

Phoebus 2A was designed to operate at 5000 MW, but unexpectedly high pressure vessel temperatures resulted in power being limited to 4200 MW. During the Phoebus 2A test series EP-IV and EP-V, the nozzle sampling system known as the elephant gun, was again used to analyze the fission product loss from the fuel elements. Five samples were taken during EP-IV, and five were planned for EP-V. As a result of an incorrect test canister assembly, only two samples were obtained from the EP-V test. All the presently recovered data regarding the fission product behavior for Phoebus 2A, which are based on the analysis of the nozzle samples, are summarized in Table 6.<sup>9</sup>

**TABLE 6. Release Fractions for Various Fission Products Measured during the Full-Power Tests of Phoebus-2A.**

Isotope Measured	Release fraction at:						
	21390*	EP-IV(June 26, 1968)			EP-VB(July 17, 1968)		
	21390*	21580	21760	21960	22053	28750	28934
<sup>115</sup> Cd**	.40	.40	.40	.40	.40	.029	.21
<sup>89</sup> Sr	.0018	.0038	.0035	.0085	.0060	.0015	.004
<sup>91</sup> Y	.0009	.0014	.0020	.0053	.0016	.00033	.0014
<sup>95</sup> Zr	.0001	.0002	.0001	.0013	.0002	---	---
<sup>111</sup> Ag	.124	.132	.178	.273	.216	.026	.24
<sup>131</sup> I	.0007	.0010	.0012	.0026	.0008	.00025	.00086
<sup>132</sup> Te	.0024	.0025	.0035	.0075	.0018	.000029	.0025
<sup>140</sup> Ba		.0017	.0017	.0060	.00017	.00043	.00087

\* Time in seconds where 0 seconds is at 6 am on run day.

\*\* <sup>115</sup>Cd values for EP-IV are assumed to be 0.40; loss of H<sub>2</sub> precluded direct calculation in all but the second EP-IV sample for which a value of 0.36 may be calculated; for EP-V <sup>115</sup>Cd values are based on amount of H<sub>2</sub> in each sample.

### PEWEE 1

The next reactor that was built and tested was Pewee 1. Although considerable work concerning fission product retention was done, we have not yet located the reports containing the detailed information. Samples of the radioactive effluent from Pewee 1 were collected from the exhaust plume just above the nozzle at five times during the full-power test of the reactor. The fractional release rates of isotopes <sup>89</sup>Sr and <sup>132</sup>Te are comparable to the rates observed during the test of Phoebus 1B. Integral losses from the reactor were a factor of 5-10 lower than had been predicted for the reported operating conditions of 2550 K fuel exit temperature for 40 minutes. This suggests that the integrity of the fuel cladding in Pewee 1, which acts as a barrier to the escape of most fission products, was significantly improved relative to past reactors like Phoebus 1B. Some of the overall release values were 0.8% for <sup>89</sup>Sr and <sup>132</sup>Te, 0.3% for <sup>95</sup>Zr, <sup>99</sup>Mo, <sup>140</sup>Ba, and <sup>141</sup>Ce and 3.6% for <sup>127</sup>Sb.<sup>11</sup> Off-site radioactivity measurements for Pewee 1 are reported in Reference 12.

### NRX-A6

As a part of the Rover/NERVA project, Westinghouse Astronautics Laboratory tested the NRX-A6 reactor for a total time of approximately one hour at full power. This test provided good data regarding fission product release. Measurements of 50 fuel element slices indicated a total gross gamma inventory release of 0.83%. The release fractions from the reactor were determined by radiochemical analysis of the fuel elements using <sup>137</sup>Cs as a reference nuclide. The fractional release of each nuclide was calculated at each designated location in the core, and then summed to give the total release from the core.<sup>13</sup> The predicted releases for the NRX-A6 reactor were calculated by a computer code that was based on the value of the NbC diffusion constant. Table 7 provides a comparison between measured and predicted fission product releases from NRX-A6 fuel elements, and Table 8 gives a summary of the predicted releases of iodine mass chains 131 and 135.

**TABLE 7. Comparison of Measured and Predicted Percent Release of Selected Fission Products from NRX-A6 Fuel Elements.**

<u>Nuclide</u>	<u>Percent Release</u>		<u>Predicted Percent</u>
	<u>Measured</u>	<u>Upper Limit</u>	
89Sr	2.96	4.6	4.5
91Y	1.67	3.6	5.6
95Zr	0.34	3.1	0.18
111Ag	33.85	57.5	35.1
115mCd	51.59	52.9	53.5
132mSn	20.13	22.3	0.10
131I	0.64	5.2	2.7
137Cs	0.00	0.00	2.9
140Ba	0.96	2.9	3.5
141Ce	1.56	7.2	1.1
144Ce	1.29	3.1	0.08
Gross Gamma	0.83	3.2	2.2

**TABLE 8. Predicted Percentage Release of Iodine Activities of the Mass Chains 131 to 135 from NRX-A6.**

<u>Mass Chain</u>	<u>Percentage Release</u>
131	2.7
132	2.8
133	2.8
134	3.0
135	2.9

In addition to the Westinghouse fission retention measures, information regarding the release of radioactive isotopes from the NRX-A6 reactor test can be found in reports of the U. S. Environmental Protection Agency.<sup>14,15</sup> These reports are based on data obtained from fission product measurements of samples of air, water, milk, and vegetation in the area surrounding the test site.

### **NF-1**

As noted earlier, an effluent cleanup system was used with NF-1. As the hot hydrogen left the reactor, it was mixed with water to reduce its temperature as it entered the Effluent Cleanup System (ECS). The H<sub>2</sub>-water mixture continued through a series of heat exchangers and gas-liquid separators. The liquid water that was removed by the separators was directed to a waste-disposal system. The hydrogen continued through a silica-gel dryer and heat exchanger into a cryogenic charcoal bed, where practically all the remaining radioactive materials were removed, and was finally exhausted to the atmosphere.<sup>16</sup>

During the NF-1 test series, experiments were performed to evaluate the ability of the low-temperature charcoal trap in the ECS to remove radioactive rare gases from the effluent. The results of these experiments were: 1) when operated at the proper temperature, the charcoal trap removed both radiokrypton and radioxenon from the gas stream; although 2) the dynamic adsorption coefficients for krypton on activated charcoal as measured in these experiments were less than those reported in the literature for krypton adsorbed from helium; 3) the number of  $^{89}\text{Kr}$  atoms entering the charcoal trap as measured by the experiments agreed with the value derived by other methods.<sup>16</sup>

Fission product retention values for NF-1 were obtained by measuring the activity due to the fission product of interest relative to a nondiffusing mass chain. For the NF-1 reactor the mass-95 chain was used as the nondiffusing standard. The mass-131 and -140 chains were measured with gamma-ray spectroscopy techniques before the fuel element wafers were dissolved. The results are shown in Table 9 which lists mass-131 and -140 retentions as a function of axial location. After the wafers were dissolved, the mass-89, -91, and -140 chains were measured by radiochemical analysis. These results are shown in Table 10.<sup>16</sup> Reference 1 contains tabulations of calculated and measured retention values for the NF-1 runs. Predicted retention of ten high-yield mass chains for the planned NF-2 runs (which were not done due to program termination) showed that only 57% of the inventory would be retained.<sup>18</sup>

**TABLE 9.  $^{131}\text{I}$  and  $^{140}\text{Ba}$  Fractional Retentions by Gamma-Ray Analysis.**

Station(b)	<u>Fractional Retention</u>			
	(a)Element 62537, Location D38		Element 93442, Location B11	
	$^{131}\text{I}$	$^{140}\text{Ba}$	$^{131}\text{I}$	$^{140}\text{Ba}$
3.25	---	---	---	---
10.25	1.001	1.003	0.987	0.999
28.25	0.602	0.887	0.569	0.729
29.25	---	---	0.594	0.726
41.75	0.294	0.683	0.466	0.835
51.25	0.167	0.799	0.418	0.630

(a) The elements that were analyzed are identified for serial number and location in the reactor.  
 (b) Values denote inches from the coolant inlet end of the fuel element.

**TABLE 10.  $^{89}\text{Sr}$ ,  $^{91}\text{Y}$ , and  $^{140}\text{Ba}$  Fractional Retentions by Radiochemical Analysis**

Element Location	<u>Station 51.25</u>		
	$^{89}\text{Sr}$ Fractional Retention	$^{140}\text{Ba}$ Fractional Retention	$^{91}\text{Y}$ Fractional Retention
D38	0.289	0.728	--
B11	0.444	0.622	0.636
C25	0.361	0.721	--

As part of the study of fission product retention in composite fuel, non-reactor, pre-test experiments were conducted with the fuel elements. Studies indicate that the actual release of fission products during a reactor test occur in several steps. The rates at which these steps occur are a function of the temperature, the gas pressure, the chemical nature of the fuel (e.g., the uranium loading), the fuel element coating, and other unidentified factors. Thus it is not possible

to predict in detail which fuel element will retain what fission product best. Therefore, the composition of the fuel elements used for pre-test experiments was varied in order to obtain a greater range of fission product release data. The effect of variations in composition is illustrated in Table 11. It is noteworthy that the low-carbide materials release  $^{131}\text{I}$ ,  $^{133}\text{I}$ , and  $^{132}\text{Te}$  more rapidly while the high carbide materials release  $^{143}\text{Ce}$  and  $^{147}\text{Nd}$  more rapidly.<sup>19</sup>

**TABLE 11. Fractional Retention of Fission Products in Several Composite Fuel Materials Following High-Temperature Anneals.**

Isotope Measure	<u>Fraction Retained in Composite Fuel Material</u>						
	<u>Material (vol. % carbide/uranium content (mg/cc))</u>						
	<u>20/70</u>	<u>20/151</u>	<u>30/70</u>	<u>30/136</u>	<u>30/151</u>	<u>35/151</u>	<u>40/77</u>
	<u>Heated at 1972 K for 1000 min or 1500 min</u>						
$^{89,91}\text{Sr}$			.72		.75		.56
$^{127}\text{Sb}$			.89				.79
$^{132}\text{Te}$			.77		.70		.97
$^{131,133}\text{I}$			.95		.63		.67
$^{133,135}\text{I}$			.86		.82		.83
Xe							
$^{140}\text{Ba}$			.83				.59
	<u>Heated at 2300 K for 50 min</u>						
$^{89,91}\text{Sr}$	.38	.62	< .72	.48	> .47	.58	.63
$^{125}\text{Sn}$	.66		< .81		> .68	.90	
$^{127}\text{Sb}$	.38	.68	.29	.44	.62	.95	.64
$^{132}\text{Te}$	.47	.22	.80	.90	> .86	.90	.92
$^{131,133}\text{I}$	.26	.31	.40	.38	> .36	.51	.53
$^{133,135}\text{I}$	.55	.62	.46	.43	> .40	.47	.47
Xe							
$^{140}\text{Ba}$	.47	.76	.86	< .78	.51	> .52	.63
$^{141,143}\text{Ba}$	.97	.95		.64		.71	.77
$^{144}\text{Ce}$							
$^{147}\text{Nd}$	.99	1.01		.61		.68	.77
	<u>Heated at 2605 K for 50 min</u>						
$^{89,91}\text{Sr}$		.26					.18
$^{125}\text{Sn}$							.31
$^{127}\text{Sb}$							.30
$^{132}\text{Te}$		.04	.16				.65
$^{131,133}\text{I}$		.08	.09				.17
$^{133,135}\text{I}$		.15	.07				.08
Xe							
$^{146}\text{Ba}$							.17
$^{143}\text{Ce}$							.54
$^{147}\text{Nd}$							.50

In addition to the study of the effect of composition, there were pretest experiments that varied the axial temperature profile, the coating of the fuel elements, and the gas environment. Fission product retention for mass chains 89, 131, and 140 have been calculated on the basis of power, temperature, and time. Table 12 summarizes the fractional retention of these three isotopes, tabulated according to approximate maximum fuel element temperature and corresponding axial location.<sup>18</sup>

**TABLE 12. Calculated Retention of Selected Fission Products as a Function of Fuel Element Temperature**

Fuel Element Temperature, K	Station, Inches	Fractional Retention		
		<sup>89</sup> Sr	<sup>131</sup> I	<sup>140</sup> Ba
2444	36.2-52.0	0.187	0.482	0.743
2333	34.9-36.2	0.366	0.630	0.795
2222	31.8-34.9	0.580	0.783	0.840
2111	29.2-31.8	0.762	0.897	0.879
2000	26.8-29.2	0.882	0.959	0.910
1889	24.7-26.8	0.948	0.985	0.936
1778	22.7-24.7	0.980	0.995	0.957
1611	18.9-22.7	0.996	0.998	0.978
1389	0.0-18.9	1.000	0.999	0.993

The effect of ZrC coating on the composite fuel elements was analyzed by mechanically removing the coating from all surfaces of several fuel elements and comparing the release of the fission products from coated and uncoated fuel elements. As expected, there was a generally enhanced release from the uncoated material compared to the coated material. Unexpectedly the release of Ce and Nd was lower from the uncoated material. Table 13 summarizes the effect of ZrC coating on fission product release from composite fuel.<sup>19</sup>

**TABLE 13. Fractional Retention of Fission Products in ZrC-Coated and Uncoated "Composite" Fuel Materials. Note the particularly large effects of coating on Sb, Te, and I.**

Temperature (K)	Heating Time (min)	Fuel Material		Clad	Fractional Retention of							
		Carbide (vol %)	Uranium (mg/cc)		Sr	Sb	Te	I	Xe	Ba	Ce	Nd
1872	800(1500)	40	77	Yes	.88	.78	.88	.77	.83	.88	.88	.71
1872	800	40	77	No	.82	.87	.78	.77	.81	.88	.88	1.0
2300	80	20	78	Yes	.38	.38	.48	.21	.82	.47	.88	.88
2300	80	20	78	No	.28	.85	.88	.88	.38	.38	1.0	-
2300	80	40	77	Yes	.83	-	.82	.88	.47	.84	.77	.77
2300	80	40	77	No	.88	.11	.84	.11	.28	.88	.88	.88
2605	80	20	151	Yes	.26	-	.88	.18	.14	-	-	-
2605	80	20	78	No	.88	.48	.82	.82	.82	.84	.71	-
2605	80	40	77	Yes	.18	.38	.88	.14	.88	.17	.84	.81
2605	80	40	77	No	.42	.81	.82	.88	.84	.88	.87	.81

Lastly, fuel elements were tested both in 1 atmosphere of helium and in vacuum. The presence of gas in the pores of the composite material and in the space around the material may be expected to appreciably affect the fractional release of fission products. The effect of pretest heating of fuel elements in a vacuum compared to heating in one atmosphere of helium was measured using a half inch slice of 20 vol % carbide material at 2300 K. The results are summarized in Table 14. A very definite effect was noted for Ce isotopes, while only small effects were found for other isotopes (the difference in time at temperature must be taken into account in comparing the fractions retained for the two samples).<sup>19</sup>

**TABLE 14. Effect of Heating in Vacuum as Opposed to Heating in Helium on Retention of Fission Products in Composite Fuel Material.**

Temperature (K)	Heating		Fractional Retention of:						
	Time (min.)	Atm.	Sr	Sb	Te	I	Xe	Ba	Ce
2300	30	Vac.	.31	<.05	.07	.11	.44	.33	.86
2300	50	He	.28	.05	.06	.08	.36	.39	1.0
1994	30	Vac	.32	.65	.93	.87	.90	.45	.99
1972	500*	He	.55	.57	.78	.77	.77	.66	.99

\*The fourth sample was 40 vol% carbide; the first three were 20 vol % carbide.

Comparing the NF-1 and laboratory data for composite fuel fission product release with the results obtained for bead-loaded graphite in the preceding five reactors, a much larger fission product loss from composite fuel is noticed. It seems clear that the pyrocarbon coating of the beads was a very effective barrier to fission product diffusion for the time and temperature of the reactor tests, while the composite fuel material is less effective. However, this conclusion must be tempered with a realization that the total duration of the NF-1 tests was nearly a factor of two greater than NRX-A6 and much longer than the tests of earlier bead-loaded reactors.

On the other hand, the composite fuel is clearly superior in corrosion resistance and its enhanced time-temperature capability compared to bead-loaded graphite fuel will provide extremely important improvements in mission performance. Therefore, we must plan to conduct both ground testing and flight operations of nuclear rocket engines so that safe and successful operation can be achieved even though an appreciable fraction of fission products leave the fuel.

With respect to testing, the successful operation of the NF-1 effluent cleanup system<sup>17</sup> provides a model with direct application to fuel element demonstration testing, which can be done economically in a nuclear furnace or similar reactor and indirect application to larger scale cleanup systems for flight prototype testing. Preliminary studies were done during the Rover program of effluent cleanup systems for Pewee and NERVA size systems. Rover Progress Reports such as LA-4779-MS, LA-4854-PR, LA-4922-PR, LA-4997-PR, LA-5054-PR, and LA-5133-PR briefly describe the design and development work on a Pewee effluent system, while the NERVA study, by the Aerotec Division of Universal Oil Products, is mentioned in LA-3962, LA-4012-MS and LA-4071-MS.

Obviously, considerable work will be necessary to design the best facility configuration for ground testing of nuclear rocket engines, but the Rover test program will provide a very strong foundation for this effort just as it does in many other areas of nuclear rocket technology.

## References

- Daniel R. Koenig, "Experience Gained from the Space Nuclear Rocket Program (Rover)," *Los Alamos National Laboratory report LA-10062-H*, (May 1986).
- E. A. Bryant, J. E. Sattizahn, and G. F. Grisham, "Radiochemical Results on Phoebus-1A," *Los Alamos Scientific Laboratory office memorandum J-11*, (November 1, 1965).
- J. E. Sattizahn, "Fission Product Losses during Full-Power Run of Phoebus-1A," *Los Alamos Scientific Laboratory office memorandum J-11*, (July 23, 1965).
- P. R. Guthals, "Phoebus-1A Air Sampling," *Los Alamos Scientific Laboratory office memorandum J-11*, (June 30, 1965).
- E. A. Bryant, "Radiochemical Analysis of Phoebus-1B Fuel Elements," *Los Alamos Scientific Laboratory office memorandum J-11*, July 7, 1967.
- E. A. Bryant, G. F. Grisham, and A. E. Norris, "Chapter 6, Phoebus-1B Post Mortem Report," *Los Alamos Scientific Laboratory office memorandum J-11*, (October 19, 1967).
- E. A. Bryant, "Fission Product Release from Phoebus-1B," *Los Alamos Scientific Laboratory office memorandum J-11*, (April 14, 1967).
- E. A. Bryant, "Release of Fission Products by Phoebus-1B," *Los Alamos Scientific Laboratory office memorandum J-11*, (April 25, 1967).
- Quarterly Status Report of the LASL Rover Program for period ending August 31, 1968. LA-4012-MS. (Distributed October 2, 1968.)
- N Division Personnel, "Pewee 1 Reactor Test Report," *Los Alamos Scientific Laboratory report LA-4217-MS*, (August 29, 1969).
- "Pewee 1 Postmortem Radiochemistry," *Los Alamos Scientific Laboratory office memorandum J-11*, (March 1, 1969).
- Environmental Surveillance Western Environmental Research Laboratory, "Final Report of December 4, 1968," *U.S. Environmental Protection Agency memorandum No. SWRHL-87 R*, (September 1971).
- W. A. Henninger, C. T. Rymer, and J. Roesmer, "Radiochemical Analysis of NRX-A6," *Westinghouse Astronuclear Laboratory report WANI-THE-1810*, (June 1968).
- Environmental Surveillance National Environmental Research Center, "Particulate Effluent Study NRX-A6, EP-111A - December 15, 1967," *U.S. Environmental Protection Agency report NERC-LV-539-21*. (March 1973).
- Environmental Surveillance Western Environmental Research Laboratory, "Final Report of Off-Site Surveillance for the NRX-A6 Reactor Test Series," *U.S. Environmental Protection Agency report SWRHL-78r*, (August 1971).
- W. L. Kirk, "Nuclear Furnace-1 Test Report," *Los Alamos Scientific Laboratory informal report LA-5189-MS*, (March 1973).



J. E. Sattizahn, "Quarterly Rover Progress Report - September 1, 1972 through November 30, 1972," *Los Alamos Scientific Laboratory office memorandum CNC-11*, (December 4, 1972).

CNC-11 memo "Calculated Release of Selected Fission Products from NF-2," (November 27, 1972).

J. W. Barnes, E. A. Bryant, G. F. Grisham, and A. E. Norris, "Release of Fission Products from 'Composite' Rover Fuel," *Los Alamos Scientific Laboratory office memorandum J-11*, (July 6, 1970)

K. Boyer, C. Fenstermacher, E. A. Bryant, and K. Meier, "Scale Model Tests of a Nuclear Rocket Exhaust Scrubber," *Los Alamos Scientific Laboratory AIAA paper No. 68-603*, (June 1968).