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FISSION-GAS-RELEASE RATES FROM IRRADIATED URANIUM NITRIDE SPECIMENS

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16. Abstract

Fission-gas-release rates from two 93 percent dense UN specimens were measured using a sweep gas facility. Specimen burnup rates averaged 4.5×10^{-3} and 3.2×10^{-3} percent/hr, and the specimen temperatures ranged from 425 to 1323 K and from 552 to 1502 K, respectively. Burnups up to 7.8 percent were achieved. Fission-gas-release rates first decreased then increased with burnup. Extensive interconnected intergranular porosity formed in the specimen operated at over 1500 K. Release rate variation with both burnup and temperature agreed with previous irradiation test results.

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SUMMARY

Fission-gas-release rates from two 93 percent dense uranium nitride (UN) specimens irradiated at high burnup rates were measured using a sweep gas facility. Specimen burnups (atom-percent fissions) reached 7.8 percent, and specimen temperatures ranged from 425 to 1502 K. The two specimen burnup rates averaged 4.5×10^{-3} and 3.2×10^{-3} percent per hour, respectively.

Fission-gas-release rates were found to first decrease, then increase with burnup. The minimum release rates occurred between 1 and 1.5 percent burnup. Postirradiation photomicrographs of a specimen operated at high temperature (>1500 K) shows large amounts of interconnected intergranular porosity. Release rate variation with both burnup and temperature agreed with previous irradiation test results.

INTRODUCTION

A program to develop methods to predict fission-gas-release rates from uranium nitride (UN) fuel pins was carried out at the NASA Lewis Research Center. The technique used was to measure fission-gas-release rates from UN specimens with a sweep gas facility, correlate the measured release rates with the test variables (temperature, fission-rate density, and burnup) through a theoretical fission gas transport release model, and then extend the model to include actual UN fuel pins through the addition of fuel/clad interaction effects.

This program, along with most of the NASA nuclear development programs, was terminated in January 1973. Fortunately, we have been able to achieve our main program goal even though it was not possible to complete all planned irradiations and postirradiation evaluations. The correlation of the data presented herein is included in reference 1.

A complete description of the sweep gas facility is contained in reference 2. The results of the first three UN specimen irradiations, a description of data analysis procedures, and the development of a preliminary gas release model are given in reference 3. The development of a gas release model which includes fuel/clad interaction effects and the correlation of this model with stable gas release data is presented in reference 1.

The main purpose of this report is to document the fission-gas-release rate data obtained too late for complete inclusion in reference 3.

TEST APPARATUS

A complete description of the sweep gas facility has been given in references 2

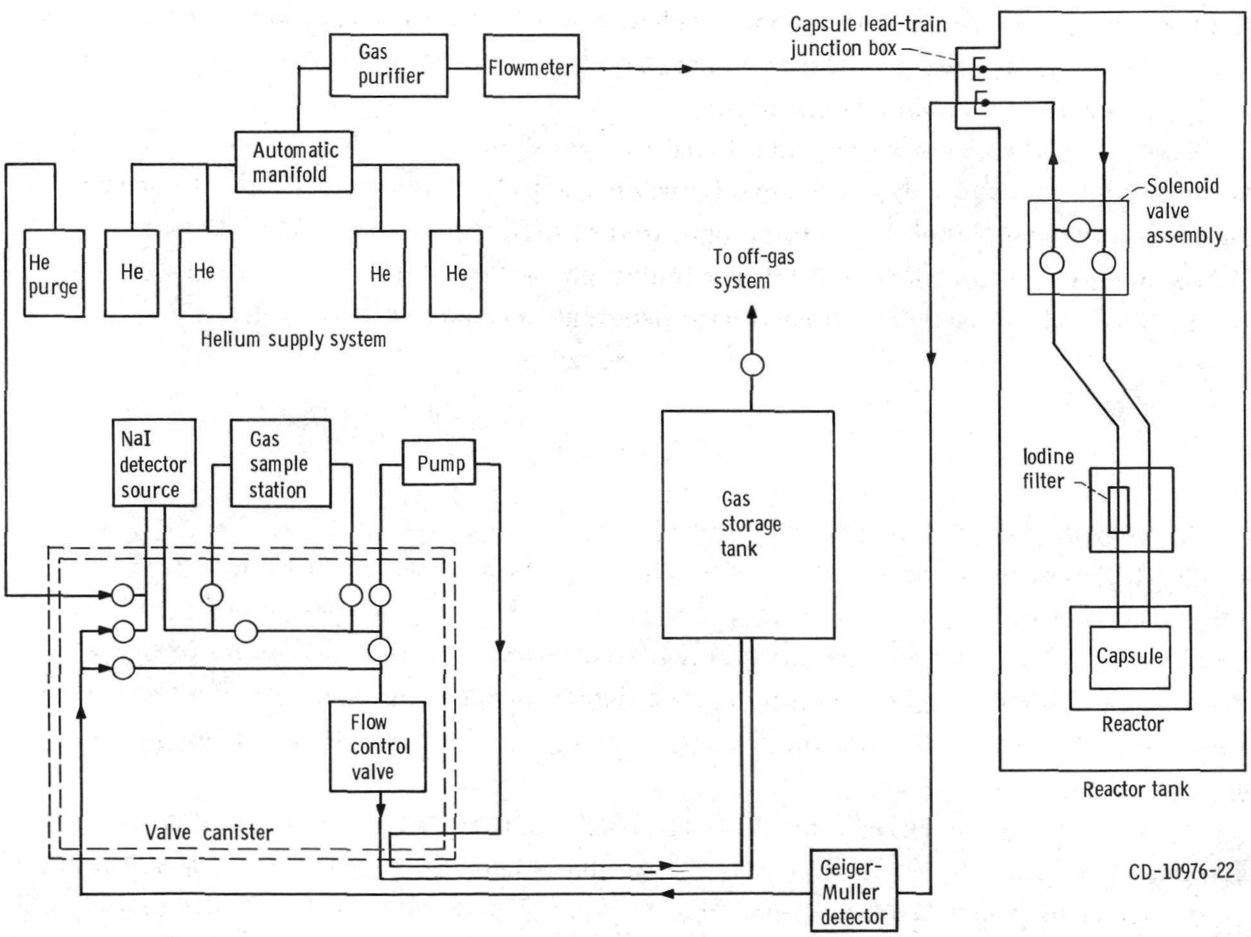


Figure 1. - Simplified schematic of sweep gas facility.

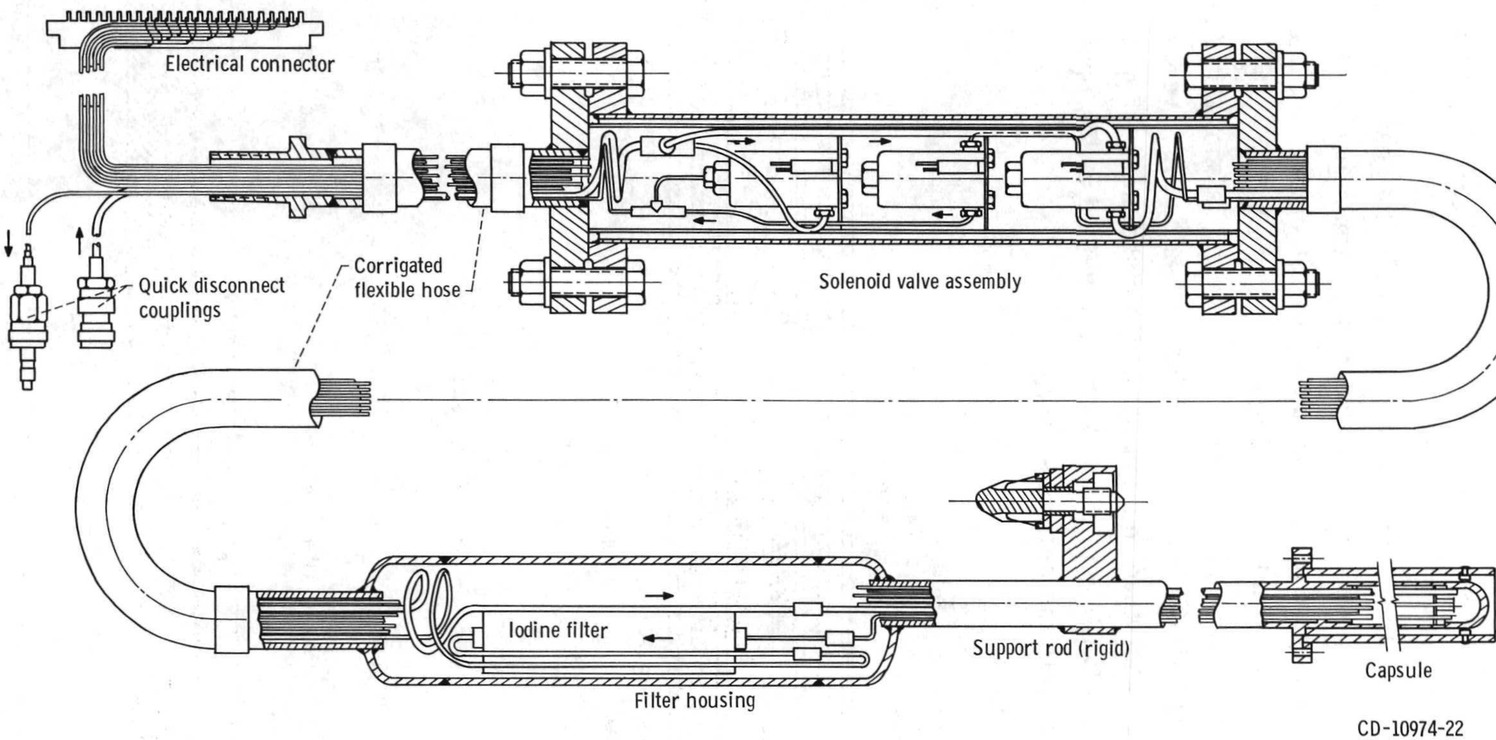


Figure 2. - Instrument lead system.

and 3. Some of the more pertinent detail is repeated in this section.

A simplified flow schematic of the sweep gas facility is shown in figure 1. Bottle-supplied helium is piped through a gas dehumidifier-purifier, a flowmeter, and an irradiation capsule containing a fuel specimen.

The portion of the sweep gas facility located inside the reactor tank consists of an instrument lead system (fig. 2) containing the irradiation capsule, the iodine filter, and the solenoid valve assembly, plus the capsule insertion device. The entire instrument lead system is replaced as a unit with each new fuel specimen.

The small-volume irradiation capsule that was used is shown in figure 3. It is designed to irradiate small-diameter fuel cylinders contained in a vented pin.

The fuel sample is contained in a cylindrical molybdenum - 5-percent-titanium - 0.08-percent-zirconium (TZM) pin (fig. 4) which is vented at the upper end to permit the escape of fission gases into the helium sweep gas.

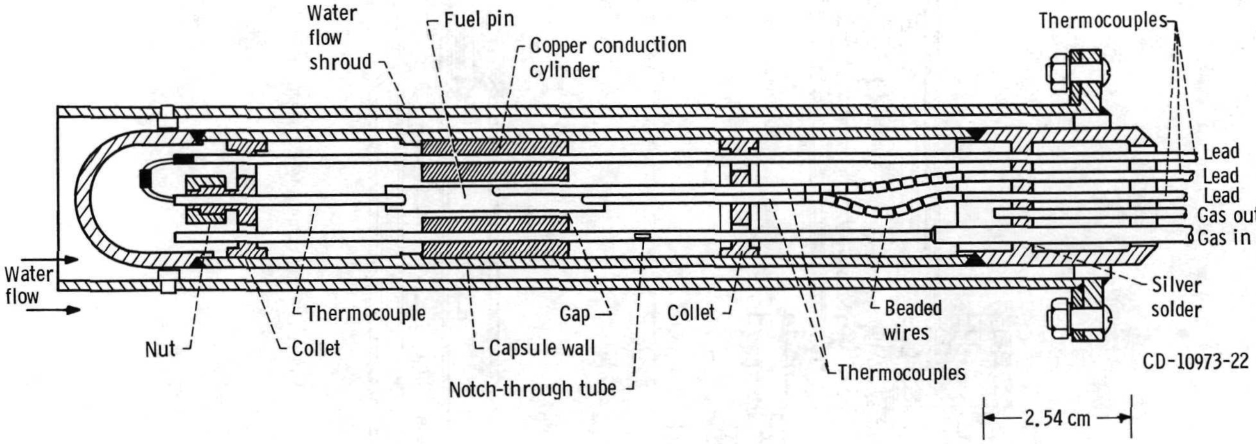


Figure 3. - Irradiation capsule.

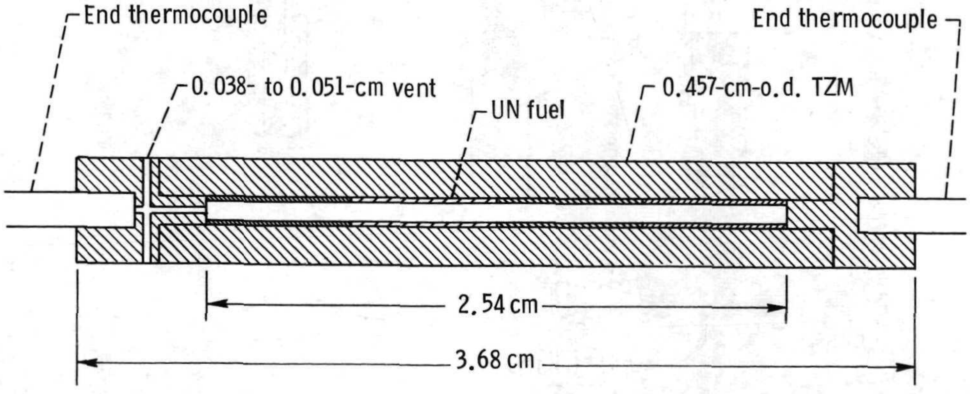


Figure 4. - Fuel pin assembly.

The fuel pin is axially supported and centered in the capsule by means of thermocouple sheathing tubes (molybdenum) attached at end end of the pin. A third thermocouple is set in a slot machined in the TZM clad.

Each fuel pin irradiated in this program contained four UN pellets having the following nominal dimensions: 0.635 centimeter in length by 0.127 centimeter in outside diameter by 0.0763 centimeter in inside diameter. The total fuel length in each pin was approximately 2.54 centimeters. Pellet size is shown relative to a common pin in figure 5,

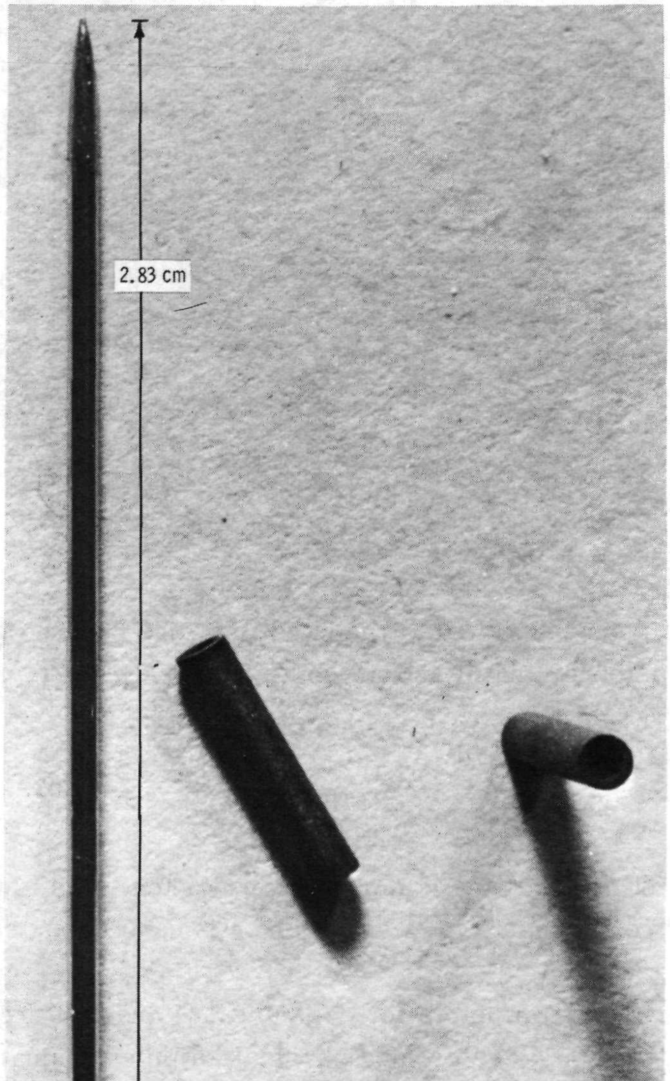


Figure 5. - Uranium nitride fuel pellets compared to 2.83-centimeter-long pin.

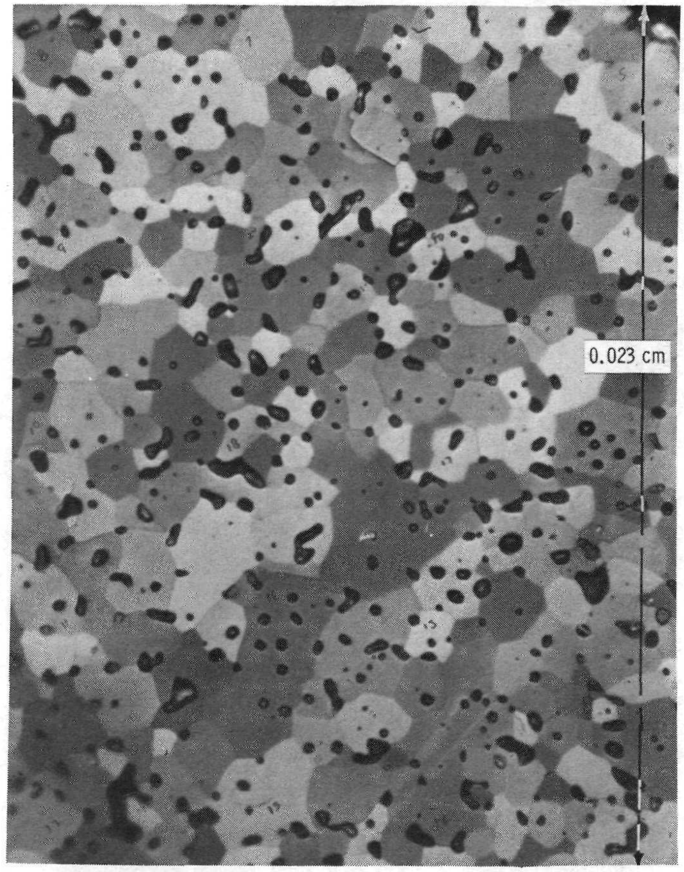


Figure 6. - Section of fuel pin before irradiation. X500.

and the typical fuel microscructure is presented in figure 6. The preirradiation density of all fuel pellets was approximately 93 percent of theoretical.

RESULTS

A concise description of all the completed irradiation tests is presented in table I. Details of the first three, capsules 121, 122, and 123 have been previously presented in reference 3.

Complete temperature-time plots for capsules 124 and 126 are given in figure 7.

The online (scintillation detector) and physical sample (germanium detector) krypton-88 (Kr^{88}) activities in the sweep gas for the two capsules are shown in figures 8 and 9. The actual release rates of fission produced gases can be calculated from the measured gas activities using

TABLE I. - OUTLINE OF IRRADIATION TESTS

Capsule	Total irradiation time, hr	Total burnup, percent	Primary test temperature, K	Temperature range (side thermocouple), K	Fission-rate density range, $\frac{\text{fissions}}{\text{cm}^3\text{-sec}}$	Number of sweep gas samples
121	1294	8.3	1780	1215 to 1789	3.34×10^{14} to 8.29×10^{14}	9
122	1923	6.6	1223	593 to 1225	$.71\times10^{14}$ to 3.64×10^{14}	19
123	1894	6.0	1505	579 to 1512	$.49\times10^{14}$ to 4.45×10^{14}	19
124	1751	7.8	Several	[a]425 to 1323	$.36\times10^{14}$ to 7.57×10^{14}	15
				[b]552 to 1357	$.35\times10^{14}$ to 3.15×10^{14}	11
126	1778	5.6	Several	593 to 1502	----------------------	13

[a]Helium sweep gas.
[b]Neon sweep gas.

$$R = \frac{\dot{Q}Ae^{\lambda\tau}}{\lambda}$$

where

R release rate, atom/sec

$\dot{Q}$ volumetric flow rate, cm^3/sec

A activity of source gas, disintegrations/cm^3-sec

λ decay constant, sec^{-1}

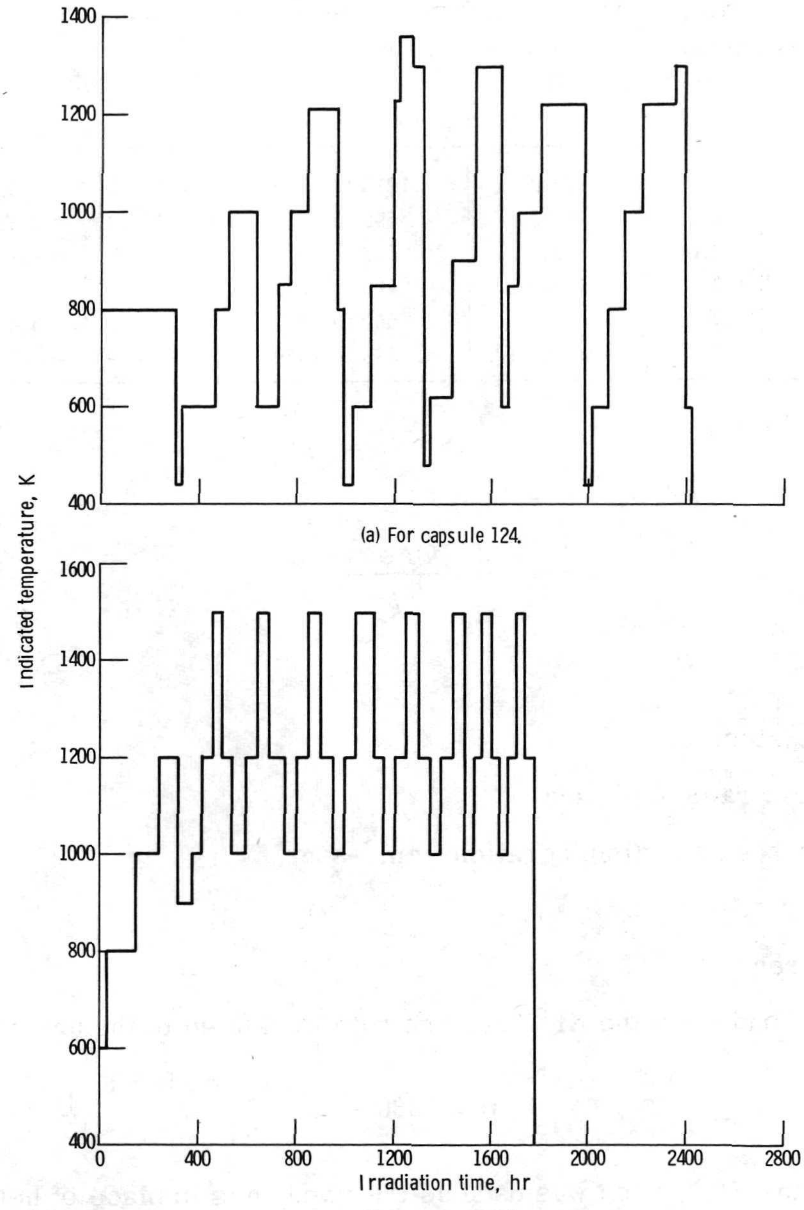
τ travel time, sec

For our operating conditions the Kr^{88} release rate is related to the activity by

$$R = 4280\ A$$

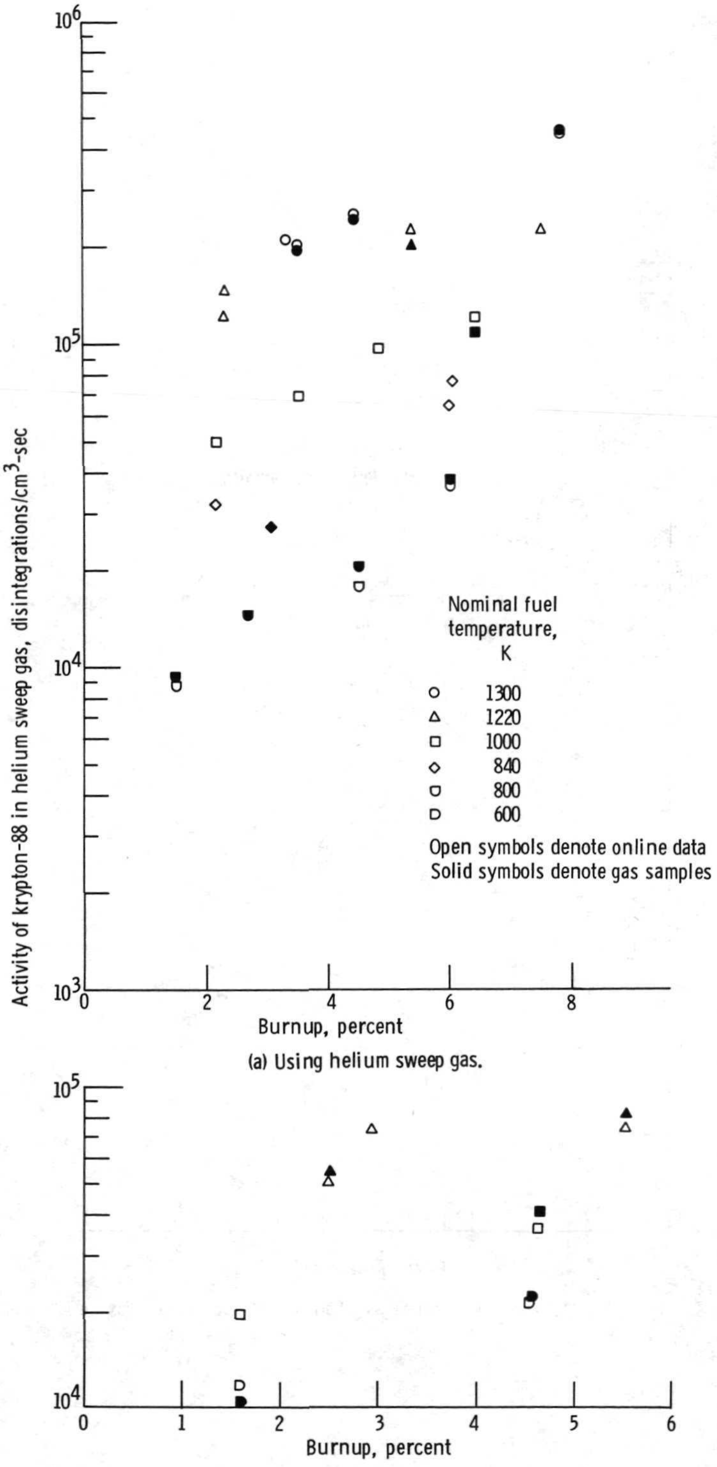
As indicated in figure 8(b), neon was used as the sweep gas in place of helium for many capsule 124 tests. By switching from one gas to the other, we could test the fuel specimen at different burnup rates while keeping the temperature constant. Tabulated values for the data points in figures 8 and 9 are given in tables II and III.

Some low-temperature - low-burnup release rate data for capsule 124 is presented in figure 10. The total burnup range in figure 10 is from 0 to 0.66 percent.



(b) For capsule 126.

Figure 7. - Side thermocouple temperature as a function of irradiation time (small variations not included).



(a) Using helium sweep gas.

(b) Using neon sweep gas.

Figure 8. - Activity of krypton-88 as a function of burnup for capsule 124.

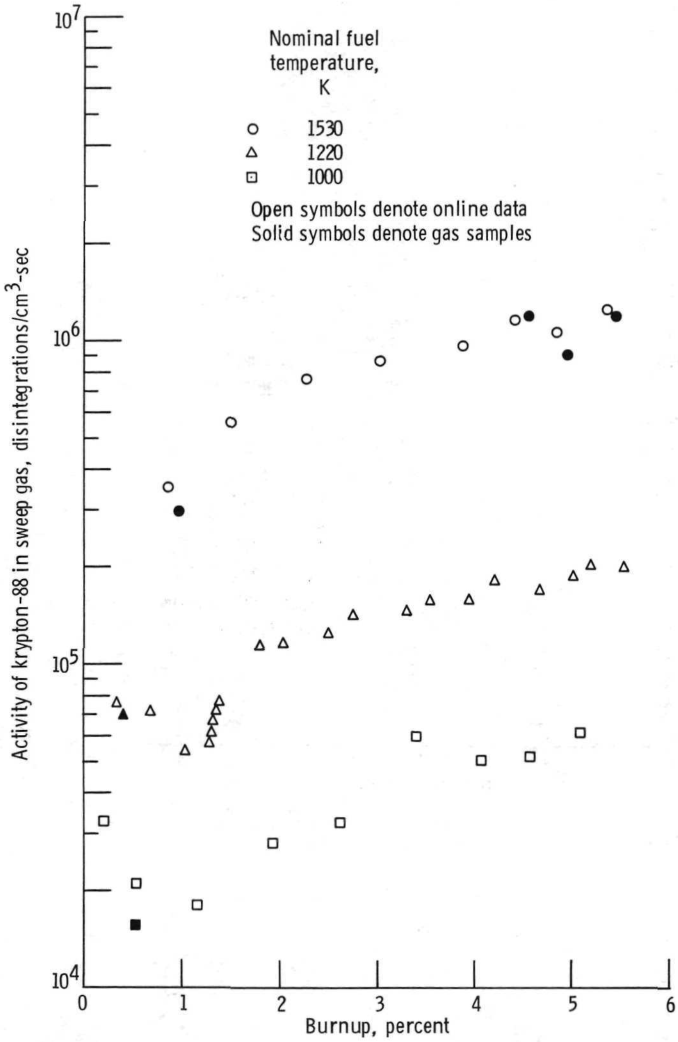


Figure 9. - Activity of krypton-88 as a function of burnup for capsule 126.

TABLE II. - KRYPTON-88 ACTIVITY FOR CAPSULE 124

(a) In helium sweep gas

Fuel maximum temperature, K	Fission-rate density, fissions/(cm^3-sec)	Fuel burnup, percent	Type of data[a]	Measured Kr^{88} activity, disintegrations/(cm^3-sec)
595	1.01×10^{14}	1.53	O	8.69×10^{3}
594	1.10	1.54	S	9.30
1002	3.81	1.84	O	53.2
1230	6.18	2.30	O	123
1230	6.18	2.38	O	148
808	2.25	2.58	O	29.2
810	2.35	2.58	S	29.2
425	.357	2.59	O	3.64
570	.904	2.66	O	11.4
480	.535	2.66	O	4.79
594	1.10	2.66	S	14.5
836	2.45	2.85	O	42.9
1306	7.28	3.33	O	212
1296	7.13	3.53	O	205
1294	7.12	3.53	S	193
1002	3.81	3.55	O	69.5
632	1.21	3.66	O	30.2
626	1.16	3.66	S	30.2
900	2.96	3.89	O	53.9
1316	7.45	4.48	O	252
1323	7.57	4.49	S	241
592	1.09	4.52	S	20.9
594	1.01	4.52	O	17.8
594	1.01	4.52	O	20.6
1003	3.82	4.87	O	97.1
1229	6.17	5.39	O	229
1232	6.33	5.40	S	202
439	.389	5.90	O	4.22
596	1.01	5.98	O	36.6
595	1.10	5.99	S	38.6
810	2.26	6.01	O	77.8
810	2.26	6.16	O	64.2
1016	4.13	6.48	S	109
1012	3.93	6.48	O	121
1226	6.14	7.53	O	229
1318	7.50	7.75	O	444
1318	7.51	7.76	S	454

[a]Sample, S; online, O.

TABLE II. - Concluded. KRYPTON-88 ACTIVITY FOR CAPSULE 124

(b) In neon sweep gas

Fuel maximum temperature, K	Fission-rate density, fissions/cm^3-sec	Fuel burnup, percent	Type of data[a]	Measured Kr^{88} activity, disintegrations/cm^3-sec
849	1.-1×10^{14}	1.59	O	12.0×10^3
851	1.07	1.60	S	10.7
999	1.55	1.64	O	19.9
1219	2.50	2.50	O	50.1
1219	2.49	2.51	S	54.4
429	.21	2.60	O	3.64
570	.39	2.61	O	5.33
595	.43	2.61	S	5.02
841	1.00	2.72	O	16.0
1219	2.50	2.90	O	74.2
1358	3.19	3.08	O	110
1363	3.15	3.10	S	113
629	.48	3.59	O	7.55
631	.50	3.60	S	7.39
895	1.18	3.72	O	19.5
1303	2.90	4.06	O	92.8
1308	2.88	4.06	S	105
833	.99	4.57	O	21.5
835	1.06	4.58	S	22.4
999	1.59	4.65	S	41.1
1000	1.55	4.65	O	36.9
1225	2.53	5.53	O	75.0
1221	2.50	5.54	S	81.8

[a]Sample, S; online, O.

TABLE III. - KRYPTON-88 ACTIVITY IN HELIUM SWEEP GAS FOR CAPSULE 126 (ONLINE DATA)

Fuel maximum temperature, K	Fission-rate density, $\frac{\text{fissions}}{\text{cm}^3\text{-sec}}$	Fuel burnup, percent	Measured Kr^{88} activity, $\frac{\text{disintegrations}}{\text{cm}^3\text{-sec}}$
593	0.54×10^{14}	0.03	4.5×10^{3}
800	1.02	.08	15.4
[a]795	.54	.11	6.8
[a]998	.89	.16	14.7
1009	1.87	.27	32.9
1231	2.93	.42	75.0
[a]1218	1.31	.46	25.8
914	1.25	.53	18.6
1009	183	.63	29.5
1223	2.91	.77	70.8
1536	5.05	.97	351
1224	2.92	1.13	53.7
999	1.79	1.23	17.9
1223	2.91	1.38	66.7
1531	5.02	1.66	560
1229	2.96	1.90	111
1003	1.81	1.99	28.0
1228	2.95	2.15	115
1531	5.00	2.42	768
1226	2.94	2.58	123
1000	1.79	2.68	32.6
1229	2.96	2.84	14.1
1534	5.03	3.23	863
1225	2.93	3.37	144
1000	1.79	3.47	58.9
1224	2.92	3.63	156
1531	5.00	3.89	962
1226	2.94	4.05	157
1008	1.82	4.14	50.0
1225	2.93	4.30	180
1531	5.00	4.57	1170
1002	1.81	4.64	51.4
1230	2.97	4.76	170
1531	5.00	4.96	1070
1229	2.96	5.09	188
1008	1.82	5.16	61.6
1223	2.91	5.28	20.2
1533	5.02	5.48	1255
1227	4.94	5.60	199

[a]Neon sweep gas used for short while.

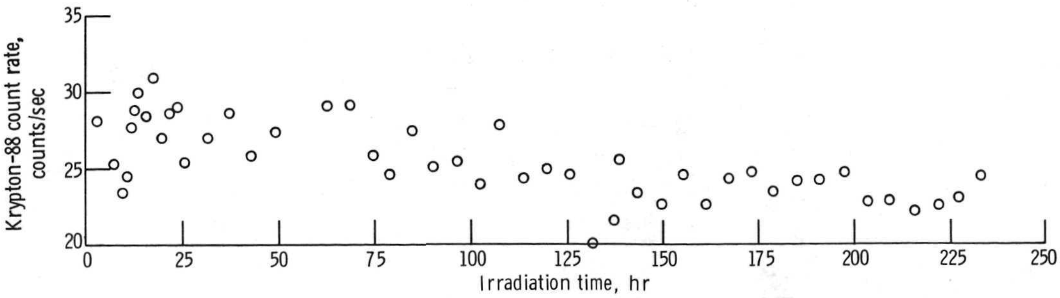


Figure 10. - Decrease in krypton-88 release rate with time from start of irradiation. Temperature of capsule 124, ~ 800 K.

Physical samples of the sweep gas were periodically obtained for accurate and detailed analysis using a Ge(Li) detector. Analyses for the samples obtained during the capsule 124 and 126 irradiations are given in tables IV and V.

The data presented in figures 8 to 10 exactly parallels that reported in reference 3. Release rates increase substantially with increasing temperature. At burnups above ~1.5 percent the release rate increases with increasing burnup. At low burnups (fig. 10) the gas release rate decreases as burnup increases.

Postirradiation photomicrographs for capsules 124 and 126 are shown in figures 11 and 12. Comparison of figure 11(b) (capsule 124, X500) and figure 12(c) (capsule 126, X500) shows striking differences. The fuel from capsule 126 has a great amount of intergranular porosity and obvious bubble formation, while the fuel from capsule 124 shows little porosity formation.

This difference in porosity formation is exactly as reported in reference 3 for capsules 122 and 123. The specimen from capsule 123, which also operated at temperatures over 1500 K showed the same large-scale formation of intergranular porosity as seen here in capsule 126. The intergranular porosity formed in capsule 123 was preferentially oriented perpendicular to the temperature gradient. The intergranular porosity in capsule 126 is not so obviously oriented.

Burnup and Gamma Scan Results

Many different radiochemical postirradiation tests were conducted on the capsule 124 and 126 fuel specimens. For capsule 124 the results are summarized in table VI(a), and for capsule 126 the results are shown in table VI(b) and figure 13.

The gamma scan results shown in figure 13 indicate flux peaking near the ends of the fuel specimen. It is not known why the uppermost group of data points (i.e., the points at the 2.46 cm position) are low. It might simply be the result of a slight inaccuracy in pin position relative to the gamma scan collimator.

TABLE IV. - FISSION GAS ACTIVITY FOR CAPSULE 124 (GAS SAMPLES)

(a) In helium sweep gas

Fuel maximum temperature, K	Fission-rate density, fissions / cm^3-sec	Fuel burnup, percent	Isotope activity, disintegrations/cm^3-sec			
			Kr^{85m}	Kr^{87}	Kr^{88}	Xe^{135}
806	2.33×10^{14}	0.53	7.96×10^3	18.2×10^3	25.2×10^3	22.2×10^4
803	2.29	.82	8.16	16.5	24.6	21.4
554	.90	.93	2.07	3.35	6.02	6.33
1008	3.99	1.42	19.6	39.3	56.5	48.3
594	1.10	1.54	3.23	5.93	9.30	8.96
810	2.35	2.58	10.0	19.0	29.2	28.5
594	1.10	2.66	5.02	8.95	14.5	14.7
1294	7.12	3.53	69.1	129	193	195
626	1.16	3.66	10.8	20.0	30.2	30.4
1323	7.57	4.49	87.9	162	241	238
592	1.09	4.52	7.62	14.2	20.9	20.3
1232	6.33	5.40	70.3	138	202	195
595	1.10	5.99	13.5	28.3	38.6	38.6
1016	4.13	6.48	38.1	71.6	109	103
1318	7.51	7.76	173	278	454	436

(b) In neon sweep gas

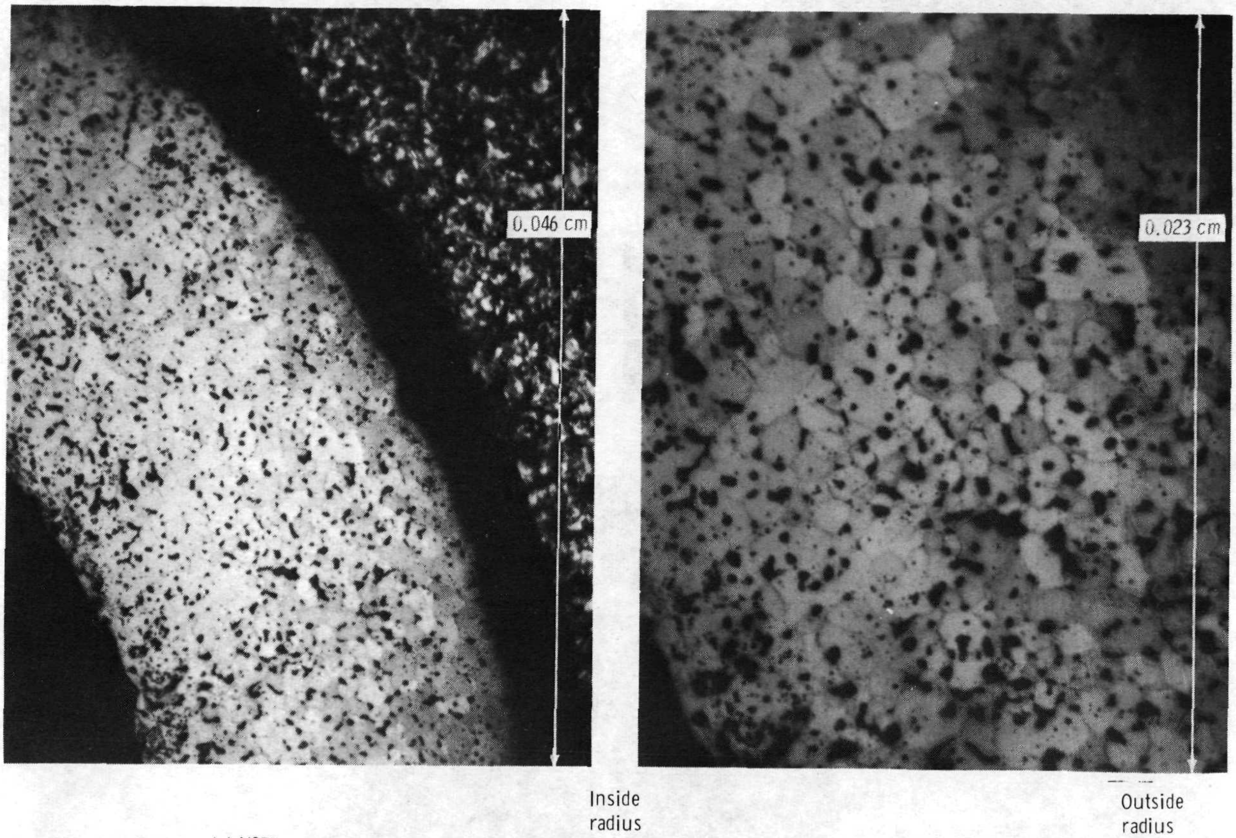
Fuel maximum temperature, K	Fission-rate density, fissions / cm^3-sec	Fuel burnup, percent	Isotope activity, disintegrations/cm^3-sec			
			Kr^{85m}	Kr^{87}	Kr^{88}	Xe^{135}
554	0.35×10^{14}	0.84	0.886×10^3	1.88×10^3	2.73×10^3	2.44×10^3
999	1.59	1.11	7.96	16.4	23.3	20.4
851	1.07	1.60	3.85	8.59	10.7	10.8
1219	2.49	2.51	19.8	36.4	54.4	57.6
595	.43	2.61	1.65	3.45	5.02	5.02
1363	3.15	3.10	40.5	75.5	113	128
631	.50	3.60	2.62	5.47	7.39	7.85
1308	2.88	4.06	35.5	67.9	105	107
835	1.06	4.58	7.85	15.0	22.4	23.7
999	1.59	4.65	14.9	27.5	41.1	43.2
1221	2.50	5.54	29.0	58.5	81.8	86.9

TABLE V. - CAPSULE 126 FISSION GAS ACTIVITY IN SWEEP GAS SAMPLE

[Helium is sweep gas unless indicated otherwise.]

Fuel maximum temperature, K	Fission-rate density, fissions/(cm^3-sec)	Fuel burnup, percent	Isotope activity, disintegrations/cm^3-sec			
			Kr^{85m}	Kr^{87}	Kr^{88}	Xe^{135}
590	0.54×10^{14}	0.03	1.67×10^{3}	3.05×10^{3}	4.66×10^{3}	4.88×10^{3}
[a]795	.54	.11	2.28	4.43	6.74	6.87
1231	2.93	.42	25.6	44.4	69.4	68.3
[a]1217	1.31	.46	9.55	17.5	27.0	17.5
920	1.25	.53	7.47	14.6	21.0	20.8
1531	5.02	.97	116	184	299	423
[b]1534	5.03	1.66	21.5	34.5	52.7	74.0
1527	4.95	2.42	87.4	130	210	300
1534	5.03	3.89	390	570	970	1300
1532	5.02	4.57	460	660	1200	1600
1522	4.90	4.96	360	540	900	1200
1537	5.06	5.48	500	680	1200	1600

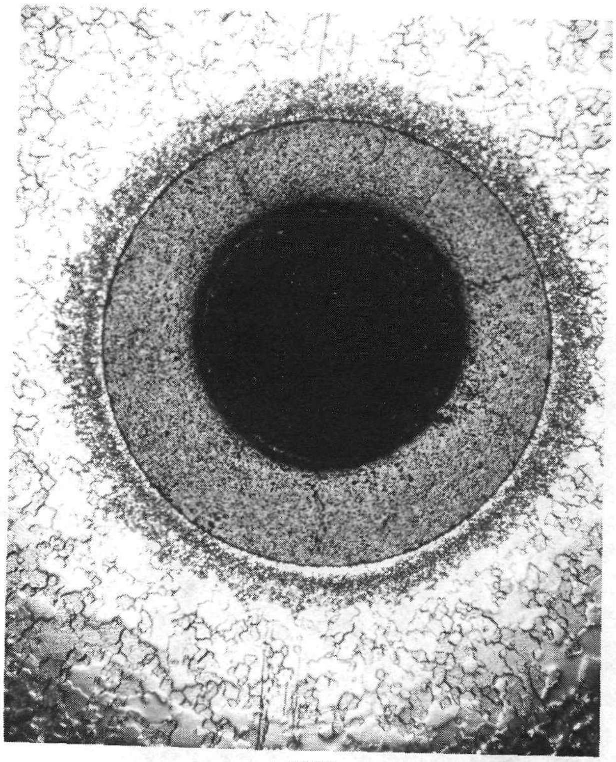
[a]Neon sweep gas.
[b]Low activity.



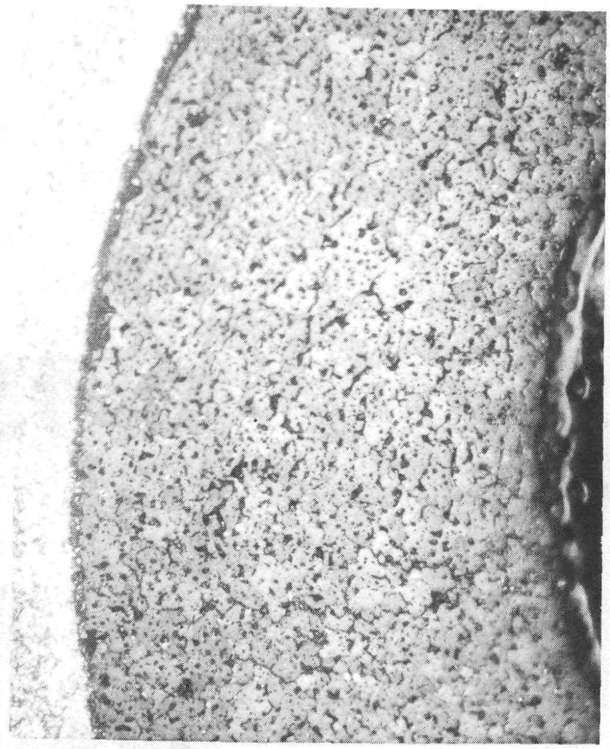
(a) X250.

(b) X500.

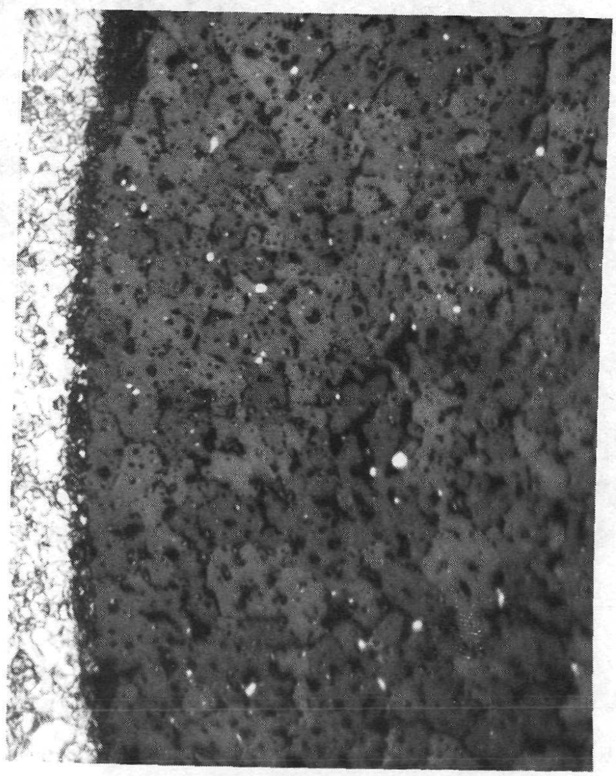
Figure 11. - Section of fuel pin in capsule 124.



(a) X250.



(b) X250



(c) X500

Figure 12. - Section of fuel pin in capsule 126.

TABLE VI. - POSTIRRADIATION RESULTS

(a) For capsule 124

Fuel segment[a]	Burnup, percent			Cs^{137} retention[b], percent
	Mass spectrometer[c]	Ce^{141}[b]	Ce^{144}[b]	
1	10.2	7.3	10.0	77
3	6.9	4.7	6.2	29
5	6.3	3.6	4.7	74

(b) For capsule 126

Pellet[d]	Burnup (mass spectrometer[e]), percent
1	7.4
4	4.9

[a]Pin was sectioned into five segments.
[b]Ge(Li) detector.
[c]U^{236}/U^{235} ratio - recommended values.
[d]Fuel was divided into four pellets.
[e]U^{236}/U^{235} ratio.

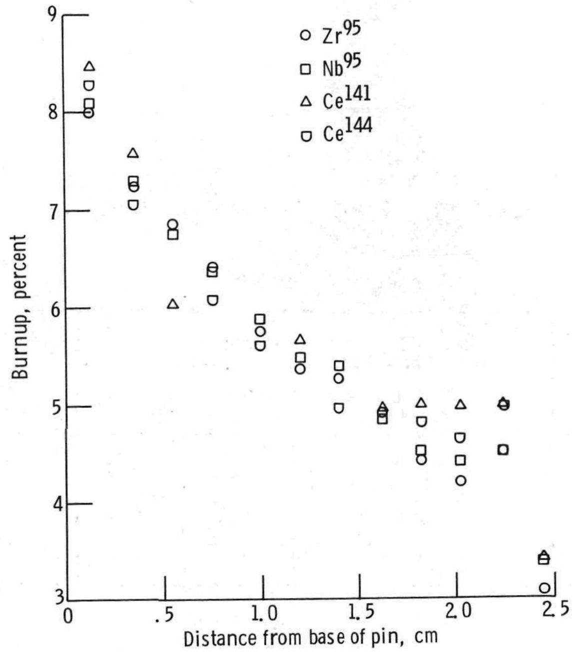


Figure 13. - Gamma scan data of burnup along fuel pin in capsule 126. Pin length, 2.54 centimeters; slit width, 0.12 centimeter.

CONCLUDING REMARKS

Data obtained during and after the irradiation testing of two 93 percent dense uranium nitride (UN) specimens in a sweep gas facility have been presented. Specimen burnups reached 7.8 percent and specimen temperatures ranged from 425 to 1502 K.

We found fission-gas-release rates to increase with increasing temperature, and to decrease then increase with increasing burnup. The minimum has been found to occur at about 1- to 1.5-percent burnup.

Large amounts of interconnected intergranular porosity formed in the fuel specimen operated at 1500 K. Fission-gas-release rate behavior agreed with previous test results reported in TN D-7171.

Lewis Research Center,

National Aeronautics and Space Administration,

Cleveland, Ohio, June 26, 1973,

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