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FISSION-GAS-RELEASE FROM UO_2, INTERIM REPORT #1

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Abstract

The evolution of fission products from UO_2 during irradiation at high temperatures is of primary interest to the Gas Cooled Reactor Project. Fuel tests consisting of UO_2 pellets encapsuled in Inconel or stainless steel have been irradiated in the LITR, ORR and ETR. The capsules were pierced in hot cells, and the gases collected in evacuated systems. Fractions of this gas, of suitable activity for counting, were taken and then analyzed by gamma spectrometry. Larger fractions of gas were analyzed by mass spectrometry. Percentage of gas release varied widely, increasing with temperature, impurity content, oxygen to uranium ratio of the UO_2, and decreasing with bulk density. For high density, stoichiometric UO_2, the gas release was generally less then 3% up to a temperature of about 2800°F, about which it was greatly accelerated.

Fuel burnups of up to 22,000 MWD/MT were obtained. Maximum measured central fuel temperatures of 3150°F were reached. The lower density, nonstoichiometric UO_2 released greater amounts of fission gas, particularly Kr^{85}.

INTRODUCTION

During the past eighteen months an extensive irradiation and evaluation program has been in existence in support of the EGCR fuel element design. The program includes investigation of UO_2 pellet geometry, single-rod prototype element tests, structural and property changes of UO_2, and retention of gaseous and solid fission products. This report will consider the results to date on the last item.

The postirradiation examinations were made on capsules irradiated in the LITR, ORR, and on one capsule irradiated in the ETR. Reactor Projects Division personnel[1] performed the irradiations with UO_2 provided by the Metallurgy Division.[2] The LITR irradiations were intended primarily as fission gas release experiments under conditions of accelerated burnup and measured elevated central UO_2 temperatures. They also inadvertently, provided an additional parameter of stoichiometry. The ORR and ETR irradiations are prototype experiments in which questions of clad deformation and pellet fragmentation are resolved by means of a controlled clad temperature. The postirradiation investigations were a group effort. Hot cell operation and equipment: H. E. Robertson, J. W. Gooch, and T. W. Fulton, Solid State Division. Analyses: J. R. Sites and T. H. Handley, Analytical Chemistry Division.

[1]D. B. Trauger, Irradiation Engineering Department; W. E. Thomas, LITR irradiations; F. R. McQuilkin, ORR irradiations.

[2]L. M. Doney and A. J. Taylor, Ceramics Laboratory.

IRRADIATION HISTORY

The LITR tests contained small hollow cylinders of UO_2. All had an ID of 0.078 in.; OD's were either 0.154 in. or 0.156 in.; lengths were either 0.2, 0.25, or 0.4 in. They were sealed in helium in a compartmented capsule, with thermocouples installed in the center hole of the UO_2 bushing (see Fig. 1). Tantalum clad tungsten-rhenium couples were used to monitor the elevated temperatures. The sample temperature is controlled by passing air over the outside of the capsule. The bottom half of the capsule or "b" sample, operated at a higher temperature than the top half, "a" sample. The fill gas in all the irradiations was either helium or argon. The burnup was calculated from the neutron activation of the monitor wire attached to each experiment and is reported in megawatt days per metric ton uranium (Mwd/MT). The fission-heat density ranged from 260 to 1090 w/cc, depending on the U^{235} enrichment and neutron flux. The integral of $kd\phi$ (w/cm) from outside-to-inside pellet temperature varied from 1.8 to 4.3, k being an average thermal conductivity of the UO_2.

These pellets were, almost without exception, cracked into at least quarters radially as the result of the thermal stress during irradiation.

The indicated temperature inside the UO_2 pellets generally declined during the irradiation. Contamination of the thermocouple with impurities and burnup of the U^{235} could be responsible for this behavior. Thermal

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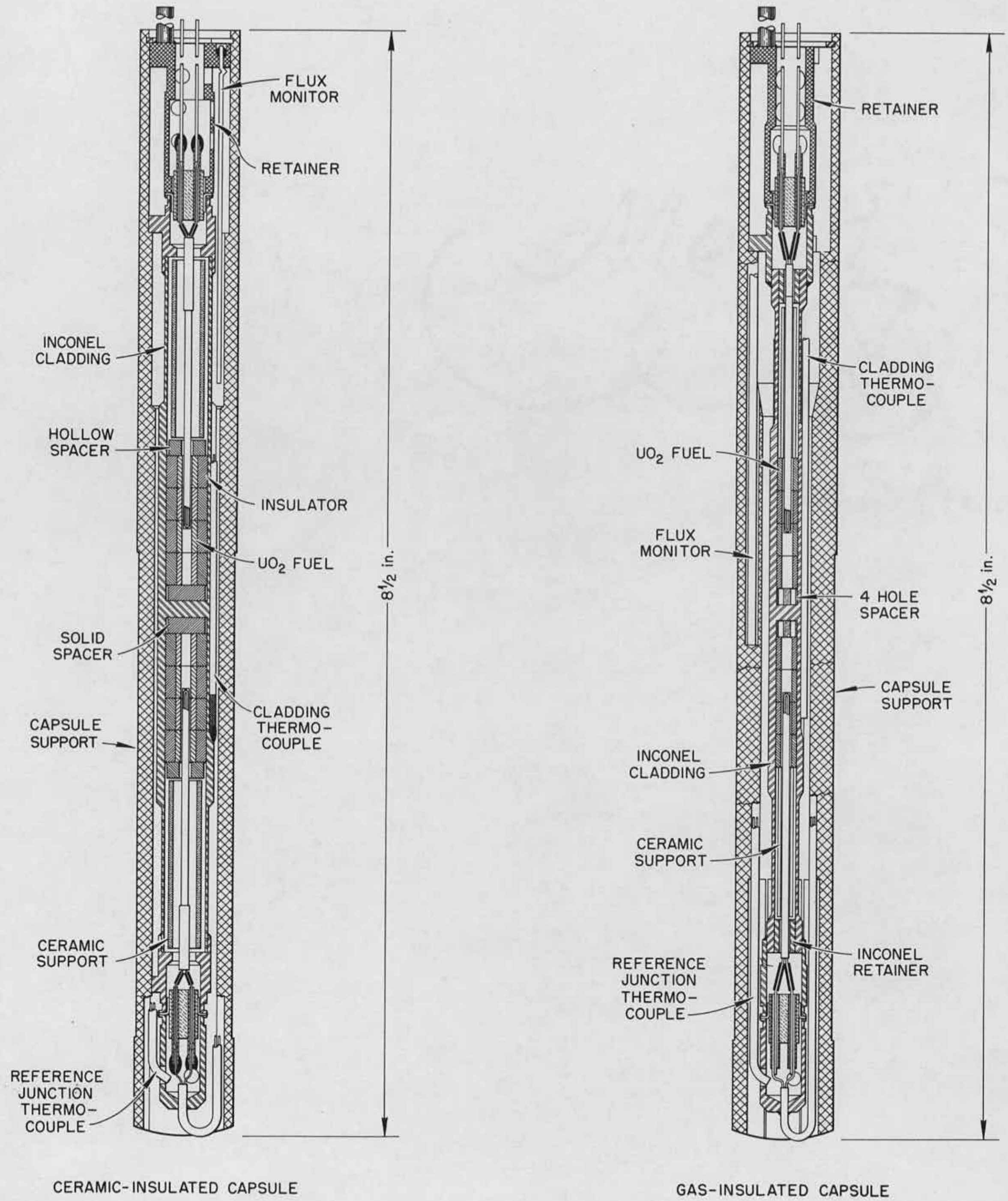

Fig. 1. Experimental Assemblies for LITR Fission-Gas-Release Experiments.

cycles were caused by changes in reactor power, with adjustments to adjacent experiments and cracking and shifting of the fuel pellets as possible contributors. An average temperature for the last three to four weeks of irradiation was selected from the records of operation and considered to be most important in studying the release of fission gases, Xe^{133} in particular. With reference to Kr^{85}, which has a half life of ten years, and the stable isotopes of Kr and Xe, a somewhat higher temperature should be used.

The ΔT across the UO_2 pellets varied, depending mainly upon size. For the small LITR pellets, the ΔT is about 100 to 400°F, while for the larger pellets irradiated in the ORR and ETR, values of 1000 to 2000°F have been calculated. The ORR irradiations contained larger pellets of UO_2: OD, 0.705 in.; ID, 0.323 in.; 0.5 in. long. Heat generation of 71 to 220 w/cc was obtained. Figure 2 shows the type of capsule used in these experiments. The center UO_2 temperature is not measured. The capsule irradiated in the ETR is similar; E-1 contained solid pellets.

FISSION-GAS-RELEASE EXPERIMENTS

LITR Irradiations

The capsules were punctured with a needle valve in the hot cell at ambient temperature, and the gas expanded into evacuated dilution tanks (Fig. 3). Samples of the gas were then taken for gamma-spectrometer and mass-spectrometer analysis. Quantitative values were

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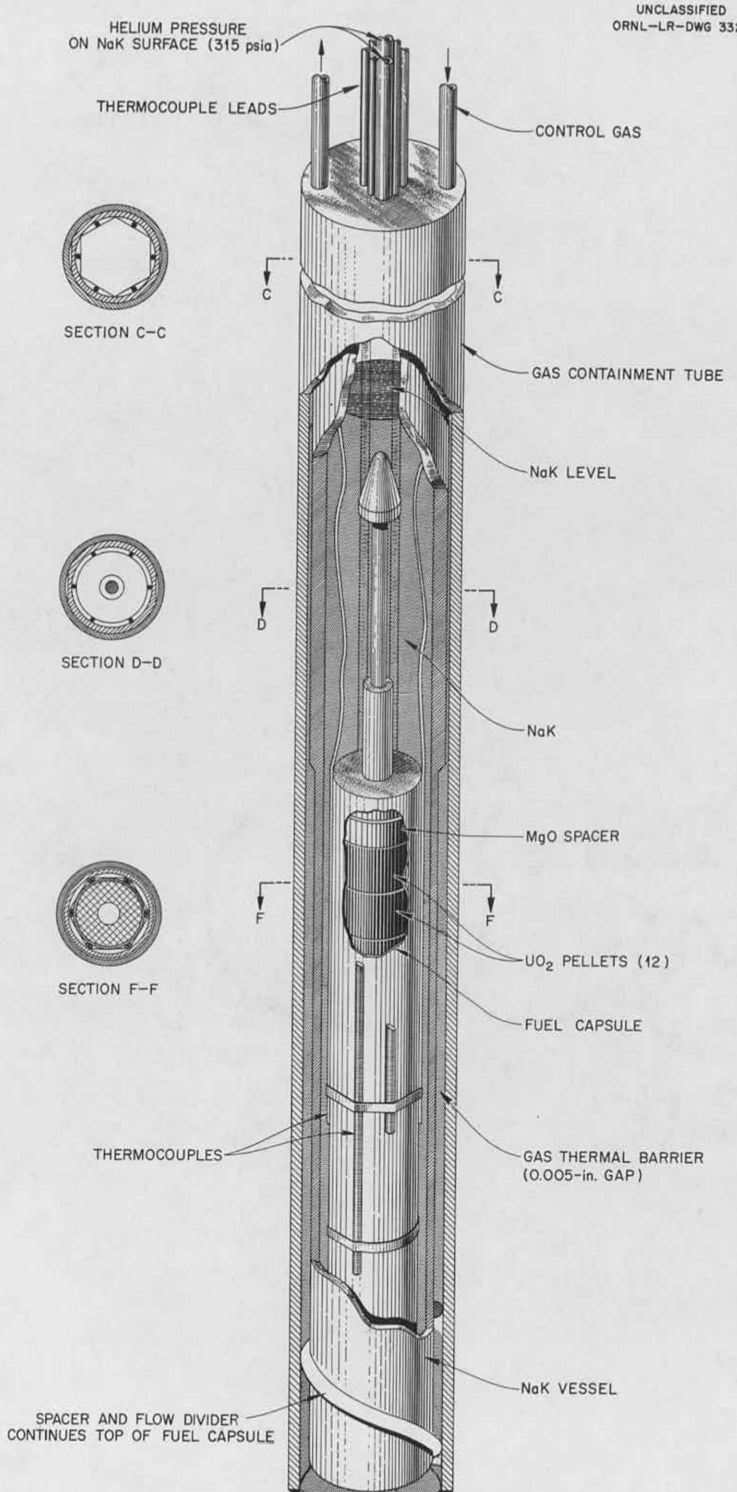

Fig. 2. Apparatus for Fuel Capsule Irradiations in the ORR and ETR.

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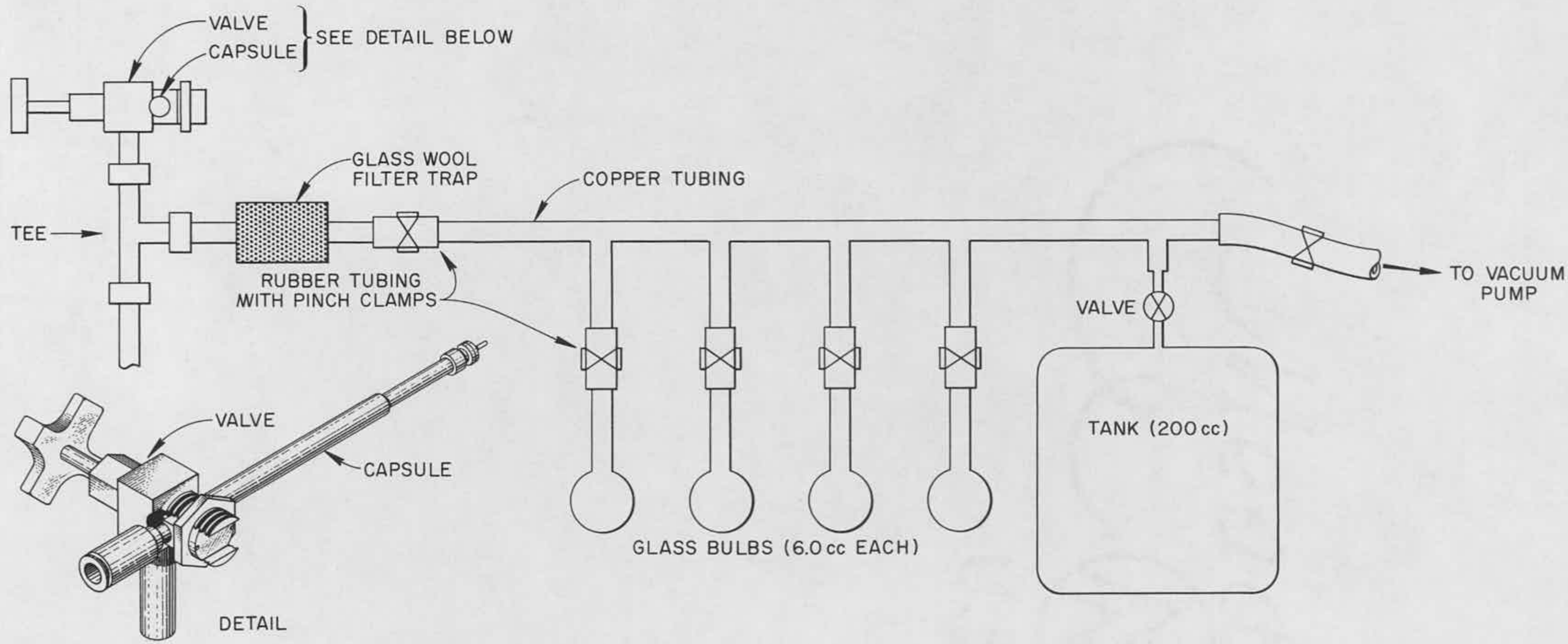

Fig. 3. Fission-Gas Collection Apparatus.

obtained from the mass ratio analysis by counting the Kr^{85}. The percentage of gas release reported is defined as "the number of atoms of isotope A found in the gas surrounding the UO_2", divided by "the total number of atoms of isotope A in the capsule". The data are adjusted to correspond to the time that the irradiation ended. For the species of shorter half life the release reflects the irradiation history of the UO_2 for only the last few days.

Discussion

Twenty-two LITR capsules have been irradiated, and as all but one were compartmented, this resulted in 43 samples. Of these, three were lost due to in-pile leakage and seven were lost during hot cell operations. Eighteen of the remaining 33 did not meet EGCR UO_2 procurement specifications[3] for oxygen/uranium ratio, uranium assay, and impurity content.

Gas samples from several experiments were analyzed for I^{131}, with release percentages of 10^{-4} to 10^{-6} obtained. These figures are of doubtful value, since the quantity of iodine and iodine compounds retained in the capsule, the gas lines, and the glass-wool particle filter is indeterminate.

The percentage of release of Xe^{131} for the LITR experiments was probably low, depending on the length of irradiation, since the gases

[3] R. M. Evans, Fuel Fabrication Costs, ORNL CF-60-5-127 (May 23, 1960); R. L. Heestand and R. M. Evans, GRR Irradiation Program Fuel Data Summary, ORNL CF-59-12-1 (Apr. 7, 1960).

were removed from the capsule and sampled immediately after reactor shutdown. A large portion of the 8-day I^{131} which was generated during the last month of irradiation and had diffused from the fuel had not decayed to stable Xe^{131}; consequently, it remained on the capsule walls during sampling.

The radioactive fission-gas-release data for UO_2 which had an O/U ratio greater than 2.02 are listed in Table 1. The capsules are listed in order of increasing operating temperature. Based on Kr^{85} data, the release was higher for the lower density material. This effect is obscured, however, by the O/U ratio, which varies considerably. All capsules in this group experienced nearly the same number of thermal cycles during the last two weeks of operation. The exception was capsule L-26 which had three times the average number of cycles.

There were no large changes in the density, which indicated no change in internal porosity.

The data from UO_2 having an oxygen-to-uranium ratio of less than 2.02 are listed in Table 2. Below 2800°F, the gas release from these higher density samples was small. The latest irradiations above this temperature show a marked increase in gas release.

Stable fission gas release data for four capsules are given in Table 3. The first three contained UO_2 which had densities of around 75% of theoretical while L-186 was 85%.

Table 1. Fission-Gas Release from Nonstoichiometric UO_2 (Ratios greater than 2.02)

Capsule No.	Bulk Density (% Theoretical)	Oxygen to Uranium Ratio	Carbon Content (ppm)	Effective UO_2 Temp. (°F)	Burnup Estimated from Co^{60} (Mwd/MT)	Immersion Density (gm/cc)		Percent of Fission Gas Evolved (Based on estimated burnup)				
						Preirrad.	Postirrad.	Kr^{85}	Kr^{85m}	Kr^{88}	Xe^{133}	Xe^{135}
L-15a	91.43	2.0285	690	1700[a]	5,900	10.05		0.059	0.019	0.004	0.010	0.004
L-14a	88.7	2.0285	690	1950[a]	5,100	10.31	9.92	0.072	0.029	0.011	0.014	0.006
L-14b	90.26	2.0285	690	2100[b]	5,100	10.46	10.04	0.13	0.057	0.019	0.029	0.017
L-2b	84.41	2.1299	2500	2200[b]	3,600	10.06	10.29	13.			9.0	
L-7b	95.15	2.0407		2200[b]	6,100	10.54		0.075	0.019	0.033	0.066	0.035
L-7xa	94.99	2.0407		2200[b]	16,000	10.51		0.074			0.16	
L-7xb	94.85	2.0407		2450[a]	16,000	10.50		4.9			0.85	
L-6a	73.63	2.024		2100[b]	14,000	10.48	10.47	1.9	0.091		0.293	0.049
L-6b	74.18	2.024		2500[b]	14,000	10.52	10.53	2.1	0.035		0.132	0.026
L-8a	84.58	2.1326	3000	2300[a]	7,100	10.19	10.13	4.6			0.007	
L-8b	85.01	2.1326	3000	2500[b]	7,100	10.18	10.33	18.			4.0	
L-11b	85.23	2.1401	630	2400[a]	22,000	10.46		40.			5.3	2.1
L-10b	95.19	2.0407		2800[a]	1,600	10.48	10.26	16.			30.	27.
L-3a	75.01	2.1178	2400	2800[b]	4,600	10.66	10.61	1.5			0.42	
L-3b	74.87	2.1178	2400	3000[b]	4,600	10.66	10.40	24.			19.	
L-18b	85.19	2.082	3100	3150[b]	6,400[c]	10.40	10.47	38.			16.	

[a]Estimated from temperature plots, with emphasis on last month of operation.
[b]Estimated temperature, from calculations and comparisons with other capsules.
[c]Calculated from estimated neutron flux.

Table 2. Fission-Gas Release from Stoichiometric UO_2 (O/U Ratios less than 2.02)

Capsule No.	Bulk Density (% Theoretical)	Oxygen to Uranium Ratio	Carbon Content (ppm)	Effective UO_2 Temp. (°F)	Burnup Estimated from Co^{60} (Mwd/MT)	Immersion Density (gm/cc)		Percent of Fission Gas Evolved (Based on estimated burnup)				
						Preirrad.	Postirrad.	Kr^{85}	Kr^{85m}	Kr^{88}	Xe^{133}	Xe^{135}
UO_2-1-1	96.	2.008		1750[a]	4,000	10.57	10.76		0.026		0.0091	0.0024
UO_2-1-2a	96.	2.008		1750[a]	11,000	10.58	10.49			0.01	0.028	0.008
UO_2-1-2b	97.	2.008		2000[b]	11,000	10.63	10.62			0.025	0.033	0.012
L-17a	93.69	2.002		1800[a]	10,600	10.43	10.52	0.94	0.04	0.04	0.20	0.05
L-17b	93.80	2.002		2000[b]	10,600	10.38	10.46	3.1	0.13	0.24	1.3	0.32
L-16a	94.35	2.0052	70	1900[a]	2,900	10.48	10.66	0.20			0.078	0.021
L-16b	94.38	2.0052	70	2050[b]	2,900	10.47	10.58	0.033[d]		$<$0.075[d]	0.033[d]	0.027[d]
L-13a	93.68	2.0158	6500	1900[a]	~ 11,000[c]	10.60	10.67	0.21			0.034	
L-13b	92.07	2.0158	6500	2200[b]	~ 11,000[c]	10.60	10.68	2.0			0.057	
L-4a	92.07	2.0158	6500	2300[b]	14,000	10.33	10.49	0.22	0.011	0.021	0.028	0.0085
L-4b	93.35	2.0158	6500	2500[b]	14,000	10.56	10.41	0.54	0.012	0.020	0.04	0.0075
L-25a	95.17	2.020	200	2150[a]	5,100	10.69		0.15			0.039	0.019
L-25b	94.90	2.020	200	2500[b]	5,100	10.73		0.15			0.22	2.0
L-16xa	93.98	2.0052	70	2700[b]	3,200	10.46		1.9				
L-16xb	94.35	2.0052	70	3000[a]	3,200	10.48		6.4				
L-17xa	94.62	2.0052	70	2800[b]	3,100	10.48	10.57				30.	4.5
L-17xb	94.83	2.0052	70	3100[a]	3,100	10.45	10.54	24.			61.	20.

[a]Estimated from temperature plots, with emphasis on last month of operation.

[b]Estimated temperature, from calculations and comparisons with other capsules.

[c]Calculated from estimated neutron flux.

[d]Probably low, since postirradiation examination indicated a small leak in the capsule during operation.

Table 3. Stable Fission Gas Release from Nonstoichiometric UO_2

Capsule No.	O/U Ratio	Effective UO_2 Temp. (°F)	Percent Released Kr^{83}	Kr^{84}	Kr^{86}	Xe^{131}	Xe^{132}	Xe^{134}	Xe^{136}	Kr/Xe* Ratio
L-6a	2.024	2100	2.2	2.0	2.0	1.4	1.6	1.6	1.7	0.21
L-6b	2.024	2500	2.3	2.2	2.2	1.1	1.4	1.4	1.4	0.27
L-3a	2.1178	2800	2.2	1.6	1.6	1.3	1.4	1.1	1.2	0.23
L-18b	2.082	3150	44	39	40	22	23	24	26	0.27
L-7xb	2.0407	2450	5.0	4.6	4.8	2.6	2.6	2.6	2.8	0.29
L-11b	2.1401	2400	49	45	45	29	32	29	30	0.25

* The naturally occurring Kr/Xe ratio from U^{235} fission is 0.157.

ORR and ETR Irradiations

Four of the prototype capsules containing hollow pellets of UO_2 were pierced in the hot cell in a manner similar to the LITR capsules. The time elapsed since the end of irradiation was such that only Kr^{85} was present of the radioactive isotopes. The ETR prototype capsule was sampled at the General Electric Vallecitos Atomic Laboratory and the gas analyzed for Kr^{85}. This capsule contained solid UO_2 pellets. The results are shown in Table 4. The center temperatures are calculated values. The nominal bulk density was 95% of theoretical. Additional mass analysis data will be obtained on capsule 0-1.

Table 4. Stable Fission Gas Release, ORR and ETR Capsules

Capsule No.	UO_2 Central Temp. (^oF)	Burnup* (Mwd/MT)	O/U Ratio	Isotope							
				Kr^{83}	Kr^{84}	Kr^{85}	Kr^{86}	Xe^{131}	Xe^{132}	Xe^{134}	Xe^{136}
ORR											
0-1	2500	1760	2.0287			0.4					
0-2	2800	1910	2.0365	1	0.8	0.9	0.8	0.5	0.6	0.6	0.7
0-3	2700	1640	2.0287	0.4	0.3	0.5	0.4	0.2	0.3	0.3	0.3
0-5	3100	3260	2.0365	0.3	0.3	0.6	0.3	0.2	0.2	0.2	0.1
ETR											
E-1	3700	1005	2.0285			5.6					

* Calculated from Co^{60} flux monitor.

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