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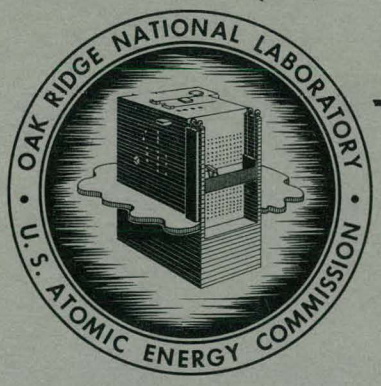
THE RELEASE OF Kr^{85} FROM UO_2 IN ORR CAPSULES

J. L. Scott



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J. L. Scott

Abstract

In an attempt to determine the validity of the method of predicting the release of fission gases from UO_2 suggested recently by Cottrell et al., a series of calculations were made of the expected release of Kr^{85} from prototype Experimental Gas-Cooled Reactor (EGCR) fuel capsule irradiated in the Oak Ridge Research Reactor (ORR). The computed values were then compared with measured values of the per cent Kr^{85} released. In the calculations, the thermal conductivity of the UO_2 was assumed to be 0.028 w/cm-°C in the temperature range 700–1600°C, and in the absence of a precise knowledge of the helium gap, the cases of a 3-mil helium gas and no gap were treated. Values of the release-rate parameter (D') were estimated from BET surface areas of the UO_2 pellets.

Results showed that the measured values of the per cent Kr^{85} released generally fell within or close to the limits set by the 3-mil helium gap and no gap conditions. There was also a definite correlation between the measured values and the 3-mil gap condition when the clad temperature was about 700°C. When the clad temperature was about 800°C, the measured values corresponded better to the no gap condition. The correspondence between measured values and the calculated range lends evidence to the method of Cottrell et al. as well as the value of the thermal conductivity of UO_2 used in the calculations. In addition, the surface area of pressed and sintered UO_2 appears to be indicative of the D' values for irradiated material at least up to 2400 Mwd/MT of UO_2.

Introduction

The release of volatile fission products from UO_2 is one of the principal factors governing the irradiation time of fuel elements in the EGCR. Upon release, the volatile fission products contribute to the internal pressure on the cladding. If the amount released is sufficiently large, the internal pressure could exceed the external pressure of 320 psia. When this occurs, a condition of instability exists because the forces on the cladding tend to increase the gas gap and this increases the UO_2 temperature and the rate of release of fission gases. Such a condition would lead ultimately to the rupture of the cladding and the release of activity to the primary coolant stream.

In order to operate fuel elements without failures and yet not be so conservative as to suffer a severe economic penalty, it is important to be able to estimate the fraction of fission gases generated which will be released under known operating conditions within reasonable limits. A method for making such estimates was given by Cottrell et al.[1] recently, but a lack of data precluded the testing of the method on fuel elements operating under EGCR conditions. Within the past few months, the percentages of Kr^{85} released from Groups I and II of the full diameter prototype EGCR fuel capsule irradiated in the ORR were reported.[2,3] Since the operating data for the two groups of capsules have also been reported,[4,5] the method may now be tested under EGCR conditions. This report discusses the method and assumptions and gives a comparison between predicted and measured values of Kr^{85} release from the UO_2 in ORR capsules.

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[1] W. B. Cottrell et al., Fission-Product Release from UO_2, ORNL-2935 (Sept. 13, 1960).

[2] GCR Quar. Prog. Rep. June 30, 1960, ORNL-2964, p. 160.

[3] GCR Quar. Prog. Rep. Mar. 31, 1961, ORNL-3102, p. 160.

[4] GCR Quar. Prog. Rep. Dec. 31, 1959, ORNL-2888, p. 100.

[5] GCR Quar. Prog. Rep. Sept. 30, 1960, ORNL-3015, p. 108.

Procedure

It was first necessary to determine the temperature distribution in the UO_2. This may be calculated by use of equations given by Cottrell et al.[1] if the operating data are known and if one selects the proper values of the thermal conductivity of the UO_2 as well as the effective gas gap between the UO_2 and the clad. For the present calculations, the operating data given in Table 1 were used.

The thermal conductivity of irradiated UO_2 is subject to a considerable uncertainty. However, Robertson et al.[6] and Bates[7] have reported values for the thermal conductivity of UO_2 which are based on a correlation of microstructures of irradiated fuel elements with known temperatures for grain growth and columnar grain formation. These values are shown in Fig. 1. Although there are significant differences between the values of the investigators for both the very high- and relatively low-temperature data, the values are in reasonable agreement in the temperature range 700–1600°C. Fortunately, the operating temperatures of the UO_2 pellets in ORR capsules were in this range.

Computations are simplified by use of a constant value of the thermal conductivity of the UO_2. In the present case, there is little loss of accuracy by use of a constant value of 0.028 w/cm-°C for the thermal conductivity of UO_2 in the temperature range 700–1600°C. Therefore, this value was used in the calculations.

For those fuel elements containing annular pellets (01–06 Groups I and II), the temperature distribution in the UO_2 was calculated by use of the equation:

$$T_r = T_b + \frac{H}{4\pi k}\left[\frac{b^2 - r^2}{b^2 - a^2} - \frac{2a^2}{b^2 - a^2}\ \ell n\ \frac{b}{r}\right]. \qquad (1)$$

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[6] J. A. L. Robertson et al., "Irradiation Behavior of UO_2 Fuel Elements," Nuclear Metallurgy 6, 45 AIME (1959).

[7] J. L. Bates, "Thermal Conductivity of UO_2 Improves at High Temperatures," Nucleonics 19(6), 83 (1961).

Table 1. Operating Data for ORR Capsules Groups I and II

Capsule	Av Surface Temperature (°C)	Av Heat Rating (w/cm)	OD of Pellets (cm)	ID of Pellets (cm)	Initial Cold Helium Gap (cm)	Type 304 Stainless Steel Clad Thickness (cm)	Irradiation Time (days)
	a	b	a	a			a
01-1	719	239	1.793	0.820	0.00763	0.0508	145.8
02-1	823	259	1.793	0.820	0.00763	0.0508	145.8
03-1	704	217	1.793	0.820	0.00763	0.0508	145.8
04-1	720	339	1.793	0.820	0.00763	0.0508	145.8
05-1	790	293	1.793	0.820	0.00763	0.0508	145.8
06-1	805	264	1.793	0.820	0.00763	0.0508	145.8
07-1	677	179	1.793	0.000	0.00763	0.0508	145.8
08-1	816	172	1.793	0.000	0.00763	0.0508	145.8
	c	c	c	c			c
01-2	712	268	1.793	0.820	0.00763	0.0508	144.9
02-2	794	274	1.793	0.820	0.00763	0.0508	144.9
03-2	714	276	1.793	0.820	0.00763	0.0508	144.9
04-2	717	277	1.793	0.820	0.00763	0.0508	144.9
05-2	803	283	1.793	0.820	0.00763	0.0508	144.9
06-2	812	269	1.793	0.820	0.00763	0.0508	144.9
07-2	700	236	1.793	0.000	0.00763	0.0508	144.9
08-2	808	254	1.793	0.000	0.00763	0.0508	144.9

[a] GCR Quar. Prog. Rep. Dec. 31, 1959, ORNL-2888, p. 100.

[b] Values from (a) were corrected by use of flux monitor data given in GCR Quar. Prog. Rep. Mar. 31, 1960, ORNL-2929, p. 150, except for 05-1. For 05-1, it was assumed that there was a typographical error and that the total burn-up was 2030 Mwd/MT of UO_2.

[c] GCR Quar. Prog. Rep. Sept. 30, 1960, ORNL-3015, p. 108.

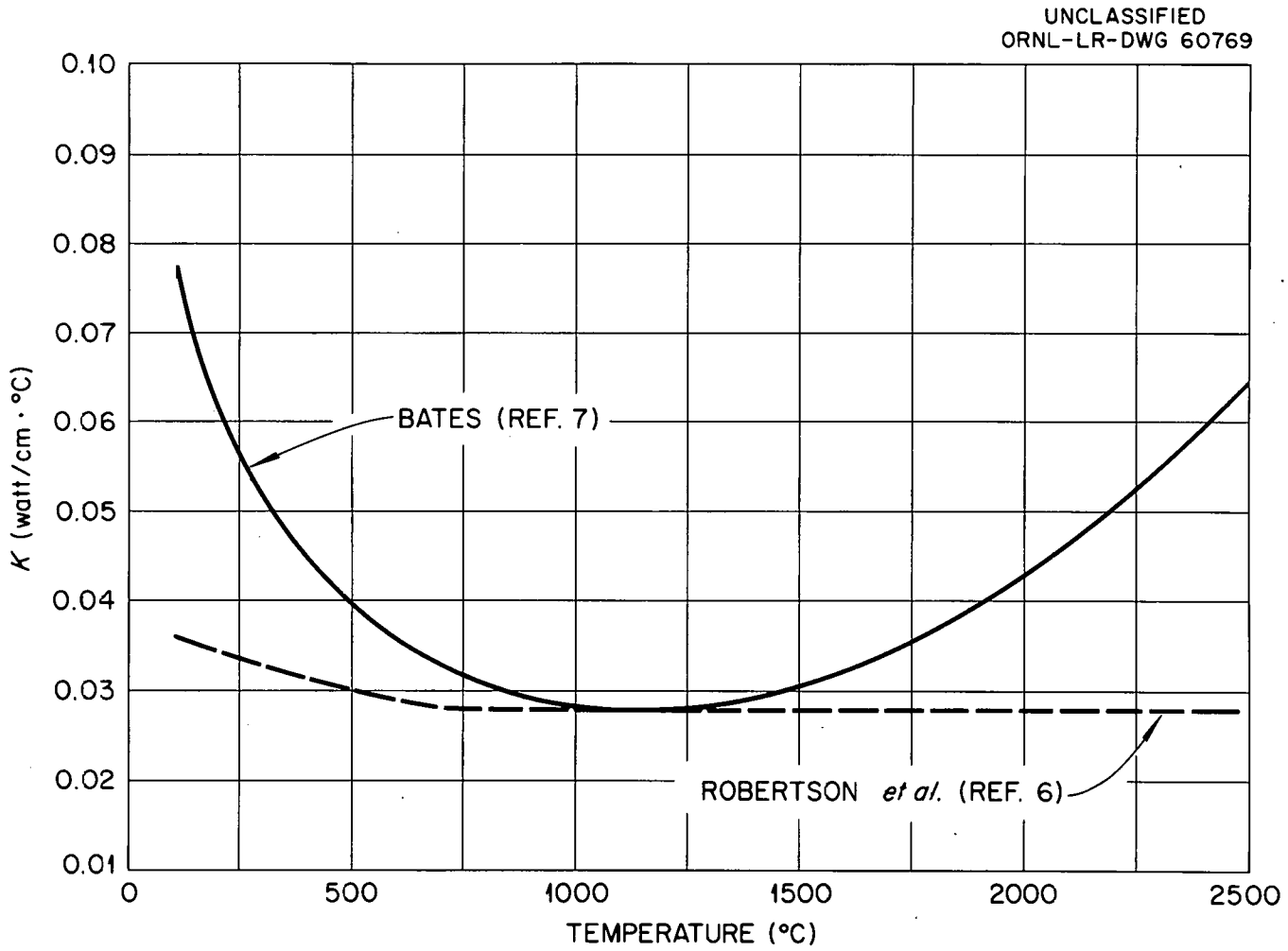

Fig. 1. Selected Values of the Thermal Conductivity of UO_2. References: J. A. L. Robertson et al., Nuclear Metallurgy 6, 45, AIME (1959). J. L. Bates, Nucleonics 19(6), 83 (1961).

In Eq. 1,

T_r = the temperature at radius, r – °C,
T_b = the temperature at the surface of the UO_2 – °C,
H = the heat rating – w/cm
k = the thermal conductivity – w/cm-°C,
b = the outer radius of the pellets – cm, and
a = the inner radius of the pellets – cm.

No corrections were made for the change in heat rating as a result of the flux gradients along the axes of the elements because a postirradiation gamma scan of capsule 01-1[ref 8] indicated that the activity was fairly constant along the axis.

For solid rods, the temperature distribution in the UO_2 was calculated by use of the equation:

$$T_r = T_b + \frac{H}{4\pi k}\left[\frac{b^2 - r^2}{b^2}\right]. \qquad (2)$$

The nomenclature in Eq. 2 is the same as that used in Eq. 1.

The next problem which arises is to decide whether there is or is not a gap between the UO_2 and the cladding material. Under the conditions of operation of 320 psia external pressure and cladding temperatures in the range 675–900°C, the 20-mil, type 304 stainless steel tubing has been observed to collapse,[9] but the phenomenon is temperature and time dependent. Since the temperature was not constant and the internal pressure was not known for certain, it is difficult to predict the time for collapse. Even if the time required for collapse were known, it would be difficult to assign a mean gap over the life of the fuel element. Thus, it was decided to compute the fraction released both with and without the initial cold helium gap and use the result to determine which condition obtained in the individual capsules. For the condition of no gap, the contact resistance to heat transfer was neglected.

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[8] GCR Quar. Prog. Rep. Mar. 31, 1960, ORNL-2929, p. 161.

[9] GCR Semiann. Prog. Rep. Dec. 31, 1958, ORNL-2676, p. 77.

The temperature gradients across the cladding and the helium gap were computed by use of the equation:

$$\Delta T = \frac{H\delta}{\pi k D} . \qquad (3)$$

In Eq. 3,

ΔT = temperature gradient – °C,

δ = cladding thickness or gap width – cm, and

D = average diameter of the cladding or gap – cm.

The other parameters were defined previously.

The values of the thermal conductivities of type 304 stainless steel and helium used in the calculations are shown in Table 2. The data for type 304 stainless steel were taken from the Metals Handbook,[10] and values for helium were the best fit of data of Glasstone[11] and Bates[7] plotted on the basis of a linear relationship between log k and log T(°K).

By use of the surface temperatures of the capsules and the heat ratings in Table 1 together with the dimensions and Eqs. 1, 2, and 3, the temperature distribution in the UO_2 in each capsule was computed for the cases of a 3-mil helium gap and no gap. An average temperature, $\overline{T}$, for the UO_2 in each capsule was then calculated by use of the equation:

$$\overline{T} = \frac{2T_a + T_b}{3} . \qquad (4)$$

A value of the release-rate parameter $D' = D/\alpha^2$, (where D is the diffusion coefficient of Kr^{85} in UO_2 and α is the radius of the equivalent sphere) was then determined for this average temperature. The use of a D' value associated with $\overline{T}$ instead of the more complicated process of summation of individual values of F over different segments of the pellets was discussed by Cottrell et al.[1]

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[10] Taylor Lyman (ed.), Metals Handbook, p. 314, American Society for Metals, Cleveland, 1948.

[11] Samuel Glasstone, Principles of Nuclear Reactor Engineering, p. 844, D. Van Nostrand, New York, 1955.

Table 2. Thermal Conductivities of Type 304 Stainless Steel and Helium

T°C	Type 304 Stainless Steel $k \frac{watts}{cm\ °C}$ (a)	Helium $k \frac{watts}{cm\ °C}$ (b,c)
100	0.163	0.00171
200	0.176	0.00198
300	0.188	0.00218
400	0.201	0.00240
500	0.213	0.00260
600	0.226	0.00279
700	0.238	0.00296
800	0.251	0.00311

[a]Taylor Lyman (ed.), Metals Handbook, p. 314, American Society for Metals, 1948.

[b]J. L. Bates, "Thermal Conductivity of UO_2 Improves at High Temperatures," Nucleonics 19(6), 83 (1961).

[c]Samuel Glasstone, Principles of Nuclear Reactor Engineering, p. 844, D. Van Nostrand, New York, 1955.

Values of D' for the individual capsules at 1400°C were estimated by use of the equation of Toner and Scott:[12]

$$\log D'_{1400°C}\ (\text{sec}^{-1}) = -14.00 + 2 \log S\ (\text{cm}^2/\text{g}), \tag{5}$$

where S is the measured surface area of the pellets in each capsule prior to irradiation. The values of D' at 1400°C were then extrapolated to $\overline{T}$ by use of the equation:

$$\ell n \frac{D'_{\overline{T}}}{D'_{1400°C}} = \frac{70{,}000}{R}\left[\frac{1}{1673} - \frac{1}{\overline{T} + 273}\right]. \tag{6}$$

In Eq. 6, R is the gas constant (1.987 cal/g mole °C). The activation energy of 70 kcal/mole used in Eq. 6 was taken from the work of Long et al.[13] In using Eqs. 5 and 6 for the present analysis, it was assumed that the diffusion coefficients for Kr^{85} in UO_2 are identical to those for Xe^{133} and that irradiation does not affect the diffusion coefficients.

The fraction of Kr^{85} released from the UO_2 was calculated by the equation:[1]

$$F = \frac{4}{\pi^{1/2}} (D't)^{1/2}\ . \tag{7}$$

In this equation,

F = the fraction of Kr^{85} released,

D' = the release-rate parameter — sec^{-1}, and

t = irradiation time — sec.

Results

The values of the temperature increases associated with the cladding, the 3-mil helium gap, and the UO_2 are shown in Table 3 for each capsule.

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[12] D. F. Toner and J. L. Scott, "Study of the Factors Controlling the Release of Xe^{133} from Bulk UO_2," Paper presented at the Annual Meeting of the American Society for Testing Materials, Atlantic City, New Jersey, June 27, 1961. (To be published in the Proceedings of the conference.)

[13] G. Long, D. Davies, and J. R. Findlay, "Diffusion of Fission Products in Uranium Dioxide and Uranium Monocarbide," Paper presented at the First Conference on Nuclear Reactor Chemistry, Gatlinburg, Tennessee, Oct. 12, 1960.

Table 3. Temperature Distribution in ORR Capsules Groups I and II

Capsule	Clad Surface Temperature (°C)	Temperature Rise in Clad (°C)	Temperature Rise in 3-mil Helium Gap (°C)	Temperature Rise in UO_2 (°C)	Central Temperature of UO_2 Without Gap (°C)	Central Temperature of UO_2 With Gap (°C)
01-1	719	8.6	136	397	1125	1261
02-1	823	8.9	139	431	1263	1402
03-1	704	7.9	118	360	1072	1190
04-1	720	12.2	182	563	1295	1477
05-1	790	10.2	149	488	1288	1437
06-1	805	9.1	134	439	1253	1387
07-1	677	6.6	100	509	1193	1293
08-1	816	5.8	83	489	1305	1388
01-2	712	9.7	145	445	1165	1312
02-2	794	9.5	139	455	1259	1398
03-2	714	10.0	149	459	1183	1332
04-2	717	10.0	149	459	1187	1336
05-2	803	9.8	143	470	1283	1426
06-2	812	9.3	136	447	1268	1404
07-2	700	8.6	128	670	1379	1507
08-2	808	8.8	129	722	1539	1668

From these values, the central temperature of the UO_2 was computed for each capsule for the case of the 3-mil helium gap and with no helium gap. These values are also shown in Table 3. With the exception of capsule 08-2 with a 3-mil helium gap, all capsules have central temperatures below 1600°C, the temperature where grain growth might be expected to occur in capsules irradiated for 6 months. The fact that no significant grain growth was observed in capsules 01-1, 02-1, 03-1, 05-1, and 06-1 on postirradiation examination[14] indicates that the actual temperatures were not markedly higher than the calculated values. Long[15] has now examined the microstructure of the UO_2 in capsule 08-2 and observed no grain growth. These temperature estimates for this capsule are probably too high. An error in the flux is suspected.

The estimated values of D' for each capsule are shown in Table 4 for the cases of a 3-mil helium gap and no gap. The measured surface areas of the pellets are also shown in Table 4. Values of $\overline{T}$ were computed by use of Eq. 4 and the temperatures given in Table 3. The values of D' were computed by use of Eqs. 5 and 6 from the measured surface areas and values of $\overline{T}$.

The estimated values of the per cent Kr^{85} release are shown in Table 5 together with measured values. The estimated values were computed by use of Eq. 5. Measured values were based on a branching ratio of 0.36 per cent for Kr^{85}. Both the cases of a 3-mil helium gap and no gap are given in Table 5. The values of the clad temperature are also listed in Table 5 because one would expect more clad collapse to occur at the higher clad temperatures. Thus, one would tend to associate the lower release data on capsules with low cladding temperatures with the 3-mil gap condition and the data from capsules with high cladding temperatures with the condition of no gap. The high estimates for the release in 08-2 are felt to be due to the actual temperature being lower than the estimated temperature as discussed previously. The reason why the measured release in 05-2 and 06-2 correspond better to the 3-mil gap condition than to the no-gap condition is not known.

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[14] E. L. Long, Jr., GCR Quar. Prog. Rep. Mar. 31, 1961, ORNL-3102, p. 163.

[15] Private communication from E. L. Long, Jr.

Table 4. Estimates of D′ Values for ORR Capsule

Capsule	Surface Area[a] (cm^2/g)	D′ (1400°C) (sec^{-1})	$\bar{T}$ (No Gap) (°C)	$\bar{T}$ (3-mil He Gap) (°C)	D′ ($\bar{T}$ - No Gap) (sec^{-1})	D′ ($\bar{T}$ - Gap) (sec^{-1})
01-1	33.8	1.14×10^{-11}	993	1128	1.28×10^{-14}	1.91×10^{-13}
02-1	80.7	6.50×10^{-11}	1119	1258	9.23×10^{-13}	9.22×10^{-12}
03-1	33.8	1.14×10^{-11}	952	1070	5.16×10^{-15}	6.44×10^{-14}
04-1	33.8	1.14×10^{-11}	1107	1289	1.30×10^{-13}	2.55×10^{-12}
05-1	80.7	6.50×10^{-11}	1125	1247	1.04×10^{-12}	1.15×10^{-11}
06-1	80.7	6.50×10^{-11}	1107	1241	7.43×10^{-13}	7.10×10^{-12}
07-1	8.78	7.70×10^{-13}	1023	1123	1.68×10^{-15}	1.18×10^{-14}
08-1	8.78	7.70×10^{-13}	1141	1224	1.69×10^{-14}	6.30×10^{-14}
01-2	23.6	5.58×10^{-12}	1017	1162	1.07×10^{-14}	1.69×10^{-13}
02-2	23.6	5.58×10^{-12}	1107	1246	6.38×10^{-14}	7.86×10^{-13}
03-2	40.6	1.65×10^{-11}	1030	1179	4.17×10^{-14}	6.68×10^{-13}
04-2	41.9	1.75×10^{-11}	1034	1183	4.81×10^{-14}	7.58×10^{-13}
05-2	34.3	1.17×10^{-11}	1126	1269	1.89×10^{-13}	1.95×10^{-12}
06-2	40.0	1.60×10^{-11}	1119	1255	2.27×10^{-13}	2.17×10^{-12}
07-2	18.4	3.39×10^{-12}	1156	1284	9.29×10^{-14}	7.05×10^{-13}
08-2	39.8	1.59×10^{-11}	1299	1428	4.11×10^{-12}	2.10×10^{-11}

[a]Measurements made by P. G. Dake and E. A. Woy of the Works Laboratory, Technical Division, Oak Ridge Gaseous Diffusion Plant.

Table 5. The Percent of Kr^{85} Released in ORR Capsules Groups I and II

Capsule	Predicted Release with no Gap (%)	Predicted Release with 3-mil He Gap (%)	Measured Release (%)	Average Clad Temperature (°C)
			[a]	
01-1	0.09	0.35	0.4	719
02-1	0.77	2.43	0.9	823
03-1	0.06	0.20	0.5	704
04-1	0.29	1.28	-	720
05-1	0.82	2.72	0.6	790
06-1	0.69	2.13	-	805
07-1	0.03	0.09	-	677
08-1	0.10	0.20	-	816
			[b]	
01-2	0.08	0.33	0.53	712
02-2	0.20	0.71	0.55	794
03-2	0.16	0.66	0.76	714
04-2	0.18	0.70	0.07[c]	717
05-2	0.35	1.11	1.51	803
06-2	0.38	1.18	1.1	812
07-2	0.24	0.67	0.51	700
08-2	1.63	3.68	0.31	808

[a] GCR Quar. Prog. Rep. June 30, 1960, ORNL-2961, p. 160.

[b] J. G. Morgan et al., GCR Quar. Prog. Rep. Mar. 31, 1961, ORNL-3102, p. 160.

[c] Capsule leaked.

Considering the uncertainty of the data used to predict the amount of Kr^{85} released and the possibility of error in the measured values, the agreement between observed release and predicted range is excellent. This agreement is perhaps fortuitous; however, it does lend credence to the reported values of the thermal conductivity of UO_2 (ref 16,17) as well as the method of estimating release of Cottrell et al.[18] In addition, the surface area of pressed and sintered UO_2 seems to be useful in estimating the D' values for irradiated material at least up to 2400 Mwd/MT of UO_2.

A word of caution is in order concerning the present method of estimating fission-gas release. When the temperature of a considerable volume of UO_2 is above 1600°C, grain growth in the UO_2 and void migration are observed to occur.[16,17] Under these conditions, the method used should be modified by assuming that all of the gas is released from the volume of UO_2 which is above 1600°C. This latter assumption may be improved in the future when these processes are better understood.

Acknowledgment

The author would like to thank D. A. Douglas, J. H. Coobs, J. W. Prados, and W. B. Cottrell for their advice and suggestions.

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[16] J. A. L. Robertson et al., "Irradiation Behavior of UO_2 Fuel Elements," Nuclear Metallurgy 6, 45 AIME (1959).

[17] J. L. Bates, "Thermal Conductivity of UO_2 Improves at High Temperatures," Nucleonics 19(6), 83 (1961).

[18] W. B. Cottrell et al., Fission-Product Release from UO_2, ORNL-2935 (Sept. 13, 1960).

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