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RELEASE OF NOBLE GASES FROM UO_2 FUEL RODS

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by

W. A. Yuill
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ABSTRACT

An analytical function has been developed for correlating fission gas capsule release data. This function, derived through a thermodynamic approach employing Gibbs-free-energy relationships, provides an excellent correlation of the capsule release data for fuel elements during reactor operation. The thermodynamic approach demonstrates that the outer regions of the UO_2 fuel control fission gas release from a fuel pin during operation and that a large temperature gradient retards the escape of fission gases. The analytical function derived predicts results which are contrary to most of the currently published models, however, it is not entirely unique as is discussed. The equation for the least squares fit of the function and the operating data for a fuel pin may be used to calculate the release fraction to within a factor of two of the observed release fraction. The function also correctly predicts the relation between the decay constant, λ_i , and the release fraction, F_i .

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RELEASE OF NOBLE GASES FROM UO_2 FUEL RODS

The behavior of fission products in uranium dioxide fuel elements is important because of the safety aspects associated with the release of fission products such as the isotopes of iodine and because of the effect fission products have on the physical properties of the uranium dioxide and cladding materials. Noble gas fission products are particularly important because of their contribution to the fuel pin internal pressure which is the cause of fuel pin swelling or rupture during a nuclear accident.

The present high level of interest in the behavior of noble gases in the fuel pins and the difference in interpretations of existing release data are reflected by the papers presented at the Topical American Nuclear Society meeting on "Fundamental Properties of Materials in Nuclear Fuel Assemblies"⁽¹⁾.

This report results from a study of iodine release from uranium dioxide fuel. Expressions derived for the release of iodine were applied to the release of noble gases. Results obtained through use of these expressions are not consistent with those from any previously discussed model for the release of noble gases from a fuel pin under operating conditions.

This analysis of the behavior of fission products in a fuel pin during operation was performed by assuming that the distribution of fission products in a fuel pin is the result of diffusional processes which are controlled by Gibbs-free-energy gradients. Equations are derived which may be used to reproduce the observed distribution of fission products within the fuel and from these concentration equations an equation is derived to relate the fractional release of fission products from a fuel pin to the power rating of the pin. The expression relating release and power rating may be used to relate the decay constant for a given isotope to the release fraction for that isotope. Noble gas release data from the General Electric Company and

from Atomic Energy of Canada Limited are treated to determine the validity of the predicted relationships between release fraction and power rating, and between release fraction and decay constant. The significance of the analysis and the results obtained from the study of the release of noble gases are discussed.

I. ANALYSIS OF THE BEHAVIOR OF FISSION PRODUCTS IN FUEL

The basic assumption in starting this analysis of the release of fission products to the fuel-cladding gap was that fission products are released from the fuel as the result of steady-state equilibrium within the fuel rather than as the result of kinetics. This assumption was based partly on an examination of the results of J. L. Bates' work^(2,3). The general shape of the curves for the concentration of fission products in the fuel, observed by Bates, suggested that these concentrations were produced during steady-state equilibrium.

The assumption that the distribution of fission products is the result of chemical equilibrium rather than a kinetic mechanism implies that the distribution of fission products within the fuel can be described by the correct solution of Equation 1, the equation for the diffusion of isotope i:

$$\frac{dC_i}{dt} = P_i(r,t) - \lambda_i C_i(r,t) + \nabla \cdot \left[\frac{D_i C_i(r,t)}{RT} (\nabla G_i(r,t)) \right], \quad (1)$$

where C_i = concentration of the isotope i

t = time

P_i = rate of production of the isotope per unit volume

λ_i = decay constant of the isotope

r = radius

D_i = diffusivity of isotope i

R = ideal gas constant

T = fuel temperature

G_i = Gibbs-free-energy of the isotope.

The diffusivity, D_i , in Equation 1, which is the result both of surface diffusivity and bulk diffusivity, is an effective diffusivity. Subsequent discussion will show that the important region for diffusion is near the outside edge of the fuel where temperature and, hence, diffusivity are low. The term P_i is assumed to be independent of r for isotopes which do not have long-lived precursors which become redistributed before decaying.

A solution of Equation 1 may be obtained for the steady-state equilibrium situation by setting $\frac{dC}{dt}$ equal to zero. Equation 1 then may be arranged to give for cylindrical geometry:

$$rC_i = \frac{1}{\lambda_i} \left[P_i r + \frac{\partial}{\partial r} \left[r \frac{D_i C_i}{RT} \left(\frac{dG_i}{dr} \right) \right] \right]. \quad (2)$$

The Gibbs-free-energy gradient in Equation 2 may be written as

$$\frac{dG_i}{dr} = \left(\frac{\partial G_i}{\partial C} \right)_T \frac{dC}{dr} + \left(\frac{\partial G_i}{\partial T} \right)_C \beta r \quad (3)$$

where β is defined by Equation 4 in terms of the center-line temperature, T_0 , and T , the temperature at r .

$$T = T_0 + \beta r^2/2. \quad (4)$$

The relationship given in Equation 3 is substituted into Equation 2 and Equation 2 is then solved for the equilibrium concentrations in a cylindrical fuel pin. A solution of the form

$$C_i(r) = \sum a_n \psi_n(r) \quad (5)$$

was assumed. The ψ_n 's are given by

$$\psi_n(r) = \cos \frac{n\pi r^2}{b^2} \quad (6)$$

where b is the radius of the fuel pellet. These solutions predict distributions for the fission products in the fuel very similar to those observed by Bates.

The release of fission products to the fuel-cladding gap may also be calculated from Equation 2 by using a solution of the type given in Equations 5 and 6. The quantity of the fission products, R_{g_i} , released to the gap can be determined by subtracting the quantity in the fuel, $2\pi \int_0^b C_i(r) r dr$, from the total inventory, I_i . If the assumption is made that the production rate is constant across the fuel pin, the amount of fission products in the gap is

$$R_{g_i} = I_i - 2\pi \int_0^b C_i(r) r dr \quad (7)$$

where

$$I_i = \frac{\pi b^2}{\lambda_i} P_i. \quad (8)$$

The use of cosines for $C_i(r)$ in Equation 7 has one important limitation. Any function for concentration based on the summation of cosines will neglect the contribution of concentration to diffusion at the boundary of the fuel because the concentration gradient, $\frac{dC}{dr}$, goes to zero at the boundary. Functions other than cosines must be used for fuel which has small temperature gradients.

The appropriate substitutions in Equation 7 lead to the following expression for the release:

$$R_{g_i} = - \frac{2\pi b^2 \beta D_{ib} C_{ib}}{R \lambda_i T_b} \left(\frac{\partial C_i}{\partial T} \right)_C. \quad (9)$$

The subscript b indicates that the functions are evaluated at the outer edge of the fuel.

II. RELEASE OF NOBLE GASES TO THE FUEL-CLADDING GAP DURING REACTOR OPERATION

A number of mechanisms have been proposed to explain the release of fission gases from fuel pins during reactor operation. A common assumption in all of the proposed models is that once the noble gases are released

from the UO_2 lattice to the center of the fuel pin, they are free to escape to the fuel-cladding gap. Plots of fission gas release, F_i , versus integral power, $\int KdT$, should be relatively smooth according to most of these models; however, Figures 1 and 2 show scatter in such a plot. The extent of this scatter indicates that other variables must affect release.

Several sets of data are treated on the basis of the relationship in Equation 9. Equation 9 is rewritten in the form

$$\frac{1}{2\pi} \frac{Rg_i}{C_{ib}} \frac{R\lambda_i T_b}{b^2 D_{ib}} = - \beta \left(\frac{\partial G_i}{\partial T} \right)_C \quad (10)$$

The rate of release may be related to the fractional release by

$$\frac{Rg_i}{C_{ib} b^2} \simeq \text{Constant } F_i \simeq \ln(1 - F_i) \quad (11)$$

where F = fraction of the isotope released (true for $F_i \ll 1$).

The potential gradient may be related to the integral power by

$$\beta \left(\frac{\partial G_i}{\partial T} \right)_C = \frac{1}{b^2} \int (\int KdT) \quad (12)$$

where $\int KdT$ = integral power rating of the fuel rod. If the two approximate functions in Equations 11 and 12 are substituted into Equation 10, Equation 13 is obtained.

$$\frac{\ln(1-F_i) T_b \lambda_i b^2}{D_{ib}} = \int (\int KdT) \quad (13)$$

Expressions have been derived which can be used to relate the integral power, $\int KdT$, to the fraction, F_i , of an isotope released and to relate the fraction released to the decay constant, λ_i , if the basic assumption used in this analysis is correct. The correctness of the original assumption concerning the importance of the Gibbs-free-energy in the behavior of fission products can be established by proving the equations derived on the basis of that assumption are valid. The validity of the equations derived is tested in the following section by using data from the literature for the release of noble gases.

Fig. 1 Percent Release Versus Integral Power for General Electric Company Data

Fig. 2 Percent Release Versus Integral Power for Atomic Energy of Canada Limited Data

III. ANALYSIS OF NOBLE GAS RELEASE DATA FROM GENERAL ELECTRIC COMPANY

The data from the General Electric Company^(4,5), listed in Table I, were used in Equation 13 to obtain the results shown in Figure 3. The surface temperatures, T_b , were calculated from the center-line temperatures also given by General Electric Company⁽⁶⁾. λ is a constant for the data given by General Electric Company and, therefore, was set equal to one for calculating the points in Figure 3. The form used for D_b was

$$D_b = \text{constant } e^{-Q/RT_b} \quad (14)$$

The heat of activation for diffusion, Q , was set equal to 72 kilocalories for calculating the points in Figure 3. This value is calculated⁽⁷⁾ from the experimental work of Parker⁽⁸⁾.

Table I is a listing of the data and results for Figures 1 and 3. Percent releases calculated from the equation for the least-squares curve drawn in Figure 3 are also listed in Table I. The releases were calculated from the least-squares curve by using the calculated surface temperature and linear power. The points in Figure 3 seem to fall into three distinct groups. The main group includes all points except the five points for burnups of 1890 and 1980 MWd/T and includes pins having four levels of burnup. Release fractions were not calculated for these five points because they appear to be displaced from the main body of data. An important observation may be made for the three points having a burnup of 1890 MWd/T. A line parallel to the least squares line for the main body of data would also describe these three points very well. The objective in calculating the percent release is to show that the curve produced by Equation 13 is sensitive to the release. The wide variation between the main body of data and the points having a burnup of 1890 MWd/T indicates that smaller variations should be anticipated for different burnup sets included in the least squares analysis. This variation undoubtedly accounts for much of the difference between the calculated and observed releases.

TABLE I

GEAP-4314 -- NOBLE GAS RELEASE DATA

Fuel Element	Rod Number	Burnup ^(a) (MWd/T)	Element Radius (cm)	T ₀ (°K)	T _b ^(b) (°K)	$\int_{T_b}^{T_0} KdT^{(a)}$ Average (W/cm)	Observed Release (%)	Calculated ^(c) Release (%)
SI-1.BP	7	1980	0.536	1038	698	13.4	0.31	(d)
	14	1980	0.536	1105	707	15.1	0.67	(d)
SII-2.GP	C	1890	0.631	2255	824	37.4	41.4	(d)
	G	1890	0.631	1788	752	30.8	43.2	(d)
	H	1890	0.631	1705	743	29.4	24.0	(d)
SII-3.GP	A	121	0.635	2955	929	46.9	14.5	18.1
	B	121	0.632	1905	809	31.0	11.1	33.3
	C	121	0.632	1938	776	33.0	4.0	2.4
	D	121	0.635	2261	827	37.5	8.7	5.4
	G	121	0.637	2355	842	38.7	11.7	7.0
	H	121	0.637	2077	797	35.0	6.3	3.2
PWL-6	B	600	0.503	1916	803	31.4	17.1	31.6
	C	600	0.503	1011	644	15.0	11.8	7.9
	D	600	0.503	1016	650	14.9	4.0	13.7
	E	600	0.503	1011	647	14.9	13.5	10.8
DP-1	B	3580	0.632	1444	722	24.0	15.4	7.8
	C	3580	0.632	1594	749	26.6	20.3	10.8
	E	3580	0.632	1677	767	27.7	14.3	17.7
	G	3580	0.632	1599	752	26.5	11.4	13.6
DP-5	A	5000	0.631	1949	812	31.8	9.16	27.7
DP-14	I	5250	0.626	1161	671	18.6	10.22	4.75

(a) Based on 182 MeV/fission.

(b) T_b calculated from method in Reference 6.(c) Calculated from least squares fit of $\ln \left[\int_{T_b}^{T_0} KdT \right] = A + B \left(\int_{T_b}^{T_0} KdT \right) + \left(\int_{T_b}^{T_0} KdT \right)^2$.

(d) These values not used in least square analysis.

Fig. 3 $f(\int KdT)$ Versus Integral Power for General Electric Company Data

IV. ANALYSIS OF NOBLE GAS RELEASE

DATA FROM AECL-1676⁽⁹⁾, -1685⁽¹⁰⁾, -1895⁽¹¹⁾, AND -1952⁽¹²⁾

Table II lists the data plotted in Figure 2 and Figure 4. The treatment of the data for Figure 4 is essentially the same as that for Figure 3. The scatter in Figure 4 is greater than that in Figure 3 but the general trends are much the same as observed for the GEAP-4314 data. Part of the scatter may be because of large uncertainties in the surface temperatures given by AECL. The surface temperature data in AECL-1676 have an uncertainty of about ± 60 degrees. A change of ten degrees in surface temperature would cause a factor of two change in the release for an activation energy of 72 kilocalories. The agreement between points from each set of data in Figure 4 is still excellent.

The data from AECL-1895 are treated separately in Figure 5 and Table III because the different sets of data may have had different variables which were not taken into account. Figure 5 was made by plotting $f(\int KdT)$ against $\int_{T_b}^{T_g} KdT$. T_g is the temperature at which an arbitrary amount of grain growth takes place. The plot was suggested by the values of $\int_{T_b}^{T_g} KdT$ for the P series of pins (Table III).

Examination of Figures 4 and 5 shows that Figure 5 has a better correlation for the data of AECL-1895. The reason why a heat rating based on grain growth temperature gives a better correlation than one based on center-line temperature is not clear. $\int_{T_b}^{T_g} KdT$ may be a better indication of the properties at the outer edge than $\int_{T_b}^{T_0} KdT$ but a more specific explanation is needed. A good discussion of heat rating of fuel pins is given by Bain⁽¹³⁾ and others^(14, 15).

Several important trends should be noted in Figure 5. First, when Equation 13 is used for a single large set of data, an excellent correlation of all the data in the set is obtained. All of the release fractions could be calculated from the least squares fit to within a factor 2 even through the releases vary by a factor of almost 20.

A second interesting trend is apparent from studying the ten points with a surface temperature of approximately 633° Kelvin. If the surface temperature is constant, then the only variable in $f(\int KdT)$ is the

TABLE II
AECL NOBLE GAS RELEASE DATA

AECL Report	Element Number	Burnup ^(a) (MWd/T)	Pellet Radius (cm)	T _b (°K)	$\int_{T_b}^{T_0} KdT$ ^(a)		Observed Release (%)	Calculated ^(b) Release (%)
					Average	(W/cm)		
1676	DFA	386	0.95	708(c)	40.3	4.95	4.95	(d)
	DFB	528	0.95	758	54.7	17.9	17.9	92.8
	DFD	658	0.95	803	68.2	33.0	33.0	55.4
	DFE	794	0.95	850	82.0	40.1	40.1	17.6
	DFH	648	0.95	783	67.8	32.6	32.6	25.4
	DFJ	531	0.95	733	54.0	16.1	16.1	47.8
1685	LI	1417	0.945	617(e)	24.7	0.49	0.49	(d)
	LJ	1920	0.941	645	33.7	4.0	4.0	24.5
	LL	2497	0.942	662	42.4	24.4	24.4	9.3
	LM	2600	0.943	661	42.4	21.9	21.9	8.6
	LO	1715	0.944	617	31.4	4.3	4.3	4.3
1895	E-15	258	1.206	663	46.8	1.2	1.2	1.8
	E-25	258	1.206	663	46.8	2.6	2.6	1.8
	G-1	476	1.132	633	29.4	9.8	9.8	20.8
	G-3	489	1.126	633	29.4	19.8	19.8	21.0
	H-1	514	1.204	653	36.1	12.6	12.6	15.7
	H-2	506	1.204	653	36.1	13.4	13.4	15.7
	H-3	506	1.203	653	36.1	11.7	11.7	15.7
	K-1	2222	1.170	633	32.4	4.0	4.0	8.8
	K-2	2222	1.170	633	32.4	4.2	4.2	8.8
	J-1	2305	1.133	633	29.4	9.0	9.0	20.7
	P-1	8377	1.204	633	35.2	20.1	20.1	3.9
	P-2	8322	1.204	633	35.2	26.8	26.8	3.9
	P-3	8313	1.203	633	35.2	25.3	25.3	3.9
	TT-1	4532	1.198	633	30.0	6.9	6.9	16.1
	TT-3	4532	1.199	633	30.0	8.0	8.0	16.0
1952	LR	2540	0.910	619	31.3	10.6	10.6	5.7
	LS	8280	0.907	616	27.1	11.9	11.9	13.8
	LW	6900	0.909	605	22.7	6.1	6.1	16.6
	LX	10280	0.908	632	34.1	23.3	23.3	8.3
	LY	6270	0.909	582	20.7	2.0	2.0	3.1
	LZ	9060	0.909	611	29.7	19.1	19.1	4.3
	MA	7632	0.908	596	25.2	3.8	3.8	3.6
	MC	9580	0.909	619	32.2	18.0	18.0	4.5

(a) Based on 182 MeV/fission.

(b) Calculated from least squares fit of $\ln \left[\int_{T_b}^{T_0} KdT \right] = A+B \left(\frac{T_0}{T_b} KdT \right) + C \left(\frac{T_0}{T_b} KdT \right)^2$.

(c) T_b's for AECL-1676 are reported to have an uncertainty as large as $\pm 60^\circ\text{C}$. The assumption was made that this uncertainty is constant for these data.

(d) The values of $\int_{T_b}^{T_0} KdT$ for 1676-DFA and 1685-LI were not included in the least square analysis.

(e) Private communication between J. F. Palmer (Chalk River) and V. F. Baston (Idaho Nuclear Corporation) July 1969.

Fig. 4 $f(\int KdT)$ Versus Integral Power for Atomic Energy of Canada Limited

Fig. 5 $f(\int KdT)$ Versus Integral Power for Various Surface Temperatures

TABLE III

AECL-1895 -- NOBLE GAS RELEASE DATA

Element Number	Burnup (MWd/T)	$\int_{T_b}^{T_g} KdT$ (W/cm)	Observed Release (%)	Calculated Release (%)
E-15	258	41.0	1.2	1.8
E-25	258	41.0	2.6	1.8
G-1	476	25.5	9.8	10.0
G-3	489	25.5	19.8	10.1
H-1	514	27.8	12.6	20.0
H-2	506	27.8	13.4	20.0
H-3	506	27.8	11.7	20.0
K-1	2222	29.6	4.0	2.3
K-2	2222	29.6	4.2	2.3
J-1	2305	25.2	9.0	11.2
P-1	8377	23.0	20.1	22.7
P-2	8322	23.0	26.8	22.7
P-3	8313	23.0	25.3	22.7
TT-1	4532	25.7	6.9	8.4
TT-3	4532	25.7	8.0	8.4

release fraction. The important result is that for the constant temperature points in Figure 5, when the surface temperature was held constant and the power level increased, the fraction of fission product gases released was decreased. Such an observation is difficult to reconcile with approaches which consider recrystallization or bubble migration as being the rate controlling step for inert gas release. This observation strongly suggests that a steep thermal gradient may actually inhibit the release of noble gases to the fuel-cladding gap. Another observation which is difficult to explain is how one pin at a power rating of 41 W/cm centimeter, E series of Table II, could have a release of only about 2 percent whereas one with a 27.8 W/cm rating, H series, could have a release of 12 to 13 percent.

V. DISCUSSION

The results of this study suggest that the release of fission gases is, to a significant extent, controlled by the rate at which the gases are able to migrate through the outer layer of the fuel pin during operation. The results further suggest that a high temperature gradient at the outer edges of the fuel may retard the escape of fission gases. These observations are contrary to much current thought but are not without some precedence. W. B. Lewis of Atomic Energy of Canada Limited⁽¹⁶⁾ has suggested that a mechanism consistent with these observations may exist. Lewis discussed the possibility that migration of noble gases in fuel may be a surface phenomenon. Some work performed as a part of this study indicates that a better fit is obtained for some of the data by using a value of Q equal to 40 kilocalories but the results of the study are not conclusive. A value of 40 kilocalories would be more consistent with the mechanism discussed by Lewis than the value of 72 kilocalories obtained from Parker's data. The model proposed also possibly explains Lewis'⁽¹⁷⁾ statement, "As an observed fact, the region most depleted in fission gases is not always the region of highest temperature."

Another important observation made by AECL experimenters was the fractionation of noble gases in fuel rods⁽¹⁸⁾. The experimental results observed at AECL were that xenon fractionates according to atomic mass in operating fuel pins as a result of the thermal gradient being present. Such fractionation would require a process such as a repeated adsorption-desorption of the original gas mixture. If a noble gas were subject to numerous reactions, its passage through the fuel would be hindered. The effect the fuel may have on the escape of noble gases after their release from the lattice is ignored in any release model which considers only the behavior of the fuel in the center of the rod.

Equation 13, derived to describe the equilibrium distribution of fission products in a fuel pin, gives a relationship between the decay constants and the release fractions for the isotopes of a given chemical species. Equation 13 indicates that the product of λ_i and F_i should be the same for all isotopes of a given chemical species. Table IV is a compilation of the release fraction, F_i , and the release fractions multiplied by the decay constant, $\lambda_i F_i$, for various isotopes of xenon, krypton, and iodine from five different fuel pins⁽¹⁹⁾. An examination of the values of $\lambda_i F_i$ for the isotopes of one chemical species shows that the values are about equal for each rod. The scatter in the data is significant in some cases but not unexpected. Long lived isotopes will have a slightly different average history than isotopes with very short half-lives. Other factors will also affect the product of $\lambda_i F_i$. Examples of such factors are the high cross section of xenon-135 for thermal neutrons and the iodine-131 precursor, tellurium-131, with a one-day half-life. In summary, the results in Table IV are not precise enough to be conclusive but when considered with the results summarized in Tables I, II, and III seem to indicate that the general approach is valid.

The correlations between $\int (\lambda_i K_d T)$ and $\int K_d T$ and between λ_i and $\lambda_i F_i$ for different isotopes which were obtained for experimental results by using Equation 13 indicate the existence of a relationship between the fraction of a noble gas released to the fuel-cladding gap and the diffusivity of the gas in the outer regions of the fuel. These correlations indicate that surface diffusion is involved in the release of noble

TABLE IV

RELATIONSHIP BETWEEN DECAY CONSTANT, λ , AND RELEASE FRACTION, F

Fuel Pin	Heat Rating (W/cm)	Variable (a)	Kr-87	Kr-88	Xe-138	Xe-135	Xe-133	I-135	I-133	I-131
CEU	49	$FX10^3$	0.67	1.1	0.24	3.9	38	0.50	1.7	25
		$\lambda FX10^6$	6.1	4.5	9.8	4.9	3.5	0.86	0.93	1.5
CEW	53	$FX10^3$	0.41	0.82	0.16	6.7	115	0.085	0.62	44
		$\lambda FX10^6$	3.7	3.4	6.5	8.4	10.5	0.14	0.34	2.6
CEV	49	$FX10^3$	----	0.79	0.12	1.6	38	----	----	39
		$\lambda FX10^6$	----	3.2	4.9	2.0	3.5	----	----	2.3
CEX	52	$FX10^3$	0.12	0.32	0.043	2.6	59	0.015	0.045	1.4
		$\lambda FX10^6$	1.1	1.3	1.7	3.2	5.4	0.026	0.025	0.084
CEV	25	$FX10^3$	0.035	0.10	0.0076	0.11	0.55	0.00046	0.0029	0.25
		$\lambda FX10^6$	0.32	0.41	0.31	0.14	0.05	0.00079	0.0016	0.015

(a) λ was calculated employing half-lives taken from the Chart of the Nuclides prepared by Battelle Northwest Laboratory for Division of Isotopes Development, U. S. Atomic Energy Commission, May 6, 1969.

gases and surface diffusion has as a driving force an effect related to the temperature gradient in the fuel. This driving force may be the anisotropy of Q mentioned by W. B. Lewis⁽²⁰⁾. The mechanism for release also requires a mechanism for adsorption of the noble gas onto the surface of the UO_2 fuel. The existence of such a redissolution mechanism is discussed by W. B. Lewis⁽²¹⁾, A. M. Ross⁽²²⁾ and M. V. Speight⁽²³⁾.

The slope of the curves in Figures 3, 4 and 5 indicates that the thermal gradients produce a force which hinders the release of noble gases. A loss or reduction of the thermal gradient could allow noble gases to escape from the fuel. The release of noble gases observed from pins during reactor shutdown⁽²⁴⁾ could be accounted for in terms of the loss of the thermal gradient which restrains the noble gases.

Some discrepancy appears in the explanation for noble gas release proposed in this report. Perhaps the most serious anomaly in the proposed model is that such a release mechanism should be very sensitive to the operation history of the reactor. The scatter in the data in Figures 3 and 4 is small in view of the range of burnups listed for each. Possibly the function in Equation 13 has a subtle dependence upon another factor not considered in the analysis. The identity of such a parameter is not obvious at this time.

VI. CONCLUSION

A function was derived which provides a good correlation for existing noble gas release data. The exact mechanism for the release has not been identified but appears to involve both surface and gaseous diffusion. The mechanism may also involve rapid adsorption and desorption from the surface of the fuel grains. The validity of the mathematical correlation can only be established by a more thorough examination of the existing release data. Much of the existing data was taken as part of studies to evaluate other models. For this reason some important parameters such as surface temperature were either not recorded or not reported. In some instances examination of records and discussion with experimenters may provide the necessary information concerning all of the parameters for an evaluation of this function.

The relationships between the release of noble gases and other fission products are not apparent from this study. The function $\left(\frac{\partial G}{\partial T}\right)_C$ will be different for each chemical species. The data in Table IV indicate that iodine is not released as readily as noble gases. This lower relative release for iodine may be due to greater attenuation of iodine within the fuel pin. The only conclusion which can be reached is that a simple multiplier will not relate the fractions of the various chemical species released.

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