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A MODEL FOR FISSION-GAS RELEASE FROM POROUS FUELS IN LOW-PERMEABILITY CONTAINERS

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John W. Prados[1]

ABSTRACT

A simple mathematical model has been developed to describe the steady-state release rate of gaseous fission products from porous ceramic fuels in low-permeability containers. The resulting equations are used to analyze experimental release-rate results obtained from a UC_2-fueled graphite fuel body enclosed in a low-permeability impregnated graphite container. The relative release rates of the fission-product species Kr^{85m}, Kr^{88}, and Xe^{133} were predicted with reasonable success. Absolute-rate predictions were not possible due to lack of information on true permeability and porosity profiles in the graphite container.

INTRODUCTION

The release of fission-product gases from porous ceramic fuels involves the transfer of the products from the solid phase to the open pore structure by diffusion and recoil, and the subsequent flow of the product gases through the pores to the surroundings. For certain fuels, such as UC_2-fueled graphite in which the fuel particles themselves are quite small (160–260 μ)[ref 2] and the matrix porosity rather high (15–20%), the rate of radioactive fission-product release is undesirably large. It has been proposed to enclose the fuel bodies in a container of very low-permeability graphite in order to reduce the release rate of radioactive material without introducing the nuclear disadvantage of a metal cladding. Since the flow

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[1] Consultant from the University of Tennessee.

[2] M. Janes, "Graphite-Matrix Nuclear Fuel Element Development at the National Carbon Company," Proceedings of the Uranium Carbide Meeting Held at the Oak Ridge National Laboratory, December 1–2, 1960, TID-7603, pp. 72–92.

of the fission-product gases would be much slower in the low-permeability container than in the fuel itself, it was hoped that much of the radioactive fission-product gases would have decayed to nonradioactive forms before final release from the container to the surroundings.

The purpose of the present report is to present approximate analytical relationships for predicting the steady-state release rates of various fission-product species from a porous fuel encased in a low-permeability container. Calculations are based on a simplified mathematical model as described in the following section. The results are compared with data reported from in-pile, fission-gas-release tests on a fuel assembly, ORNL-MTR-48-2, which consisted of a cylindrical UC_2-fueled graphite specimen enclosed in a low-permeability graphite container.[2,3]

MATHEMATICAL MODEL EMPLOYED

The calculation of fission-gas-release rates was based upon the following simplifying assumptions:

1. Release of fission products from the fuel particles to the open porosity of the fuel body is instantaneous.
2. Flow in the fuel-body pores is sufficiently rapid that fission-gas composition is uniform throughout the fuel body.
3. Flow of a given component through the low-permeability container is proportional to the gradient of partial pressure (or concentration) of the component. This is equivalent to neglecting viscous flow effects and assuming that transfer through the pores occurs by Knudsen flow alone.
4. The partial pressures of all fission products are zero at the outside of the container.
5. The conversion of fission-product species by neutron absorption can be neglected.
6. Steady-state exists throughout the system.

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[3] GCR Quar. Prog. Rep. June 30, 1960, ORNL-2964, pp. 154-56.

Assumption 1 seems reasonable in the case of UC_2-fueled graphite due to the extremely small size of the fuel particles themselves. If this assumption were invalid, the calculated release rates would be consistently lower than those observed. Assumption 2 will be valid if the low-permeability container can maintain the over-all release rate at such a small value that no appreciable partial pressure gradients are set up in the relatively large pores of the fuel body itself. Assumption 3 should hold for the very fine pore diameters encountered in low-permeability graphite.[4] A continuous sweep of coolant gas over the outside of the container combined with the relatively low over-all release rates should support Assumption 4. Assumption 5 holds for the species[5] Kr^{85m}, Kr^{88}, and Xe^{133}, but fails for Xe^{135}.

MATHEMATICAL RESULTS

The application of the law of conservation of mass for a given fission-product species in the pores of the fuel body yields

$$V_f \epsilon_f \frac{dC_f}{dt} = B - V_f \epsilon_f \lambda C_f - n_f A_f \ , \qquad (1)$$

where

V_f = total volume of fuel body,

ϵ_f = fuel body open porosity, $\frac{\text{open pore volume}}{\text{total fuel body volume}}$,

C_f = concentration of a given fission-product species in the pores, moles/pore volume,

B = production rate of species, moles/time,

λ = decay constant for species, time^{-1},

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[4] L. W. Graham, J. G. Campbell, and D. R. Perels, "The Development and Production of Impermeable Graphite for the High-Temperature Gas-Cooled Reactor," Paper presented at the Project Dragon Graphite Symposium, Durley Hall, Bournemouth, England, November 16–19, 1959.

[5] J. O. Blomeke and M. F. Todd, Uranium-235 Fission Product Production as a Function of Thermal Neutron Flux, Irradiation Time, and Decay Time, Part 1, ORNL-2127 (August 19, 1957), pp. 27, 35.

n_f = molar flux of species from fuel body into container, moles/area-time,

A_f = contact area between fuel body and container, and

t = time.

At steady-state, the rate of production of a given species in the fuel equals its conbined rate of loss by decay and flow into the container; hence, the concentration, C_f, becomes constant, and its time derivative vanishes.

The combined processes of Knudsen flow and decay in the container can be described by the partial differential equation

$$\epsilon\frac{\partial C}{\partial t} = K\nabla^2 C - \epsilon\lambda C. \tag{2}$$

If the container is thin enough in the direction of diffusion to approximate a flat-slab geometry, Eq. 2 reduces to

$$\epsilon\frac{\partial C}{\partial t} = K\frac{\partial^2 C}{\partial X^2} = - \epsilon\lambda C, \tag{3}$$

where

ϵ = open porosity of low-permeability container,

C = concentration of species in pores of container,

K = permeability coefficient for Knudsen flow $(\text{length})^2/\text{time}$, and

X = length coordinate in direction of flow.

At steady-state, the time derivative vanishes, leaving the ordinary differential equation

$$\frac{d^2 C}{dX^2} - \frac{C}{L^2} = 0, \tag{4}$$

where

$$L = \sqrt{\frac{K}{\lambda_\epsilon}} \quad .$$

This equation can be solved subject to the boundary conditions, $C(0) = C_f$ and $C(\delta) = 0$, where δ is the thickness of the container. Since, by the

Knudsen flow equation, the flux, n, at any point in the container is given by[6]

$$n = -K \frac{\partial C}{\partial X}, \tag{5}$$

one may combine Eqs. 1 and 5 together with the solution to Eq. 4 to eliminate C_f and arrive at the expression for R, the rate of release of a given fission-product species from the outer container boundary in moles per unit time.

$$R = \frac{B}{\frac{\epsilon_f V_f}{\epsilon A_f \delta} (\frac{\delta}{L}) \sinh \frac{\delta}{L} + \cosh \frac{\delta}{L}} . \tag{6}$$

This relation is frequently expressed in terms of the ratio of steady-state release rate to production rate:

$$\frac{R}{B} = \frac{1}{\frac{\epsilon_f V_f}{\epsilon A_f \delta} (\frac{\delta}{L}) \sinh \frac{\delta}{L} + \cosh \frac{\delta}{L}} . \tag{7}$$

It is of interest to note that the character of Eq. 7 is determined by the magnitude of the parameter, $\frac{\delta}{L}$. From the definition of L, one sees that

$$\frac{\delta}{L} = \sqrt{\frac{\delta \epsilon \lambda}{(\frac{K}{\delta})}} , \tag{8}$$

and that $\frac{\delta}{L}$ is then a measure of the relative rates of decay and flow through the sleeve. This is borne out by Eq. 7 since for large $\frac{\delta}{L}$ (decay rapid relative to flow) $\frac{R}{B}$ will be small while for very small $\frac{\delta}{L}$, $\frac{R}{B}$ approaches unity.

Note further that for $\frac{\delta}{L}$ less than about 0.1 $\sinh \frac{\delta}{L} \rightarrow \frac{\delta}{L}$, $\cosh \frac{\delta}{L} \rightarrow 1$, and Eq. 7 becomes

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[6] P. C. Carman, Flow of Gases Through Porous Media, pp. 3, 69, Academic Press, New York, 1956.

$$\frac{R}{B} \cong \frac{1}{\frac{\epsilon_f V_f \delta}{\epsilon A_f L^2} + 1}$$

or

$$\frac{R}{B} = \frac{1}{\frac{\epsilon_f V_f \lambda}{A_f (\frac{K}{\delta})} + 1} \; . \qquad (9)$$

In Eq. 9, the parameter of primary interest is the ratio of $\frac{K}{\delta}$, simply that of the container's permeability to its thickness. This is the parameter governing flow without decay through a material of low permeability. It should be emphasized, however, that in the general case of Knudsen flow (or diffusion) accompanied by decay, the parameter $\frac{\delta}{L}$ rather than $(\frac{K}{\delta})$ determines the ratio of release to production rate for a given geometry. Hence, the $\frac{R}{B}$ ratio would be lower for a thick container of higher permeability than for a thin container of lower permeability even though the $\frac{K}{\delta}$ ratios were the same in both cases.

Low-permeability graphite containers are usually manufactured by an impregnation process which leaves thin layers of very low-permeability material on the inner and outer surfaces of the container, while much higher permeability exists in the bulk of the container.[4,7] In analyzing the diffusional process through such a system, it is assumed that the container is bounded by inner and outer thin layers of approximately flat-slab geometry and that the internal bulk of the container offers negligible resistance to diffusion. Under these conditions, the material balance, Eq. 1, is unchanged; Eq. 4 must be solved for both inner and outer coatings, and a material balance similar to Eq. 1 must be set up for the bulk of the container. At steady-state, the release to generation rate ratio, $\frac{R}{B}$, is given by

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[7] E. Fitzer and K. W. F. Etzel, "Contributions to the Development of Fine-Grain Graphite of Low Gas Permeability," Paper presented at the Project Dragon Graphite Symposium, Durley Hall, Bournemouth, England, November 16–19, 1959.

$$\frac{R}{B} = \frac{1}{\left[\frac{\epsilon_f V_f}{\epsilon A_f \delta}\left(\frac{\delta}{L}\right) \sinh \frac{\delta}{L} + \cosh \frac{\delta}{L}\right]\left[\frac{\epsilon_s V_s}{\epsilon A_o \delta}\left(\frac{\delta}{L}\right) \sinh \frac{\delta}{L} + \Bigl(1 + \frac{A_f}{A_o} \cosh \frac{\delta}{L}\right] - \frac{A_f}{A_o}}, \tag{10}$$

where

V_s = total volume of container material,

ϵ_s = open porosity of container, and

A_o = outer surface area of container.

In deriving Eq. 10, it has been assumed that the porosity and thickness of the low-permeability layer are the same for both inner and outer surfaces.

The limiting form assumed by Eq. 10 at small values of $\frac{\delta}{L}$ (< 0.1) is:

$$\frac{R}{B} = \frac{1}{\left[\frac{V_f \epsilon_f \lambda \delta}{A_f K} + 1\right]\left[\frac{V_s \epsilon_s \lambda \delta}{A_o K} + 1 + \frac{A_f}{A_o}\right] - \frac{A_f}{A_o}} . \tag{11}$$

It will be noted that Eqs. 10 and 11 both show the proper asymptotic behavior; that is, as $\frac{\delta}{L} \to 0$, $\frac{R}{B} \to 1$; and as $\frac{\delta}{L} \to \infty$, $\frac{R}{B} \to 0$.

In applying Eqs. 10 and 11 to analyze the results obtained from fuel assembly, ORNL-MTR-48-2, it was assumed that the open porosity was the same for both container bulk material and fuel. Geometrical information was obtained from the appropriate progress report.[3]

COMPARISON WITH EXPERIMENTAL RESULTS

In order to compare measured values of the release ratios, $\frac{R}{B}$, with those predicted from Eqs. 10 and 11, it is necessary to have reasonable values for the parameters δ, ϵ, ϵ_s, and K. This, in turn, requires permeability and porosity profiles on fuel and container, which were not available for the fuel assembly under consideration. However, prior estimates indicated that the ratio $\frac{\delta}{L}$ was probably well below 0.1 for all isotopes considered, hence, permitting the use of Eq. 11. For the limiting case of Eq. 11, one sees that the release ratio $\frac{R}{B}$ depends only on the parameters V, A, λ, ϵ_f and $\frac{K}{\delta}$. Of these, V, A, and λ are known with confidence, ϵ_f may be estimated fairly well, and only $\frac{K}{\delta}$ is unknown. Hence, it was decided to calculate $\left(\frac{K}{\delta}\right)$ values from Eq. 11 using the experimentally measured values of $\frac{R}{B}$ for the various isotopes. Agreement between $\frac{K}{\delta}$ values for different isotopes would provide reasonable support for the postulated release model.

Results of $\frac{R}{B}$ measurements[2,8] from fuel assembly ORNL-MTR-48-2 together with $\frac{K}{\delta}$ values calculated from Eq. 11 are presented in Table 1. Only results

Table 1. Calculated Results[a,b] from ORNL-MTR-48-2

Isotope	Measured[a,b] $\frac{R}{B}$ x 100	$\left(\frac{K}{\delta}\right)$ x 10^7 cm/sec, from Eq. 11	$\left(\frac{K}{\delta}\right)$ average x 10^7 cm/sec
Kr^{85m}	0.31	2.41	2.50
Kr^{85m}	0.49	3.08	
Kr^{85m}	0.30	2.37	
Kr^{85m}	0.25	2.16	
Kr^{88}	0.17	2.76	2.64
Kr^{88}	0.18	2.84	
Kr^{88}	0.12	2.31	
Xe^{133}	17	1.05	1.57
Xe^{133}	35	2.45	
Xe^{133}	20	1.21	

[a] M. Janes, "Graphite-Matrix Nuclear Fuel Element Development at the National Carbon Company," <u>Proceedings of the Uranium Carbide Meeting Held at the Oak Ridge National Laboratory, December 1–2, 1960</u>, TID-7603, pp. 72–92.

[b] <u>GCR Quar. Prog. Rep. Dec. 31, 1960</u>, ORNL-3049, pp. 246–47.

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[8] <u>GCR Quar. Prog. Rep. Dec. 31, 1960</u>, ORNL-3049, pp. 246–47.

obtained after the system had apparently reached steady-state were used in the comparison. Two abnormally low values were rejected. An average value of $(\frac{K}{\delta})$ was calculated for each isotope. These $\frac{K}{\delta}$ values were used along with reasonable values for ϵ and δ to estimate the maximum value of $\frac{\delta}{L}$. This was found to be about 0.05, indicating that the approximation of Eq. 10 by Eq. 11 was justified.

Although the average $\frac{K}{\delta}$ value for Xe^{133} is somewhat lower than those of the other two species, the discrepancy is not as serious as it might first appear. From the theoretical relations leading to the Knudsen flow equations, one would expect the permeability, K, to be inversely proportional to the square root of the molecular weight, M_i, of the flowing gas.[5] If one arbitrarily takes the average value of $(\frac{K}{\delta})$ for Kr^{85m} as a "standard," he may then estimate the corresponding values for Kr^{88} and Xe^{133} by a simple ratio of the square roots of the molecular weights,

$$(\frac{K}{\delta})_i = (\frac{K}{\delta})_{Kr^{85m}}\sqrt{\frac{85}{M_i}} \ . \qquad (12)$$

Predicted values for $\frac{R}{B}$ can then be back-calculated from Eq. 11 from these computed $\frac{K}{\delta}$ values. The results of such a calculation are shown below in Table 2.

Table 2. Predicted $\frac{R}{B}$ Ratios Based on Average $\frac{K}{\delta}$ for Kr^{85m}

Isotope	$(\frac{K}{\delta})$ [Based on Kr^{85m}] x 10^7 cm/sec	$(\frac{R}{B})$ [Predicted from Kr^{85m} value] x 100
Kr^{85m}	2.50	0.33
Kr^{88}	2.46	0.14
Xe^{133}	2.00	32

It should be noted that the proposed multiregion container would give significantly better correlation of the results than would the assumption of a container material of uniform permeability and porosity throughout. This is illustrated by calculations based on a single-region container, using

Eq. 7. Release-rate ratios were calculated from the observed Kr^{85m} value and are compared in Table 3 with those observed and with those computed from the previously developed multiregion model.

Table 3. Release-Rate Ratios Observed and Calculated from Two Models

	$\frac{R}{B} \times 100$		
Isotope	Average value observed	Calculated from recommended model (Eqs. 10 and 11)	Calculated assuming uniform container (Eq. 7)
Kr^{85m}	0.35	0.33	0.34
Kr^{88}	0.16	0.14	0.056
Xe^{133}	24	32	45

Relative values of the release-rate ratios calculated from Eqs. 10 and 11 (multiregion container) are seen to be in considerably better agreement with those observed than are the ratios calculated from Eq. 7 (single-region container).

DISCUSSION

If the calculated $\frac{R}{B}$ values from Table 2 are compared with the experimental values of Table 1, it will be seen that the value for the Kr^{88} lies at about the mean of the experimental values, while that for Xe^{133} lies above the mean but within the experimental spread. It was felt that the small number of available data points did not justify a statistical test for the significance of this deviation. On the surface, it appears that the model represents the experimental behavior with reasonable accuracy, but considerably more voluminous experimental results are needed before definite conclusions on the model's validity can be drawn. Of course, an absolute test of validity will require independent measurements of K, δ, and ϵ, which were lacking from the present study. These cannot be computed from over-all gas transmission measurements through the container.

For deriving more precise mathematical models to describe fission-gas release through coated fuels or for using the present models in systems where the asymptotic relations ($\frac{\delta}{L} < 0.1$) leading to Eq. 11 do not hold, one needs information on the premeability and open-porosity profiles for fuel, container bulk, and coating. In these cases, the parameter $\frac{K}{\delta}$ alone does not determine the release-rate ratio. It is strongly recommended that the following data be obtained for any fuel-sleeve assembly under evaluation:

1. Open porosity and permeability of the fuel, and
2. Open porosity and permeability of the container as a function of position (e.g., as a function of radius for a cylindrical sleeve).

Such information will permit calculation of release-rate ratios from the general Eq. 10 for the present model, and will be of considerable use in setting up more exact methods of calculation should the present method prove inadequate.

SUMMARY OF CONCLUSIONS

An approximate mathematical model has been developed to describe the release of fission-product gases from a porous fuel through a low-permeability container. The model shows reasonable internal consistency based on comparison of relative release rates of different isotopic species predicted by the model and observed from an experimental fuel assembly.

In order to provide an absolute test of the model's validity and to permit extension of results to more general classes of fuel assemblies, it will be necessary to obtain accurate information on permeability and open-porosity profiles on fuel and container specimens of interest. An experimental program to develop such information is recommended.

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