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DIFFUSION OF RADIOACTIVE GASES
THROUGH POWER REACTOR GRAPHITE

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INTRODUCTION

This calculation of gaseous diffusion through graphite was done in connection with a gas-cooled reactor design problem as described by Wm. B. Cottrell, Reactor Projects Division, ORNL, to A. S. Berman, Flow Research Department, Technical Division, ORGDP.

The problem arose from consideration of the feasibility of using unclad fuel elements. A cross section of such an element, in the shape of a right circular cylinder, is shown schematically in figure 1. The coolant in the primary flow system outside the graphite shell is to be maintained at a specified low radiation level by forcing a small portion (the "sweep flow") of the primary flow inward through the shell, thus retarding the outward diffusion of radioactive fission products, diluting such products in the annulus between the concentric shell and core, and removing them to a disposal unit.

Figure 1 - Cross Section of Cylindrical Fuel Element (Schematic)

The present study is concerned only with the problem of retarding the diffusion of gaseous fission products through the shell. It seeks to point out the pertinent parameters, their relationship to the properties of the graphite and the gas coolant, and the effect of their variation on the concentration in the primary system and the diffusion rate into the primary system of the fission products.

SUMMARY

An approximate differential equation is derived which relates the concentration and diffusion of gaseous fission products to gas and graphite properties. The fission products are assumed, for simplicity, to consist solely of krypton, and the coolant is taken to be helium at 1000°F. and $p_e = 21$ atmospheres.

In applying the solution of the equation to the present problem, use is made of the flow parameters, B_0 and K_0 , and the porosity, ϵ , which were measured in a previous experimental investigation of the flow of helium through a fine-pore graphite⁶. These parameters are used with the mutual diffusion coefficient D_{12} for a krypton-helium mixture to estimate the effective coefficient D_1 for diffusion of krypton through the graphite pores and through the helium contained therein.

The fraction, θ , of the generated radioactive krypton which escapes into the primary system and the mole fraction of krypton, N_e , at the external surface of the graphite are computed for the graphite used in the above-mentioned experimental investigation as well as for similar graphite differing only in pore size. The sensitivity of these results to the estimated effective diffusion coefficient D_1 is also computed.

The main results of these computations are:

- (1) The role of the empirical parameters, ϵ , B_0 , and K_0 , and their relationship to hypothetical parameters, (associated with a particular model for porous media), such as the mean hydraulic pore radius, m , and the tortuosity, L_e/L , are illustrated.
- (2) The lack of information needed for accurate estimation of D_1 prevents reliable calculation of the rate of escape of gaseous fission products into the primary system and of the concentration of radioactivity so produced. However, a conservative estimate of D_1 is made and used.
- (3) The desirable upper limits of θ and N_e , taken to be 10^{-5} and 10^{-10} , respectively, are found to be incompatible with each other if the present conservative estimate of D_1 is used with the graphite properties indicated above⁶. It is conceivable that either a more favorable value of D_1 or the development of improved graphite could remedy this situation.

A more detailed summary of results is given on pages 13-15 where reference is made to figures 2, 3, and 4.

THE CONVECTION-DIFFUSION EQUATION

The problem of gaseous diffusion in flow passages so small that gas-wall as well as gas-gas intermolecular collisions contribute to the diffusion resistance has been treated in the classified literature dealing with the gaseous diffusion process for the separation of uranium isotopes. Much of this work has been summarized and reviewed^{1*}. Related work of an unclassified nature has also been reported by Pollard and Present², Present and deBethune³, and Visner⁴.

The method used here is derived from these sources with the additional consideration of the effect of pressure diffusion⁵ which seems to have been neglected** in previous work.

Although some uncertainty (and to our knowledge no experimental information!) exists concerning the applicability of the theory of pressure diffusion to the present problem, we attempt to take the effect into account because it does represent an additional possible mechanism which enhances the rate of escape of fission products into the primary system.

Consider a gas stream, consisting of a mixture of components designated by the subscripts 1 and 2, in which a total pressure gradient as well as partial pressure gradients exist. Equations (23) of reference 1, p. 82, as modified to take pressure diffusion into account***, can be expressed as

$$\vec{G}_1 = N_1 \vec{G}^T - D_1 \vec{a}_{12} \quad (1)$$

$$\vec{G}_2 = N_2 \vec{G}^T - D_2 \vec{a}_{21} \quad (2)$$

whence, by adding and solving for $\vec{G}^T$,

$$\vec{G}^T = \vec{G} + D_1 \vec{a}_{12} + D_2 \vec{a}_{21} \quad (3)$$

where $\vec{G}_1$ and $\vec{G}_2$ are the molar flux vectors ($\frac{\text{moles}}{\text{cm}^2 \text{ sec.}}$) for the two

* Bibliographic references are listed on page 16.

** The effect of pressure diffusion becomes appreciable for gas mixtures with components of widely different molecular weights. This effect refers to the tendency of the heavier molecules to diffuse in the direction of rising pressure.

*** See footnote, p. 14.

components, $\vec{G}$ is the resultant total flow; and $-D_1 \vec{d}_{12}$, $-D_2 \vec{d}_{21}$ are the respective "diffusive" fluxes which are superimposed on the "convective" fluxes, $N_1 \vec{G}$, $N_2 \vec{G}$. Here, $\vec{d}_{12}$ and $\vec{d}_{21}$ are effective "driving forces" for the diffusion process, when no thermal diffusion occurs.

$$\vec{d}_{12} = \vec{\nabla} n_1 - \frac{N_1 M_1}{N_1 M_1 + N_2 M_2} \vec{\nabla} n = n \vec{\nabla} N_1 + \frac{n_1 n_2}{p} (M_2 - M_1) \vec{\nabla} \ln p \quad (4)$$

where n_1 and n_2 are partial molar densities, n is the total molar density, N_1 and N_2 are mole fractions, M_1 and M_2 molecular weights, p is the total density of the mixture, and p is the corresponding* pressure.

D_1 and D_2 are generalized diffusion coefficients which, in the limit of zero pressure (or small flow channels), approach the free-molecule or Knudsen values D_{k1} and D_{k2} . At high pressures (or for large flow passages) they both approach the value D_{12} . The significance of equations (1), (2), and (3) is clarified by considering their limiting forms for a porous material. With the ratio of mean free paths for gas-wall and gas-gas collisions denoted by m/λ , the following table shows the limiting values approached by the terms in these equations.

Variable in Equations (1),(2),(3)	Limiting Value	
	$(m/\lambda) \rightarrow 0$	$(m/\lambda) \rightarrow \infty$
D_1	D_{k1}	D_{12}
D_2	D_{k2}	D_{12}
$\vec{G}$	0	$\vec{G}$

In proceeding to obtain a differential equation in the mole fraction, N_1 , a considerable simplification results from taking gas 1 to be present in trace quantities**. Then,

$$\vec{d}_{12} \simeq n \vec{\nabla} N_1 - N_1 \left(\frac{M_1}{M_2} - 1 \right) \vec{\nabla} n$$

so that on substituting $\vec{G}$ from equation (3) into equation (1) and neglecting terms of order N_1^2 or $N_1 \vec{\nabla} N_1$, there is obtained

* $p = n_1 M_1 + n_2 M_2$, $p = nRT$.

** This is a well justified approximation here since we will later set N_1 equal to the mole fraction of gaseous fission products in the "sweep flow" and will find that, inside the annulus, this quantity is of the order of a part per million.

$$\vec{G}_1 = N_1 \left[\vec{G} + D_1 \left(\frac{M_1}{M_2} - 1 \right) \vec{\nabla} n \right] - n D_1 \vec{\nabla} N_1. \quad (5)$$

If $M_1 = M_2$ and $D_1 = D_{12}$, this equation becomes identical with the more familiar one which is valid at high pressures in the absence of a total pressure gradient.

In applying equation (5) to the flow through the graphite cylindrical shell illustrated in figure 1, it is assumed that $\vec{\nabla} n$ and $\vec{\nabla} N_1$ have radial components only (i.e., $\vec{\nabla}$ is replaced by d/dr) and that $\vec{G}$ and $\vec{G}_1$ as well as N_1 are functions of the radial coordinate, r , only. Then, defining F_1 as the net outflow of radioactive gas and F as the "sweep flow" of inert carrier gas in moles per second, equation (5) can be written as

$$\Gamma_1 = g N_1 - \frac{dN_1}{dz} \quad (6)$$

where $z = \ln r$

$$g = \Gamma + \left(\frac{M_1}{M_2} - 1 \right) \frac{1}{p} \frac{dp}{dz}$$

$$\Gamma = F / 2\pi L n D_1$$

$$\Gamma_1 = F_1 / 2\pi L n D_1$$

L = length of cylindrical shell

p = gas pressure in shell

It will be seen that the coefficient g in equation (6) varies only slightly with r through the shell; so it can be replaced by an average value appropriate to the pressure conditions which will be assumed:

$$\bar{g} = \frac{F}{2\pi L n \bar{D}_1} + \left(\frac{M_1}{M_2} - 1 \right) \frac{1}{\bar{p}} \frac{\Delta p}{\Delta z} \quad (6-a)$$

where $\Delta p = p_e - p_i$, $\Delta z = z_e - z_i = \ln \frac{r_e}{r_i}$,

the subscripts e and i denoting, respectively, conditions at the exterior and interior surfaces of the shell. Similarly, Γ_1 can be replaced by

$$\bar{\Gamma}_1 = \frac{F_1}{2\pi L n \bar{D}_1}. \quad (6-b)$$

With these approximations to g and Γ_1 , the solution of equation (6) is

$$N_1(z) = \frac{\bar{\Gamma}_1}{\bar{g}} + C e^{\bar{g}z}. \quad (7)$$

The constant of integration, C , is evaluated by specifying that

$$N_1(z_1) = N_1. \quad (7-a)$$

Now let the generation rate of gaseous fission products in the core of the fuel element be $-F_g$ (moles/sec.), and assume that all of this flows into the annulus and is completely mixed with the carrier gas there so that $N_1 = N_i$ everywhere in the annulus. (The minus sign used with F_g is consistent with the sign convention to be used in a material balance for the annulus; i.e., outward flows from the annulus are taken to be positive). Also let $\theta = F_1 / -F_g$ so that θ denotes the fraction of the gaseous fission products generated in the core which escapes through the shell. Then, denoting $N_1(z_e)$ by N_e gives, from equation (7) and the boundary condition (7-a),

$$\frac{N_e}{N_i} = e^{\bar{g}\Delta z} - \theta \frac{\bar{\Gamma}_1}{\bar{g}} (1 - e^{\bar{g}\Delta z}). \quad (8)$$

For a shell of specified size, equation (8) gives the ratio of external to internal radioactive mole fractions if the pertinent flow properties of the carrier (i.e., coolant) gas and of the shell material are available for the calculation of $\bar{g}$ and $\bar{\Gamma}_1$.

In applying equation (8) to a reactor design problem, N_e and θ might be specified on the basis of personnel safety or other considerations, and the resulting relationship among N_i , $\bar{g}$, $\bar{\Gamma}_1$, and Δz could be used as an aid in specifying desirable coolant and shell properties.

An additional relationship among N_i and the flows F_g , F_1 , and F is furnished by the material balance for the annulus. Thus, if F_o is the flow from the annulus to the disposal unit (figure 1), conservation of total material requires that

$$F_g + F_1 + F + F_o = 0,$$

while the conservation of radioactive component gives

$$F_g + F_l + N_i F_o = 0.$$

Eliminating F_o from these two equations and neglecting the small quantities F_g and F_l when they are added to F gives $N_i F = F_g + F_l$ whence, since $F_l = -\theta F_g$,

$$N_i = (1 - \theta) \frac{F_g}{F}. \quad (9)$$

Using this result to replace N_i in equation (8) gives

$$N_e = \frac{F_g}{F} \left\{ e^{\frac{\bar{g}\Delta z}{\bar{F}}} - \theta \left[\frac{\bar{F}}{\bar{g}} (1 - e^{\frac{\bar{g}\Delta z}{\bar{F}}}) + e^{\frac{\bar{g}\Delta z}{\bar{F}}} \right] + \theta^2 \frac{\bar{F}}{\bar{g}} (1 - e^{\frac{\bar{g}\Delta z}{\bar{F}}}) \right\} \quad (10)$$

CALCULATION OF SWEEP FLOW AND DIFFUSION RATES

Two limiting forms of equation (10) are of interest and lead to simplified computations. If it is required that there be no outflow of radioactive gas, then $\theta = 0$, and

$$N_e = \frac{F_g}{F} e^{\frac{\bar{g}\Delta z}{\bar{F}}}. \quad (10-a)$$

If, on the other hand, such radioactive gas as does escape into the primary system is diluted to essentially zero external concentration, then $N_e = 0$, and solving equation (10) for θ gives that either $\theta = 1$, or

$$\theta = \frac{\bar{g}}{\bar{F}} \frac{e^{\frac{\bar{g}\Delta z}{\bar{F}}}}{1 - e^{\frac{\bar{g}\Delta z}{\bar{F}}}}. \quad (10-b)$$

The foregoing results will be applied in the following manner.

- (1) The escape ratio, θ , will be assigned some desirable low value, and the external mole fraction, N_e , as well as the sweep flow, F , will be calculated for various graphite properties and pressure conditions. Equation (10-a) can be used if

$$\theta \left[1 + \frac{\bar{F}}{\bar{g}} \frac{1 - e^{\frac{\bar{g}\Delta z}{\bar{F}}}}{e^{\frac{\bar{g}\Delta z}{\bar{F}}}} \right] \ll 1. \quad (10-a')$$

- (2) The external mole fraction, N_e , will be assigned some desirable low value* and values of θ will be calculated. Equation (10-b) can be used if

$$\frac{FN_e}{F_g} e^{-\bar{g}\Delta z} \ll 1. \quad (10-b')$$

Evaluation of the Flow and Diffusion Parameters

The parameters $\bar{g}$ and $\bar{\Gamma}$ have been defined, in connection with equations (6), (6-a), and (6-b), in terms of certain flow rates and a generalized diffusion coefficient, D_1 . The flow of coolant gas through the graphite shell, as a function of pressure conditions, is given by the following equation⁶:

$$F \left(\frac{\text{moles}}{\text{sec.}} \right) = -K \left(\frac{\text{cm.}^2}{\text{sec.}} \right) \times \text{area} (\text{cm.}^2) \times \frac{dn}{dr} \left(\frac{\text{moles/cm.}^3}{\text{cm.}} \right)$$

or, replacing "area" by $2\pi Lr$ and $r \frac{dn}{dr}$ by $\frac{\Delta n}{\Delta z}$,

$$\frac{F}{2\pi L} = -\bar{K} \frac{\Delta n}{\Delta z}, \quad (11)$$

where $\bar{K}$ can be expressed in terms of empirical parameters, B_0 and K_0 , which are independent of the gas, by

$$\bar{K} = \frac{B_0 (\text{cm.}^2)}{\eta (\text{poise})} P \left(\frac{\text{dynes}}{\text{cm.}^2} \right) + \frac{4}{3} K_0 (\text{cm.}) \bar{v} \left(\frac{\text{cm.}}{\text{sec.}} \right) \quad (12)$$

where $\bar{v}$ is the mean molecular speed of the gas and η its viscosity.

Hence, the parameters needed to apply equation (10) are given by

$$\bar{\Gamma} = \frac{\bar{K}}{nD_1} \frac{\Delta n}{\Delta z} = - \frac{\bar{K}}{pD_1} \frac{\Delta p}{\Delta z} \quad (13)$$

* Whether or not any specified value of N_e can be realized in a steady state depends on the material balance and mixing conditions for the primary flow system. The pertinent calculations will not be done here.

and

$$\bar{g} = \left(-\frac{\bar{K}}{\bar{D}_1} + \frac{M_1}{M_2} - 1 \right) \frac{1}{\bar{p}} \frac{\Delta p}{\Delta z} \quad (14)$$

in terms of the flow coefficient, $\bar{K}$, and diffusion coefficient, $\bar{D}_1$.

Since neither a rigorous theoretical formulation nor suitable experimental values of D_1 are available in the case of diffusion through fine porous media of gases with widely different molecular weights, this quantity is estimated in the manner suggested by Carman⁷. Thus,

$$\frac{1}{D_1} = \frac{q}{\epsilon D_{12}} + \frac{3}{4K_o \bar{v}_1} = \frac{q}{\epsilon \bar{p} D_{12}} \bar{p} + \frac{3}{4K_o \bar{v}_1}, \quad (15)$$

where D_{12} is the "high pressure" coefficient of mutual diffusion ($\bar{p} D_{12}$ is independent of $\bar{p}$), ϵ is the porosity or void fraction of the graphite, and q is a tortuosity factor. The appropriate value of q is uncertain, and the most "pessimistic" (in the sense that a high value of D_1 is produced) of the possibilities⁸ mentioned by Carman is used here. Thus, q will be equated to (L_e/L) where the latter quantity as well as ϵ and K_o are evaluated by Hutcheon⁶ et al. in the paper previously cited in connection with equations (11) and (12).

Numerical Examples

On the basis of the capillary model of porous media which is employed by Hutcheon and by Carman, they relate* the empirical flow parameters B_o and K_o of equation (12) to a "mean pore size", m , and the "tortuosity factor", q , by the equations

$$B_o = \frac{\epsilon m^2}{2.5 q^2} \quad \text{and} \quad K_o = \frac{0.59}{q^2} \epsilon m. \quad (16)$$

In the numerical examples which will now be given, we use Hutcheon's value of q^2 , as obtained from his experimental determination of B_o and K_o for the flow of helium through a "fine-pore commercial graphite", as well as his value of ϵ as obtained from density measurements in helium and in mercury. However, in addition to making calculations with his value of the "pore size", m , we also show the effect of doubling and of halving this value.

* See page 269 of reference 6.

Also, while retaining Hutcheon's values of the flow parameters, we will arbitrarily vary the diffusion coefficient, D_1 , in order to illustrate the sensitivity of the results to changes in this parameter.

The working formulas and computation procedure can be summarized as follows:

$$\textcircled{1} \quad (12)^* \quad \bar{K} = A\bar{p} + B \text{ with } \bar{p} \text{ in atmospheres.}$$

$$A = 1.013 \times 10^6 \left(\frac{\text{dynes/cm.}^2}{\text{atmosphere}} \right) \frac{B_0}{\eta},$$

$$B = \frac{4}{3} K_0 (\text{cm.}) \bar{v}_2 \left(\frac{\text{cm.}}{\text{sec.}} \right)$$

$$\textcircled{2} \quad \begin{matrix} (15), \\ (16) \end{matrix} \quad \frac{1}{\bar{D}_1} = C\bar{p} + D$$

$$C = \frac{q}{\epsilon(\bar{p}D_{12})},$$

$$D = \frac{3}{4K_0 \bar{v}_1}$$

$$\textcircled{3} \quad (14) \quad \bar{g}\Delta z = (E - \textcircled{1}\textcircled{2}) \frac{\Delta p}{\bar{p}}$$

$$E = \frac{M_1}{M_2} - 1$$

$$\textcircled{4} \quad (11) \quad -F \left(\frac{\text{moles}}{\text{sec.}} \right) = H \textcircled{1} \frac{\Delta p}{\Delta z}$$

$$H = \frac{2\pi L(\text{cm.})}{RT \left(\frac{\text{atm. cm.}^3}{\text{mole}} \right)}$$

$$\textcircled{5} \quad (10-a) \quad N_e = I \exp \textcircled{3} \quad \text{for } \theta < 10^{-5}$$

$$I = \frac{-Fg}{\textcircled{4}}$$

* Numbers in parentheses indicate the equations from which the formulas are obtained.

$$\textcircled{6} \quad (10-b) \quad \theta = J \frac{\exp \textcircled{3}}{1 - \exp \textcircled{3}} \quad \text{for } N_e < 10^{-10}$$

$$J = \frac{E - \textcircled{1}\textcircled{2}}{-\textcircled{1}\textcircled{2}}$$

The calculations were carried out for helium at $T = 811^\circ \text{K.} (1000^\circ \text{F.})$ with the external pressure fixed at $p_e = 21$ atmospheres. To simplify the calculations, all of the gaseous fission products were replaced by an equivalent amount* of krypton.

The values used** for the pertinent properties of krypton (subscript 1) and helium (subscript 2) are as follows:

$$\bar{v}_1 = 4.525 \times 10^4 \frac{\text{cm.}}{\text{sec.}}$$

$$M_1 = 83.8 \text{ g./mole}$$

$$\bar{v}_2 = 20.7 \times 10^4 \frac{\text{cm.}}{\text{sec.}}$$

$$M_2 = 4.003 \text{ g./mole}$$

* The most abundant gaseous fission products were specified by Mr. Cottrell to be xenon (37%) and krypton (12%). The use of the diffusion properties of krypton again produces (cf. p. 10) a conservative estimate of D_1 . The "equivalent" amount of krypton was estimated by taking 50% of the specified fission rate (i.e., 50% of 6.5×10^{15} fissions per element per second) to be the rate of production of radioactive gas molecules and then assuming that all such molecules were krypton. Thus,

$$\begin{aligned} F_g &= 0.5 \times 6.5 \times 10^{15} \left(\frac{\text{molecules}}{\text{second}} \right) \times \frac{1}{6.06 \times 10^{23}} \left(\frac{\text{moles}}{\text{molecule}} \right) \\ &= 0.535 \times 10^{-8} \left(\frac{\text{moles}}{\text{second}} \right). \end{aligned}$$

** The mean molecular speed, $\bar{v}$, as obtained from the Maxwellian distribution of molecular speeds, is $(8RT/\pi M)^{1/2}$. The viscosity of helium, η , at 1000°F. is interpolated from Hutcheon's table VII (p. 268 of ref. 6). The diffusion coefficient at 1 atmosphere, pD_{12} , for a krypton-helium mixture is computed according to Hirschfelder⁹ et al.

$$\eta = 3.80 \times 10^{-4} \text{ poise}$$

$$\frac{M_1}{M_2} - 1 = 19.93$$

$$pD_{12} = 3.46 \frac{\text{atm. cm.}^2}{\text{sec.}}$$

$$RT = 6.656 \times 10^4 \frac{\text{atm. cm.}^3}{\text{mole}}$$

$$= 6.745 \times 10^{10} \frac{\text{ergs}}{\text{mole}}$$

With these gas properties, the following graphite specifications were used*:

$$r_2 = 5.26 \text{ cm.}, r_1 = 3.99 \text{ cm.}, \Delta z = \ln \frac{r_2}{r_1} = 0.276, 2\pi L = 1436 \text{ cm.},$$

$$\epsilon = 0.17, q^2 = 19.7.$$

From the foregoing numerical values, the constants in formulas (3) and (4) are found to be $E = 19.93$, and $H = 0.0781$. The remaining parameters, A, B, C, D, I, J, in the formulas (1) through (6) are readily computed when the pressure conditions Δp , $\bar{p}$ and the graphite properties K_0 , B_0 are specified. The results of such computations are illustrated in figures 2 and 3.

The Results of the Computations

Variation of External Mole Fraction with Sweep Flow. The variation of the external concentration of radioactive gas, N_e , with the sweep flow to the disposal unit, F , when the escape ratio, θ , is less than 10^{-5} , is shown in figure 2 as a set of curves of N_e vs. F for three values of the pore size (i.e., "mean hydraulic radius"), m . The broken curve (b) corresponds to the value $m = 0.215$ microns obtained by Hutcheon for the graphite samples which he studied. The other broken curves, (a) and (c), correspond respectively to pore sizes one-half and twice that of (b)**.

* Dimensions are as specified by Mr. Cottrell. The void fraction, ϵ , and tortuosity factor, $q = L_e/L$, are as measured by Hutcheon⁶.

** It should be remembered that the pore size, m , has been varied while keeping the porosity, ϵ , and tortuosity, q , constant. In preparing porous media with various properties, it will probably be difficult to vary a single graphite parameter independently in this manner.

The solid curves in figure 2 show the values of the pressure difference across the shell, Δp , which are required to achieve a given sweep rate, F . The external pressure, p_e , is taken to be 21 atmospheres so that the changes in Δp reflect changes of the pressure, p_1 , in the annulus, the latter presumably being brought about by varying the pumping speed at the disposal unit (cf. figure 1).

The following results are illustrated in figure 2:

- (1) Decreasing the pore size does not decrease N_e .
- (2) The rate of decrease of N_e with increasing F is retarded as the pore size is decreased by the pressure diffusion effect. It is clear that this must be so if one remembers that a given sweep flow, F , demands a rapidly increasing pressure gradient as the pore size is decreased. The effect is shown in a possibly exaggerated manner* by the appearance of the minimum in the broken curve (a).
- (3) If $N_e = 10^{-10}$ is taken as a desirable "safe" value, this value is incompatible with $\theta \leq 10^{-5}$ for the gas and graphite properties and the sweep rates now being considered.
- (4) Even these insufficient sweep rates correspond to volumetric flow rates which may call for a prohibitively large disposal unit; e.g., $F = 0.01$ moles/sec. at 1000°F . corresponds to $33.3 \text{ cm}^3/\text{sec.}$ at 20 atm. or $666 \text{ atm.cm}^3/\text{sec.}$ from each fuel element.

Variation of Escape Ratio with Sweep Flow. With the value of the external mole fraction, N_e , fixed at 10^{-10} (cf. result (3) above) the variation of the escape ratio, $\theta = F_1/F_g$, with the sweep flow, F , is shown in figure 3 as a set of curves of θ vs. F for the same three values of the pore size, m , as used previously. The figure illustrates that:

- (1) Decreasing the pore size can either decrease or increase the escape ratio, depending on the magnitude of the sweep flow.
- (2) If $\theta = 10^{-5}$ is taken as a desirable value of the escape ratio consistent with the over-all material balance, radioactivity deposition rate, half-life, etc., (cf. footnote, pg. 9), this value is incompatible with $N_e \leq 10^{-10}$ for the gas and graphite properties and the sweep rates now being considered.

*The pressure diffusion terms have been included in equations (1), (2), and (3) in a manner which, strictly speaking, is theoretically justified only when $D_1 = D_{12}$; i.e., when $m/\lambda \rightarrow \infty$. It is possible that appropriate experiments would show these terms to be less important as the pore size is decreased.

The incompatibility, shown in figures 2 and 3, of simultaneous low values* of θ and N_e must not be taken as a well-established conclusion because of the various incomplete or uncertain features of the present treatment. For example, the method of setting "safe" values of θ and N_e warrants much closer scrutiny than is attempted here. (e.g., we have ignored the relationship of these values to the material balance in the primary system). Also, our method of introducing the pressure diffusion effect lacks experimental verification. Even more important is the previously mentioned uncertainty in our estimates of the effective diffusion coefficient, D_1 . It is not inconceivable that the values used here are an order of magnitude too high. For these reasons, figures 2 and 3 should be interpreted as only qualitative indications of the kind of results obtainable from the calculation methods which are being suggested here. Hence, these results are useful for planning and interpreting further research rather than for current reactor design work.

Effect of the Uncertainty in the Values of D_1 . In order to illustrate the sensitivity of the results to the estimated value of the diffusion coefficient, N_e has been computed for various arbitrary values of D_1 . Thus, while formulas (1) and (4) were evaluated as in the calculations for curve (b) of figure 2, formula (2) was replaced by arbitrary values of D_1 , and these were used in (3) and (5). The results are shown in figure 4 as graphs of N_e vs. D_1 for $\theta < 10^{-5}$ and for three values of Δp and sweep flow. The results of the previous calculations, cf. curve (b) of figure 2, are indicated on these graphs as corresponding to "theoretical" values of D_1 . It is seen that it may be possible to attain the required low values of N_e and θ and, hence, that the existing uncertainty of the correct values of the diffusion coefficient should not be tolerated if this type of design for unclad fuel elements is to be given serious consideration.

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*The value $\theta = 10^{-5}$ was included in the specifications furnished by Mr. Cottrell. The value $N_e = 10^{-10}$ was estimated by converting the lowest atomic concentration which he specified (1.6×10^{12} atoms/cm.³ for Sr^{90}) to a mole fraction at $P_e = 21$ atmospheres and introducing an additional safety factor of roughly 0.1.

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VARIATION OF EXTERNAL MOLE FRACTION, N_e , AND OF PRESSURE DROP, ΔP , WITH SWEEP FLOW TO DISPOSAL UNIT, F .

FIGURE 2

VARIATION OF ESCAPE RATIO, θ ,
WITH SWEEP FLOW TO DISPOSAL UNIT, F .

FIGURE 3

VARIATION OF EXTERNAL MOLE FRACTION, N_e , WITH DIFFUSION COEFFICIENT, D_i

FIGURE 4