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GAMD-7535 (Rev.)

**FREVAP-8 FORMULATION FOR ESTIMATING THE RELEASE OF
METALLIC FISSION PRODUCTS FROM HTGR FUEL ELEMENTS**

by

L. R. Zumwalt

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ERRATA

GAMD-7535 (Rev.), FREVAP-8 FORMULATION FOR ESTIMATING THE RELEASE OF METALLIC FISSION PRODUCTS FROM HTGR FUEL ELEMENTS

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March 24, 1967

The following errata should be noted in General Atomic Report
GAMD-7535 (Rev.):

Page 2, middle of page --

Change $\bar{C}_{chik}$ to read $\bar{C}_{Chik}$

Page 8, middle of page, second line of definition of Λ_{Ghj} --

Change $\Lambda_{Ghi} = 1$ to read $\Lambda_{Ghl} = 1$

Page 2-A, Table A1, title --

Change Constant to read Constants

Page 4-A, Eq. 12A, right side --

Change u_{h-1} to read $(u_h - 1)$

Page 5-A, second line before Eq. 15A --

Change $\bar{C}_{chik}$ to read $\bar{C}_{Chik}$

Page 1-D, Eq. 2D, right side --

Change $\frac{Q_{th}}{R_q}$ to read $\frac{Q_{dh}}{R_q}$

Page 6-D, Table D1, second column heading --

Change D_{oh} to read D_h^o

FREVAP-8 FORMULATION FOR ESTIMATING THE RELEASE OF METALLIC FISSION PRODUCTS FROM HTGR FUEL ELEMENTS

This is a fission product release model for formulating a digital computer code for two absorber regions taking into account multicomponent sorption behavior and assuming steady-state diffusion is attained. Reference is made to earlier formulations of FREVAP.*

In this calculation we have an additional index, h (or h'), for chemical element species, which is necessary for the treatment of the effect of the competition of various chemical species (elements) for sorption sites in graphite or amorphous carbon.

The basic limiting process considered in FREVAP is the evaporation of fission product nuclides into the coolant stream at the graphite-coolant interface. This process in turn is limited by the sorption of fission products in the graphite between the fuel holes and the coolant holes and in the carbon sorbent material surrounding the fuel particles in the fuel hole. Finally the process is limited by the release of fission products from the fuel particles themselves.

Our basic consideration is of the graphite between the fuel volume and the coolant passageways in horizontal segment k of a given assumed independent core region (radial zone). As an approximation, the graphite is assumed to have an infinite slab geometry, with location expressed by one dimension (x). At the fuel-graphite interface, $x = 0$, and at the graphite-coolant interface, $x = L$ (i.e., the thickness of the graphite equals L [cm]).

The continuity equation for isotope i of chemical element h in axial segment k of a given fuel element radial region (patch) is given by:

$$\frac{M_{Gk}}{10^6} \left[\frac{d \bar{C}_{hik}}{dt} + \lambda_{hi} \bar{C}_{hik} \right] + \frac{M_{Ck}}{10^6} \left[\frac{d \bar{C}_{Chik}}{dt} + \lambda_{hi} \bar{C}_{Chik} \right] = S_{hik} - \left\{ (n_{He}/P_{He})_k \zeta_{hik} P_{bhik} - (n_{He}/P_{He})_{(k-1)} \zeta_{hi(k-1)} P_{bhi(k-1)} \right\} \quad \{1\}$$

* Zumwalt, L. R., et al, "FREVAP-6 Code for Estimating Metallic Fission Product Release" USAEC Report GAMD-7656, General Atomic Division, General Dynamics Corp., 1967.

where

M_{Gk} = total quantity of graphite in segment k of a given fuel element region (radial zone) [gm]

$C_{hik}(x)$ = the concentration of isotope i of chemical element h at location x in the graphite of segment k, [μ moles/gm]

and then

$$\bar{C}_{hik} = \frac{1}{L} \int_0^L C_{hik}(x) dx = \text{the average concentration of i in the graphite of segment k } [\mu\text{moles/gm}]$$

t = time [days]

λ_{hi} = decay constant of isotope hi [days^{-1}]

M_{Ck} = total quantity of sorbent carbon material in the fuel volume of segment k of a given fuel element region (radial zone) [gm]

$\bar{C}_{chik}$ = average concentration in the carbon of f. p. nuclide of chemical element h [μ moles/gm]

S_{hik} = rate of release of isotope hi from the fuel particles in segment k [moles/day]

n_{Hek} = rate of flow of the helium coolant in the given fuel element region (radial zone), segment k [moles/day]

P_{Hek} = pressure of coolant in segment k [atm]

$(n_{He}/P_{He})_k \equiv n_{Hek}/P_{Hek}$ and likewise for (k-1)

(Note $P_{He} \gg P_{chi}$ = partial pressure of isotope hi in coolant)

$\zeta_{hik} \equiv \frac{P_{chik}}{P_{bhik}} = \text{ratio of partial pressure of isotope hi in the coolant stream to the equilibrium pressure of hi at the graphite-coolant interface for segment k. } \{2\}$

Note: The sum of the equilibrium pressures of isotopes of the chemical element h (i.e., Cs, Sr, Ba, etc.) sorbed by graphite is given (empirically) by the Freundlich isotherm. The equilibrium pressure of isotope hi at the graphite-coolant interface in the case of a system with more than one h is given approximately by (see Appendix A -- Eq. {14A}) ,

$$P_{bhi} = P_{GLmh} \frac{C_{hi}(L)}{C_{mh}} \left[\sum_{h'} \sum_{i'} \frac{C_{h'i'}(L)}{C_{mh'}} \right]^{(u_{GLh}^{-1})} \quad [\text{atm}] \quad \{3\}$$

where

$C_{hi}(L)$ = concentration of hi in graphite at $x = L$ (the graphite-coolant interface) $[\mu\text{moles/gm}]$

C_{mh} = "monolayer" concentration of h in the graphite (a constant, based on the size of the atoms and the specific internal, BET, surface area of the graphite)--see Appendix A for values of C_{mh} $[\mu\text{moles/gm}]$

$$\ln P_{GLmh} = A_{mh} + B_{mh} \left(\frac{10^3}{T_{GL}} \right) \quad \{4\}$$

with

$$A_{mh} = a_h + c_h \ln C_{mh} \quad \{5\}$$

$$B_{mh} = b_h + d_h \ln C_{mh} \quad \{6\}$$

and

$$u_{GLh} = c_h + d_h \left(\frac{10^3}{T_{GL}} \right) \quad \{7\}$$

T_{GL} = temperature of the graphite at $x = L$ $[^{\circ}\text{K}]$.

As discussed in more detail in Appendix A, a_{Gh} , b_{Gh} , c_{Gh} , and d_{Gh} are empirical constants expressing the Freundlich isotherm equation for the equilibrium partial pressure of a single chemical element h sorbed at concentration C_h and graphite temperature T_G [$^{\circ}K$],

$$\ln P_h = a_h + b_h \left(\frac{10^3}{T} \right) + \left[c_h + d_h \left(\frac{10^3}{T_G} \right) \right] \ln C_h \quad \{8\}$$

P_h = partial pressure of h [atm] .

The concentration of h is the sum of the concentrations of the isotopes h_i

$$C_h = \sum_i C_{hi} \quad [\mu\text{moles/gm}] . \quad \{9\}$$

Values of the empirical constants are given in Appendix A.

Let

$$\eta_{hik} \equiv \frac{C_{hik}(L)}{\bar{C}_{hik}} = \text{ratio of the concentration of } h_i \text{ in graphite at coolant interface to the average concentration in segment } k . \quad \{10\}$$

Then per equations {3}, {4}, and {10}, for segment k

$$P_{bhik} = \exp \left[A_{mh} + B_{mh} \left(\frac{10^3}{T_{GLk}} \right) \right] \frac{\eta_{hik} \bar{C}_{hik}}{C_{mh}} \left[\sum_{h'} \sum_{i'} \eta_{h'i'k} \frac{\bar{C}_{h'i'}}{C_{mh'}} \right]^{(u_{GLhk}-1)} \quad [\text{atm}] \quad \{3'\}$$

likewise, for segment $k-1$,

$$P_{h(k-1)} = \exp \left[A_{mh} + B_{mh} \left(\frac{10^3}{T_{GL(k-1)}} \right) \right] \frac{\eta_{hi(k-1)} \bar{C}_{hi(k-1)}}{C_{mh}} \left[\sum_{h'} \sum_{i'} \eta_{h'i'(k-1)} \frac{\bar{C}_{h'i'}}{C_{mh'}} \right]^{(u_{GLh(k-1)}-1)} \quad [\text{atm}] .$$

The zeta factor is given for segment k by (see formulation of FREVAP-4 Jan-Mar 1966 PSC Quarterly Report)

$$\zeta_{hik} = \frac{\left(\frac{P_{bhi(k-1)}}{P_{bhik}} \right) \zeta_{hi(k-1)} + H_{hjk} \frac{(A/\ell)_k}{R_g T_{cjk} (n_{He}/P_{He})_k}}{\left[1 + \frac{H_{hjk} (A/\ell)_k}{R_g T_{cjk} (n_{He}/P_{He})_k} \right]} \quad \{11\}$$

and for k = 1 (assuming complete plateout of hi in the coolant circuit)

$$\zeta_{hil} = \frac{H_{hjl} \frac{(A/\ell)_1}{R_g T_{cjl} (n_{He}/P_{He})_1}}{\left[1 + \frac{H_{hjl} (A/\ell)_1}{R_g T_{cjl} (n_{He}/P_{He})_1} \right]} \quad \{11'\}$$

where

H_{hjk} = mass transfer coefficient for h, usually as the monatomic molecule (for transport across boundary layer at graphite-coolant interface) for segment k in time interval j [cm/day]
(See Appendix B for the equation for H_{hjk} , etc.)

T_{cjk} = coolant temperature for segment k, time interval j [$^{\circ}$ K]

R_g = gas constant
= 82.06 $\left[\frac{\text{cm}^3 \text{-atm}}{\text{mole } ^{\circ}\text{K}} \right]$

ℓ_k = length of coolant channel in segment k [cm]

(A/ℓ) = area to length ratio of coolant channels [cm] .

Time (t) is considered to be divided up into intervals--designated by an index j--over which the various temperatures in the reactor core and the production rate of fission products (reactor power) are approximately constant. Calculational steps will be divided up into these time intervals and appropriate values of temperature and f. p. production rate assigned. The index j is generally omitted in this mathematical formulation to simplify the equations, with a few exceptions, but it should be considered to be present (where appropriate). The f. p. source term S_{hik} (S_{hijk}) may be quite complex; however, in the case of "steady-state" release from the fuel particles (which has been used as a tentative approximation) it is given as follows:

$$S_{hijk} = R_{hjk} N_{hik}(t) \quad [\text{moles/day}] \quad \{12\}$$

where

$$\ln R_{hjk} = \ln R_{oh} - \frac{E_{ah}}{1.987} \left(\frac{10^3}{T_{Fjk}} \right) \quad \{13\}$$

R_{oh} = empirical release constant for chemical element h (characteristic of the given fuel particles) $[\text{day}^{-1}]$

E_{ah} = empirical activation energy of release of chemical element h for the given fuel particles $[\text{kcal/mole}]$

T_{Fjk} = average fuel temperature - time interval j, region k $[^{\circ}\text{K}]$

and

$N_{hik}(t)$ = the quantity of isotope h_i [moles] in the fuel particles at time t which is determined from

$$\frac{dN_{hik}(t)}{dt} = B_{hijk} - \lambda_{hi} N_{hik}(t) - R_{hjk} N_{hik}(t), \quad [\text{moles/day}] \quad \{14\}$$

where

B_{hijk} = production rate of f. p. isotope h_i in the fuel particles in segment k during time interval j [moles/day].

See Appendix C for modification of FREVAP for more than one type of fuel particle.

We need to express the continuity equation set {1} entirely in terms of $\bar{C}_{hik}$'s. For this, besides equations {3'} and {3''}, we need a relationship between $\bar{C}_{Chik}$ and $\bar{C}_{hik}$.

Let

$$\xi_{hik} \equiv \frac{C_{hik}^{(0)}}{\bar{C}_{hik}} \quad \{15\}$$

The ξ factor ξ_{hik} and the η factor η_{hik} (defined by equation {10}) depend on the diffusion and decay behavior of the isotopes h_i in the graphite (segment k) in the region $x = 0$ to $x = L$.

Under conditions of quasi thermodynamic equilibrium the relationship between $\bar{C}_{Chik}$ and $C_{hik}^{(0)}$ is approximately as follows:

$$\bar{C}_{Chik} = w_{hk} C_{hik}^{(0)} \quad \{16\}$$

If the fuel temperature (T_{Fk}) and the graphite interface temperature (T_{Gok}) are the same (T_k)

$$w_{hk} = K_h(T_k) \quad \{17\}$$

where

$K_h(T_k)$ = distribution coefficient, i. e., the sorbent capacity of the carbon material in the fuel holes relative to the sorbent

capacity of graphite at temperature T_k

$$K_h(T_k) = E_h + F_h \left(\frac{10^3}{T_k} \right)$$

The equation for w_{hk} is

$$w_{hk} = K_h(T_{Fk}) \frac{C_h(T_{Fk})}{C_h(T_{G0k})}$$

where the concentration ratio $C_h(T_{Fk})/C_h(T_{G0k})$ is as given in Appendix A (pg. 6-A).

Recent experimental data (see PSC Chemistry Quarterly Progress Report Oct-Dec. 1966) has indicated the sorption capacity of graphite and activated carbon changes with irradiation. Accordingly the sorption capacity factors

Λ_{Ghj} = relative sorption capacity of fuel element graphite for h in time step j. (For the initial time step, $\Lambda_{Ghi} = 1$ corresponding to the relative sorption capacity of unirradiated graphite.),

Λ_{Chj} = relative sorption capacity of carbon in the fuel region for h in time step j. (For the initial time step, $\Lambda_{Ch1} = 1$),

are introduced into the continuity Eq. {1} and substituting {15} and {16} in {1} we obtain for the continuity equation,

$$\frac{\Lambda_{Gj} M_{Gk} + \Lambda_{Cj} w_{hk} \xi_{hijk} M_{Ck}}{10^6} \left[\frac{d \bar{C}_{hijk}}{dt} + \lambda_{hi} \bar{C}_{hijk} \right] = S_{hijk} - \left\{ (n_{He}/P_{He})_k \zeta_{hik} P_{bhik} - (n_{He}/P_{He})_{(k-1)} \zeta_{hi(k-1)} P_{bhi(k-1)} \right\} \quad \{1'\}$$

where S_{hijk} , P_{bhik} and $P_{bhi(k-1)}$ are given by {12}, {3'}, and {3''}, respectively.

It is to be noted FREVAP-8 is much like FREVAP-6; however, now each equation of the set of continuity equations {1'} has a summation of $\eta \bar{C}$ for all the intermediate, long-lived, and stable isotopes of all the chemical elements of interest (actually the elements comprise Sr, Cs, Rb, and Ba at present, however more may be included as data becomes available). Accordingly, the FREVAP-8 calculation must carry out concentration and release calculations for all the fission product chemical elements at once. The HAMDE code* is used to solve {1'}.

The following gives the basis and formulae for calculating ξ_{hi} and η_{hi} (subscript k omitted).

The differential equation for $C_{hi}(x)$, the concentration of hi in graphite between $x = 0$ and $x = L$, which corresponds to steady-state diffusion, is as follows (see Appendix D for more details):

$$\frac{d^2 C_{hi}}{dx^2} = \Gamma_h \left(\frac{d C_{hi}}{dx} \right) + \frac{\lambda_{hi}}{D_{Gh}} C_{hi} \quad \{18\}$$

and at the fuel-graphite interface $x = 0$

$$C_{hi} = C_{hi}(0), \text{ and}$$

$$\frac{d C_{hi}(0)}{dx} = - \frac{10^6 F_{hi}}{\rho D_{Goh}} + \Gamma_h C_{hi}(0) \quad \{19\}$$

and define

$$\alpha = \alpha_{hi} \equiv \sqrt{\left(\frac{\Gamma_h}{2} \right)^2 + \frac{\lambda_{hi}}{D_{Gh}}} + \frac{\Gamma_h}{2} \quad \{20\}$$

$$\beta = \beta_{hi} \equiv \sqrt{\left(\frac{\Gamma_h}{2} \right)^2 + \frac{\lambda_{hi}}{D_{Gh}}} - \frac{\Gamma_h}{2} \quad \{20'\}$$

* H. D. Brown, Jr., and J. E. Gratteau, Private Communication, General Atomic

Note: α and β should have the subscripts hi (i.e., α_{hi} , β_{hi}) but are not given these to simplify writing the following equations:

Define

$$\Omega_{hi} \equiv \frac{10^6 F_{hi}}{\rho D_{G0h} C_{hi}(0)} \quad \{21\}$$

Let us consider special case 1.

where

$$\alpha = \beta = \sqrt{\lambda_{hi}/D_{Gh}} \quad \left[\frac{1}{\text{cm}} \right], \text{ i.e., } \Gamma_h = 0. \quad \{22\}$$

(See Eq. {2D}, Appendix D for equation for $\ln D_{Gh}$.)

The steady-state diffusion equations for this case are

$$C_{hi}(x) = \frac{C_{hi}(0)}{\alpha} \left\{ \alpha \cosh \alpha x - \Omega_{hi} \sinh \alpha x \right\} \quad \left[\frac{\mu \text{ moles}}{\text{gm}} \right] \quad \{23\}$$

and

$$\frac{d C_{hi}(x)}{dx} = C_{hi}(0) \left\{ \alpha \sinh \alpha x - \Omega_{hi} \cosh \alpha x \right\} \quad \left[\frac{\mu \text{ moles}}{\text{gm cm}} \right]. \quad \{24\}$$

From the boundary condition at $x = L$ (see {39}),

$$\Omega_{hi} = \alpha \left[\frac{\psi_{hi} \cosh \alpha L + D_{GLh} \alpha \sinh \alpha L}{\psi_{hi} \sinh \alpha L + D_{DLh} \alpha \cosh \alpha L} \right] \quad \left[\frac{1}{\text{cm}} \right] \quad \{25\}$$

where based on Eq. {3'} for P_{bhik} ,

$$\psi_{hi} \equiv \exp \left[A_{mh} + B_{mh} \left(\frac{10^3}{T_{GLk}} \right) \right] \frac{1}{C_{mh}} \left[\sum_{h'} \sum_{i'} \eta_{h'i'k} \frac{\bar{C}_{h'i'}}{C_{mh'}} \right]^{(u_{GLh} - 1)} \\ \times \frac{10^6 H_h}{\rho R_g T_c} (1 - \zeta_{hi}) \quad \left[\frac{\text{cm}}{\text{sec}} \right]. \quad \{26\}$$

Let

$$\varphi_{hi} \equiv \frac{\psi_{hi} L}{D_{GLh}} \quad (\infty > \varphi_{hi} > 0) \quad (\text{dimensionless}) \quad \{27\}$$

then

$$\Omega_{hi} = \alpha \left[\frac{\varphi_{hi} \cosh \alpha L + \alpha L \sinh \alpha L}{\varphi_{hi} \sinh \alpha L + \alpha L \cosh \alpha L} \right] \left[\frac{1}{\text{cm}} \right] . \quad \{25'\}$$

Now

$$\bar{C}_{hi} = \frac{1}{L} \int_0^L C_{hi}(x) dx = \frac{C_{hi}^{(0)}}{\alpha L} \left\{ \sinh \alpha L - \Omega_{hi} \left[\frac{\cosh \alpha L - 1}{\alpha} \right] \right\}$$

$$\xi_{hi} = \frac{C_{hi}^{(0)}}{\bar{C}_{hi}} = \frac{\alpha L}{\sinh \alpha L - \left[\frac{\varphi_{hi} \cosh \alpha L + \alpha L \sinh \alpha L}{\varphi_{hi} \sinh \alpha L + \alpha L \cosh \alpha L} \right] (\cosh \alpha L - 1)}$$

$$\begin{aligned} \xi_{hi} &= \frac{\alpha L \left[(\varphi_{hi}/\alpha L) \sinh \alpha L + \cosh \alpha L \right]}{(\varphi_{hi}/\alpha L) (\cosh \alpha L - 1) + \sinh \alpha L} \\ &= \frac{\alpha L \left[(\varphi_{hi}(\alpha L) \tanh \alpha L + 1) \right]}{(\varphi_{hi}/\alpha L) \left[1 - \frac{1}{\cosh \alpha L} \right] + \tanh \alpha L} \end{aligned} \quad \{28\}$$

As $\alpha L \rightarrow 0$ (i. e. $\lambda_{hi} \rightarrow 0$),

$$\xi_{hi} = \frac{\varphi_{hi} \left[\alpha L + (\alpha L)^3 / 3! + \dots + \alpha L + (\alpha L)^3 / 2! + \dots \right]}{\varphi_{hi} (\alpha L) / 2! + (\alpha L)^3 / 4! + \alpha L + (\alpha L)^3 / 3! + \dots}$$

$$\rightarrow \frac{\varphi_{hi} (\alpha L) + \alpha L}{\varphi_{hi} (\alpha L) / 2 + \alpha L} \rightarrow \frac{\varphi_{hi} + 1}{\varphi_{hi} / 2 + 1} \quad \{28'\}$$

(Note: if $\alpha L = 0$ and $\varphi_{hi} = 1$, $\xi_{hi} = \frac{4}{3}$,

If $\varphi_{hi} = 0$, $\xi_{hi} = 1$, and if $\varphi_{hi} = \infty$, $\xi_{hi} = 2$)

and when $\alpha L \gg 1$

$$\xi_{hi} \approx \frac{\alpha L \left[(\varphi_{hi} / \alpha L) + 1 \right]}{(\varphi_{hi} / \alpha L) \left[1 - 2 e^{-\alpha L} \right] + 1} \rightarrow \alpha L \quad \{28''\}$$

$$\eta_{hi} = \frac{\alpha L \cosh \alpha L - \left[\frac{\varphi_{hi} \cosh \alpha L + \alpha L \sinh \alpha L}{\varphi_{hi} \sinh \alpha L + \alpha L \cosh \alpha L} \right] \sinh \alpha L}{\sinh \alpha L - \left[\frac{\varphi_{hi} \cosh \alpha L + \alpha L \sinh \alpha L}{\varphi_{hi} \sinh \alpha L + \alpha L \cosh \alpha L} \right] (\cosh \alpha L - 1)}$$

$$= \frac{\alpha L}{(\varphi_{hi} / \alpha L) (\cosh \alpha L - 1) + \sinh \alpha L}$$

$$= \frac{\alpha L / \cosh \alpha L}{(\varphi_{hi} / \alpha L) \left[1 - \frac{1}{\cosh \alpha L} \right] + \tanh \alpha L} \quad \{29\}$$

As $\alpha L \rightarrow 0$ (i.e. $\lambda_{hi} \rightarrow 0$),

$$\eta_{hi} \approx \frac{\alpha L}{\varphi_{hi} \left[(\alpha L) / 2! + (\alpha L)^3 / 4! + \dots \right] + \alpha L + (\alpha L)^3 / 3! + \dots} \rightarrow \frac{1}{\varphi_{hi} / 2 + 1} \quad \{29'\}$$

(Note: if $\alpha L = 0$ and $\varphi_{hi} = 1$, $\eta_{hi} = \frac{2}{3}$; if $\varphi_{hi} = 0$, $\eta_{hi} = 1$ and if $\varphi_{hi} = \infty$,

$\eta_{hi} = 0$).

and when $\alpha L \gg 1$

$$\eta_{hi} = \frac{2 \alpha L e^{-\alpha L}}{(\varphi_{hi}/\alpha L) + 1} \rightarrow 0 \quad \{29''\}$$

It is of interest to note that the ratio of hi concentration in graphite at coolant interface to that of the fuel interface is given by:

$$\frac{C_{hi}(L)}{C_{hi}(0)} = \frac{\eta_{hi}}{\xi_{hi}} = \frac{1}{\cosh \alpha L [(\varphi_{hi}/\alpha L) \tanh \alpha L + 1]} \quad \{30\}$$

The ratio $\rightarrow 0$ when $\alpha L \gg 1$ and $\rightarrow \frac{1}{\varphi_{hi} + 1}$ when $\alpha L \rightarrow 0$

Now let us consider the general case where $\Gamma_h \neq 0$:

Γ_h is determined by the effect of thermal diffusion.

Thus,

$$\Gamma_h = \frac{Q_{th}}{R_q} G_T \quad [1/\text{cm}] \quad \{31\}$$

where

Q_{th} = heat of transport of h[kcal/mole]

$R_q = 1.987$ [calories/mole $^{\circ}\text{K}$]

and

$$G_T = \frac{\left(\frac{10^3}{T_{GL}}\right) - \left(\frac{10^3}{T_{G0}}\right)}{L} \quad \{32\}$$

The solution for $C_{hi}(x)$, $\frac{d C_{hi}}{dx}$, and $\bar{C}_{hi}$ (see Appendix D for details) when $\Gamma_h \neq 0$ are as follows:

$$C_{hi}(x) = \frac{C_{hi}(0)}{(\alpha + \beta)} \left\{ \alpha e^{\alpha x} + \beta e^{-\beta x} - \Omega_{hi} \left(e^{\alpha x} - e^{-\beta x} \right) \right\} \quad \{33\}$$

$$\frac{dC_{hi}(x)}{dx} = \frac{C_{hi}(0)}{(\alpha+\beta)} \left\{ \alpha^2 e^{\alpha x} - \beta^2 e^{-\beta x} - \Omega_{hi} (\alpha e^{\alpha x} + \beta e^{-\beta x}) \right\}. \quad \{34\}$$

$$\text{Integrating to obtain } \bar{C}_{hi} = \frac{1}{L} \int_0^L C_{hi}(x) dx \quad \{35\}$$

$$\bar{C}_{hi} = \frac{C_{hi}(0)}{L(\alpha+\beta)} \left\{ e^{\alpha L} - e^{-\beta L} - \Omega_{hi} \left[\frac{\beta(e^{\alpha L} - 1) + \alpha(e^{-\beta L} - 1)}{\alpha\beta} \right] \right\}. \quad \{36\}$$

Thus we have for ξ_{hi} and η_{hi}

$$\xi_{hi} = \frac{C_{hi}(0)}{\bar{C}_{hi}} = \frac{(\alpha+\beta)L}{\left[e^{\alpha L} - e^{-\beta L} \right] - \Omega_{hi} \left[\frac{\beta(e^{\alpha L} - 1) + \alpha(e^{-\beta L} - 1)}{\alpha\beta} \right]} \quad \{37\}$$

and

$$\eta_{hi} = \frac{C_{hi}(L)}{\bar{C}_{hi}} = \frac{L \left\{ \left[\alpha e^{\alpha L} + \beta e^{-\beta L} \right] - \Omega_{hi} \left[e^{\alpha L} - e^{-\beta L} \right] \right\}}{\left[e^{\alpha L} - e^{-\beta L} \right] - \Omega_{hi} \left[\frac{\beta(e^{\alpha L} - 1) + \alpha(e^{-\beta L} - 1)}{\alpha\beta} \right]}. \quad \{38\}$$

To obtain Ω_{hi} , we utilize the flux boundary condition at the graphite-coolant interface given (for segment k) by:

$$\text{Flux} = \frac{H_h P_{bhik}}{R_g T_{ck}} (1 - \zeta_{hik}) = - \frac{\rho D_{GLhk}}{10^6} \left\{ \frac{d C_{hik}(L)}{dx} - \Gamma_h C_{hik}(L) \right\} \left[\frac{\text{moles}}{\text{cm}^2 \text{ sec}} \right] \quad \{39\}$$

where

$$\ln D_{GLh} = \ln D_h^o - \frac{10^3}{T_{GL}} \frac{Q_{hd}}{R_q} + \delta_h \left[\sum_{h'} \sum_{i'} \eta_{h'i'} \bar{C}_{h'i'} \frac{C_{mh}}{C_{mh'}} \right] \quad \{40\}$$

and where P_{bhik} is given by Eq. {3'}.

Substituting {33}, {34}, and {3'} into {39} (letting $x = L$)

$$\begin{aligned} & \frac{C_{hi}(0)}{(\alpha + \beta)} \left\{ \alpha e^{\alpha L} + \beta e^{-\beta L} - \Omega_{hi} (e^{\alpha L} - e^{-\beta L}) \right\} \\ & \left\{ \exp \left[A_{mh} + B_{mh} \left(\frac{10^3}{T_{GLk}} \right) \right] \frac{1}{C_{mh}} \left[\sum_{h'} \sum_{i'} \eta_{h'i'k} \frac{\bar{C}_{h'i'}}{C_{mh'}} \right] \frac{10^6 H_h}{\rho R_g T_c} (1 - \zeta_i)^{-D_{GL} \Gamma_h} \right\}^{(u_{GLhk} - 1)} \\ & + \frac{C_i(0)}{(\alpha + \beta)} \left\{ \alpha^2 e^{\alpha L} - \beta^2 e^{-\beta L} - \Omega_i (\alpha e^{\alpha L} + \beta e^{-\beta L}) \right\}^{-D_{GL}} = 0 \quad \{42\} \end{aligned}$$

and define ψ_{hi} and φ_{hi} as in {26} and {27}, respectively

$$\Omega_{hi} = \frac{(\alpha e^{\alpha L} + \beta e^{-\beta L}) \varphi_{hi} + L \left[(\alpha^2 e^{\alpha L} - \beta^2 e^{-\beta L}) \Gamma_h (\alpha e^{\alpha L} + \beta e^{-\beta L}) \right]}{(e^{\alpha L} - e^{-\beta L}) \varphi_{hi} + L \left[(\alpha e^{\alpha L} + \beta e^{-\beta L}) - \Gamma_h (e^{\alpha L} - e^{-\beta L}) \right]} \quad \{43\}$$

or since $\Gamma_h = \alpha - \beta$

$$\Omega_{hi} = \frac{(\alpha e^{\alpha L} + \beta e^{-\beta L}) \varphi_{hi} + \alpha \beta L (e^{\alpha L} - e^{-\beta L})}{[(e^{\alpha L} - e^{-\beta L}) \varphi_{hi} + L (\beta e^{\alpha L} + \alpha e^{-\beta L})]} \quad \{43'\}$$

The general expressions for ξ_{hi} and η_{hi} in terms of α , β , L and φ_{hi} are obtained by substituting {43'} into {37} and {38}, respectively.

Let us consider the expressions for ξ_{hi} and η_{hi} in certain cases.

In special case 2.

$$\Gamma_h > 0 \text{ and } \lambda_{hi} \rightarrow 0$$

Then

$$\alpha = \Gamma_h \text{ and } \beta \rightarrow 0$$

And

$$\xi_{hi} = \frac{\alpha L [1 - e^{-\alpha L}] \varphi_{hi} + \alpha L e^{-\alpha L}}{\varphi_{hi} [\alpha L - 1 + e^{-\alpha L}] + \alpha L (1 - e^{-\alpha L})} \quad \{37'\}$$

As

$$\alpha L \rightarrow 0, \xi_{hi} \rightarrow \frac{\varphi_{hi} + 1}{\varphi_{hi}/2 + 1} \quad (\text{same as } \{28'\})$$

When $\alpha L \gg 1$

$$\xi_{hi} \rightarrow \frac{\varphi_{hi}}{\varphi_{hi}(\alpha L - 1)/\alpha L + 1} \quad \{37''\}$$

Also

$$\eta_{hi} = \frac{(\alpha L)^2}{\varphi_{hi} [\alpha L - 1 + e^{-\alpha L}] + \alpha L (1 - e^{-\alpha L})} \quad \{38'\}$$

As $\alpha L \rightarrow 0$,

$$\eta_{hi} \rightarrow \frac{1}{\varphi_{hi}/2 + 1} \quad (\text{same as } \{29'\})$$

When $\alpha L \gg 1$

$$\eta_{hi} = \frac{\alpha L}{\varphi_{hi}(\alpha L - 1)/\alpha L + 1} \quad \{38''\}$$

In special case 3.

$$\Gamma_h < 0 \text{ and } \lambda_{hi} \rightarrow 0$$

Then

$$\beta = -\Gamma_h = |\Gamma_h|, \alpha \rightarrow 0$$

And

$$\xi_{hi} = \frac{\beta L [(1 - e^{-\beta L}) \varphi_{hi} + \beta L]}{(1 - e^{-\beta L} - \beta L e^{-\beta L}) \varphi_{hi} + \beta L (1 - e^{-\beta L})} \quad \{37''\}$$

As $\beta L \rightarrow 0$,

$$\xi_{hi} \rightarrow \frac{\varphi_{hi} + 1}{\varphi_{hi}/2 + 1} \quad (\text{same as } \{28'\})$$

When $\beta L \gg 1$

$$\xi_{hi} \rightarrow \frac{\beta L [\varphi_{hi} \beta L]}{\varphi_{hi} + \beta L} = \beta L \quad \{37^{iv}\}$$

Also

$$\eta_{hi} = \frac{(\beta L)^2 e^{-\beta L}}{[1 - e^{-\beta L} - \beta L e^{-\beta L}] \varphi_{hi} + \beta L (1 - e^{-\beta L})} \quad \{38''\}$$

As $\beta L \rightarrow 0$,

$$\eta_{hi} \rightarrow \frac{1}{\varphi_{hi}/2 + 1} \quad (\text{same as } \{29'\})$$

When $\beta L \gg 1$

$$\eta_{hi} \approx \frac{\beta L e^{-\beta L}}{\varphi_{hi}/\beta L + 1} \rightarrow 0$$

In carrying out FREVAP calculations for ξ_{hi} and η_{hi} , we take $\varphi_{hi} = 0$ for the first time step $j = 1$. In subsequent time steps we calculate φ_{hi} (ψ_{hi} and D_{GL}) from the η_{hi} , $\bar{C}_{hi}$ and ζ_{hi} values obtained at the end of the previous time step. It is believed these calculations need to be done only over relatively long time step intervals. However, the short time steps of HAMDE may be used.

The quantity of a given fission product isotope hi , $(QR)_{hi}$, released from a given radial zone is obtained by integrating the amount of hi carried by the coolant stream from the last horizontal segment (k') of the reactor core. Thus, in the n^{th} time interval (assumed not completed), at time t , $(QR)_{hi}$ is given by:

$$(QR)_{hi} = (n_{He}/P_{He})_{k'} \left\{ \sum_{j=1}^{n-1} \int_{t_{j-1}}^{t_j} \zeta_{hijk'} P_{bhijk'} \exp[-\lambda_{hi}(t-\tau)] d\tau + \int_{t_{n-1}}^t \zeta_{hink'} P_{bhink'} \exp[-\lambda_{hi}(t-\tau)] d\tau \right\} \quad [\text{moles}] \quad \{44\}$$

where

$P_{bhijk'}$ (a function of time, τ) is given by Eq. {3'} with k' replacing k . The term $P_{bhink'}$ is for the last time step, which has the index n ,

and

$\zeta_{hijk'}$ (a function of time, τ) is given by Eq. {11}. It is for time step j and segment k' . Likewise $\eta_{hink'}$ is for time step n and segment k' .

The equations and parameters for calculating the factor $\frac{H_{hjk}(A/\ell)\ell_k}{R_g T_{cjk}(n_{He}/P_{He})_k}$,

which appears in the equation for $\zeta_{hijk'}$, {11}, are given in Appendix B.

APPENDIX A

Partial Pressure Equations for Metallic Fission Product Isotopes Sorbed by Graphite or Amorphous Carbon

We have found that for the sorption of certain metallic elements (including Rb, Sr, Cs, Ba) at low concentrations in graphite or amorphous carbon (i.e., below monolayer coverage at internal BET surface area), and for a single element, the Freundlich sorption isotherm is followed over the entire range of experimental data.* The data is expressed empirically by the equation

$$\ln P_h = v_h + u_h \ln C_h \quad \{1A\}$$

where

$$v_h = a_h + b_h \left(\frac{10^3}{T} \right) \quad \{2A\}$$

$$u_h = c_h + d_h \left(\frac{10^3}{T} \right) \quad \{3A\}$$

P_h = equilibrium partial pressure of chemical element h [atm.]

C_h = concentration of chemical element h sorbed in graphite
[μmoles/gm]

T = temperature of the sorbent (graphite or amorphous carbon)
[°K]

a_h, b_h, c_h, d_h are empirical constant characteristic of the sorbent material.

The constants obtained for TS-688 graphite (needle coke, National Carbon Co.) are given in Table A1.

* "Public Service Company of Colorado 330-MW(e) High-Temperature Gas-Cooled R and D Program, Quarterly Progress Reports," USAEC Reports GA-7634, GA-7453, and GA-7086, General Atomic Division, General Dynamics Corp. (1967, 1966). Also, C. E. Milstead, A. B. Riedinger, and L. R. Zumwalt, "Cesium-Graphite Sorption Isotherms Determined by a Static Method", Carbon, 4, 99-106 (1966).

Table A1

Freundlich Isotherm Constant for Alkali Metal and
Alkaline Earth Fission Products Sorbed by
TS-688 Graphite

Element h	a _h	b _h	c _h	d _h
Rb	14.1	-34.1	-9.0	16.8
Sr	10.00	-38.1	0.079	4.10
Cs	26.6	-55.7	-14.5	24.2
Ba	6.48	-33.7	-1.304	7.55

More conventionally the Freundlich isotherm is expressed by the equation*

$$\frac{C_h}{C_{oh}} = (k_h P_h)^{\frac{1}{n}} \quad \{4A\}$$

Now define

$$P_{oh} \equiv \frac{1}{k_h} \text{ (a constant for a given h)} \quad [\text{atm}]$$

and

$$C_{oh} \equiv \text{the concentration at which } P_h = P_{oh} \text{ which is independent of temperature (i.e., n)} \quad [\mu \text{ moles/gm}]$$

and

$$n = u_h = c_h + d_h (10^3/T)$$

We can write the conventional Freundlich isotherm equation as,

$$\ln (P_h/P_{oh}) = u_h \ln (C_h/C_{oh}) \quad \{5A\}$$

* See for example, Hayward, D. O., and Trapnell B. M. W., "Chemisorption," 2nd Ed. Butterworth, Washington (1964).

Substituting {1A} into {5A}

$$\ln P_{oh} = v_h + u_h \ln C_{oh}$$

and from {2A} and {3A}

$$\ln P_{oh} = a_h + b_h \left(\frac{10^3}{T} \right) + \left[c_h + d_h \left(\frac{10^3}{T} \right) \right] \ln C_{oh}$$

Since P_{oh} and C_{oh} are constants (independent of T)

$$\ln C_{oh} = - \frac{b_h}{d_h} \quad \{6A\}$$

and

$$\ln P_{oh} = a_h - c_h \left(\frac{b_h}{d_h} \right) \quad \{7A\}$$

One might assume that C_{oh} is the concentration corresponding to a monolayer of coverage of the internal surface area of the sorbent (chemisorption by its very nature cannot extend to more than a monolayer – see Hayward and Trapnell, loc. cit.). However the value of C_{oh} calculated from {6A} or

$$C_{oh} = \exp(-b_h/d_h) \quad \{6A'\}$$

does not agree well with the monolayer coverage, C_{mh} , calculated from known atomic diameters by assuming atoms to cover the BET area ($\sim 1.0 \text{ m}^2/\text{g}$ for TS-688 graphite) in hexagonal close packing. The comparison is given in Table A2.

In view of the wide variation (and unrealistically high values in the case of Sr and Ba) of C_{oh} , it would seem better to define surface coverage in terms of C_{mh} , the "geometric" monolayer concentration. Also the use of C_{mh} gives fairly reasonable values for the vapor pressure (calc. from {9A}) of h in the monolayer state.

Table A2
Comparison of C_{mh} and C_{oh}

h	Atomic Diameter* (Å)	C_{mh} ($\frac{\mu\text{moles}}{\text{gm}}$)	C_{oh} ($\frac{\mu\text{moles}}{\text{gm}}$)
Rb	4.88	8.1	6.8
Sr	4.31	10.4	8800.
Cs	5.25	7.0	9.8
Ba	4.35	10.2	84.

*Barrett, C.S., "Structure of Metals," McGraw-Hill Book Co., 1952.

Then the Freundlich sorption equation for vapor pressure is given by

$$P_h = P_{mh} \left\{ \frac{C_h}{C_{mh}} \right\}^{u_h} \quad [\text{atm.}] \quad \{8A\}$$

where

$$P_{mh} = \exp \left[A_{mh} + B_{mh} \left(\frac{10^3}{T} \right) \right] \quad [\text{atm.}] \quad \{9A\}$$

with

$$A_h = a_h + c_h \ln C_{mh} \quad \{10A\}$$

$$B_h = b_h + d_h \ln C_{mh} \quad \{11A\}$$

where in the case of TS-688 graphite as sorbent a_h , b_h , c_h , d_h are as given in Table 1A and C_{mh} as given in Table 2A.

The vapor pressure equation for a single isotope of h(hi) is given by

$$P_{hi} = P_{mh} \frac{C_{hi}}{C_{mh}} \left\{ \sum_{i'} \frac{C_{hi'}}{C_{mh}} \right\}^{u_h-1} \quad [\text{atm.}] \quad \{12A\}$$

where

$$C_{hi} = \text{concentration of hi} \quad [\mu\text{moles/gm}]$$

and the concentration of h is given by

$$C_h = \sum_{i'} C_{hi'} \quad [\mu\text{moles/gm}] \quad \{13A\}$$

In the case multicomponent sorption we assume the following equation approximates the partial pressure of P_{hi}

$$P_{hi} = P_{mh} \frac{C_{hi}}{C_{mh}} \left\{ \sum_{h'} \sum_{i'} \frac{C_{h'i'}}{C_{mh'}} \right\}^{(u_h - 1)} \quad [\text{atm.}] \quad \{14A\}$$

where h' represents all the sorbed chemical elements present and $h'i'$ represents all of the isotopes including hi .

Equation for w_{hk}

w_{hk} is a distribution coefficient (a function of chemical element h , and fuel temperature, T_F , and graphite-fuel interface temperature, T_{G0}) so that under conditions of quasi thermodynamic equilibrium conditions ($P_{hi} \approx \text{constant}$), the relationship between $\bar{C}_{chik}$ and $C_{hik}(0)$ is approximately as follows (Eq. {16} of text)

$$\bar{C}_{Chik} = w_{hk} C_{hik}(0) \quad \{15A\}$$

Under isothermal conditions

$$w_{hk} = K_h(T_k)$$

where

$K_h(T_k)$ = distribution coefficient, i. e. the sorbent capacity of the carbon material in the fuel holes relative to the sorbent capacity of the fuel element graphite (for chemical element h , at temperature (T_k))

where

$$C_{hi} = \text{concentration of } h_i \quad [\mu\text{moles/gm}]$$

and the concentration of h is given by

$$C_h = \sum_{i'} C_{hi'} \quad [\mu\text{moles/gm}] \quad \{13A\}$$

In the case multicomponent sorption we assume the following equation approximates the partial pressure of P_{hi}

$$P_{hi} = P_{mh} \frac{C_{hi}}{C_{mh}} \left\{ \sum_{h'} \sum_{i'} \frac{C_{h'i'}}{C_{mh'}} \right\}^{(u_h - 1)} \quad [\text{atm.}] \quad \{14A\}$$

where h' represents all the sorbed chemical elements present and $h'i'$ represents all of the isotopes including hi .

Equation for w_{hk}

w_{hk} is a distribution coefficient (a function of chemical element h, and fuel temperature, T_F , and graphite-fuel interface temperature, T_{G0}) so that under conditions of quasi thermodynamic equilibrium conditions ($P_{hi} \approx \text{constant}$), the relationship between $\bar{C}_{chik}$ and $C_{hik}(0)$ is approximately as follows (Eq. {16} of text)

$$\bar{C}_{Chik} = w_{hk} C_{hik}(0) \quad \{15A\}$$

Under isothermal conditions

$$w_{hk} = K_h(T_k)$$

where

$K_h(T_k)$ = distribution coefficient, i. e. the sorbent capacity of the carbon material in the fuel holes relative to the sorbent capacity of the fuel element graphite (for chemical element h, at temperature (T_k))

K_{hk} is determined experimentally and is taken to depend on the temperature and two empirical constants E_h and F_h which, in general, are different for each h .

Thus

$$K_h(T_k) = E_h + F_h \left(\frac{10^3}{T} \right) \quad \{16A\}$$

where

T_k = the temperature of the graphite and the carbon segment k . [$^{\circ}K$]
There may be little temperature dependence of K_h and thus $K_h \approx E_h$ (a constant). A typical value of K_h is 300.

In the case where the carbon temperature $T_{Fk} \neq T_{GOk}$ (graphite temperature) we calculate the ratio of the concentration of isotope hi that it would have at temperature T_{Fk} relative to the value it would have at temperature T_{GOk} with P_{hi} being unchanged.

For this condition we obtain from {14A} the ratio (which is independent of i)

$$\frac{C_h(T_{Fk})}{C_h(T_{GOk})} = \frac{P_{mh}(T_{GOk}) \left\{ \sum_{h'} \sum_{i'} \frac{\xi_{h'i'} \bar{C}_{h'i'}}{C_{mh'}} \right\}^{(u_h(T_{GOk}) - 1)}}{P_{mh}(T_{Fk}) \left\{ \sum_{h'} \sum_{i'} \frac{\xi_{h'i'} \bar{C}_{h'i'}}{C_{mh'}} \right\}^{(u_h(T_{Fk}) - 1)}}$$

Where $P_{mh}(T)$ is given by Eq. {9A} and $u_h(T)$ is given by Eq. {3A}

The values of $\xi_{h'i'}$ and $\bar{C}_{h'i'}$ in the brackets are taken from the previous values in the Hamde code calculation and considered constant in the immediate Hamde calculation.

APPENDIX B

The Mass Transport Coefficient and the Factor in the Equation for Zeta

In accordance with the analogy between mass transfer and heat transfer, the following is the equation for H_{hjk} in the case of turbulent flow*

$$H_{hjk} = 0.023 \left[\frac{8.64 \times 10^4}{\pi(d_o^2 - d_i^2)} \right] \left[\frac{m_{jk}}{P_{Hejk}} \right] \left[\frac{R_g T_{cjk}}{Sc_{hjk}^{0.67} Re_{jk}^{0.20}} \right] \quad [\text{cm/day}] \quad \{1B\}$$

d_o = outside diameter of coolant channel [cm]

d_i = inside diameter of coolant channel [cm]

(this = 0 in PSC fuel element case)

m_{jk} = coolant channel mass flow rate, time step j, segment k
[g/sec]

P_{Hejk} = pressure of coolant (He), time step j, segment k [atm]

R_g = gas constant, 82.06 [cm³-atm/mole °K]

T_{cjk} = temperature of coolant gas, time step j, segment k

Sc = Schmidt number

$$Sc_{hjk} = \frac{\mu_{Hejk}}{\rho_{Hejk} D_{Hejk}} \quad [\text{dimensionless}] \quad \{2B\}$$

where

$$\mu_{Hejk} = 4.17 \times 10^{-6} [T_{cjk}]^{0.678}, \text{ the viscosity of helium } \left[\frac{\text{g}}{\text{cm sec}} \right] \quad \{3B\}$$

* T. K. Sherwood and R. L. Pigford, "Absorption and Extraction," McGraw-Hill, New York, 1952. W. M. Rohsenow and H. Choi, "Heat, Mass and Momentum Transfer," Prentice-Hall Inc., New Jersey (1961).

$$\rho_{\text{Hejk}} = \text{density of coolant (He)} = 4 P_{\text{Hejk}} / R_g T_{\text{cjk}} \quad \left[\frac{\text{g}}{\text{cm}^3} \right] \quad \{4B\}$$

$$D_{\text{Hehjk}} = \frac{D_{\text{oHeh}} (T_{\text{cjk}})^{1.75}}{P_{\text{Hejk}}} \quad \text{the mutual diffusion coefficient of h in helium,} \\ [\text{cm}^2/\text{sec}] \quad \{5B\}$$

$$D_{\text{oHeh}} = \text{mutual diffusion coefficient of h in helium at 1 atm and } 0^\circ\text{C} \\ [\text{cm}^2/\text{sec}]$$

Re = Reynolds number

$$\text{Re}_{\text{jk}} = \frac{4 m_{\text{jk}}}{\pi (d_o + d_i) \mu_{\text{Hejk}}} \quad [\text{dimensionless}] \quad \{6B\}$$

From {1B} the factor in the zeta equation {11} is

$$H_{\text{hjk}} \frac{(A/\ell) \ell_k}{R_g T_{\text{cjk}} (n_{\text{He}}/P_{\text{He}})_{\text{jk}}} \\ = \left[\frac{0.023}{(n_{\text{He}}/P_{\text{He}})_{\text{jk}}} \right] \left[\frac{8.64 \times 10^4}{\pi (d_o^2 - d_i^2)} \right] \left[\frac{m_{\text{jk}}}{P_{\text{Hejk}}} \right] \left[\frac{(A/\ell) \ell_k}{\text{Sc}_{\text{hjk}}^{0.67} \text{Re}_{\text{jk}}^{0.20}} \right] \quad \{7B\}$$

APPENDIX C

Modification of FREVAP Program for more than one type of Fuel Particle

For the case where we have more than one type of fuel particle we assume their release rates can be given by different R values.

Let

f_{kn} = fraction of type n particles in segment k

then

$$\sum_n f_{kn} = 1 \quad \{1C\}$$

and the equation for the moles of isotope h_i , N_{hikm} , in fuel particles of type m in segment k is given by:

$$\frac{d N_{hikn}}{dt} = f_{kn} B_{hijk} - (\lambda_{hi} + R_{hjk n}) N_{hikn} \quad \{2C\}$$

where

B_{hijk} = total production rate of isotope h_i in segment k

and

$$\ln R_{hjk n} = \ln R_{ohn} - \frac{E_{ahn}}{R_q} \left(\frac{10^3}{T_{Fjk}} \right) \quad \{3C\}$$

with

R_{ohn} = Empirical release constant for chemical element h in type n fuel particles $[\text{day}^{-1}]$

E_{ahn} = Empirical activation energy for release of h from n type particles $[\text{kcal/mole}]$

R_q = Gas constant = 1.987 $[\text{cal/mole } ^\circ\text{K}]$

T_{Fjk} = mean temperature of fuel particles in k segment and time
step j:

The source term is then

$$S_{hjk} = \sum_n R_{h j k n} N_{h i k n} \quad \{4C\}$$

APPENDIX D

Equations for Diffusion of Metallic Fission Product Isotopes in Graphite

The equation for the flux of isotope h_i (neglecting any effect due to thermodynamic activity coefficient gradients) when a thermal gradient is present is given by:

$$J_{hi} = - \frac{D_h \rho}{10^6} \left\{ \frac{d C_{hi}}{dx} - C_{hi} \frac{10^3 Q_{th}}{R_q} \frac{d(1/T)}{dx} \right\} \left[\frac{\text{moles}}{\text{cm}^2 \text{ sec}} \right] \quad \{1D\}$$

ρ = density of graphite $[\text{gm}/\text{cm}^3]$

D_h = tracer diffusion coefficient $[\text{cm}^2/\text{sec}]$

Q_{th} = heat of transport $[\text{kcal}/\text{mole}]$

C_{hi} = conc. of h_i (a function of time, t , and position, x) $[\mu\text{moles}/\text{gm}]$

R_q = gas constant, $1.987 [\text{calories}/\text{mole } ^\circ\text{K}]$

We take that as a reasonable approximation based on empirical data on the tracer diffusion coefficient that

$$\ln D_h \approx \ln D_{Gh}^o = \ln D_h^o - \frac{Q_{th}}{R_q} \left(\frac{2 \times 10^3}{T_{G0} + T_{GL}} \right) + \delta_h \sum_{h'} \sum_i \bar{C}_{h'i'} \left(\frac{C_{mh}}{C_{mh'}} \right) \quad \{2D\}$$

where D_h^o , Q_{dh} (activation energy for diffusion), δ_h are empirical constants (typical values of these constants are listed in Table D1, pg. 6-D)) and

$$\frac{10^3 d(1/T)}{dx} \approx G_T \equiv \frac{\left(\frac{10^3}{T_{GL}} \right) - \left(\frac{10^3}{T_{G0}} \right)}{L} \quad \{3D\}$$

where

T_{G0} = graphite temperature at $x = 0$ $[^\circ\text{K}]$

T_{GL} = graphite temperature at $x = L$ $[^\circ\text{K}]$

L = thickness of graphite $[\text{cm}]$

Then approximately

$$J_{hi} = - \frac{D_{\bar{G}h} \rho}{10^6} \left\{ \frac{d C_{hi}}{dx} - C_{hi} \left(\frac{Q_{ht}}{R_q} \right) G_T \right\}$$

Let

$$\Gamma_h = \left(\frac{Q_{th}}{R_q} \right) G_T \quad \{5D\}$$

then

$$J_{hi} = - \frac{D_{\bar{G}h} \rho}{10^6} \left\{ \frac{d C_{hi}}{dx} - C_{hi} \Gamma_h \right\} \quad \{6D\}$$

The continuity equation for the diffusion of isotope hi in the graphite is given (when λ_{hi} = decay constant of hi [sec^{-1}]) by

$$\begin{aligned} \frac{\partial C_{hi}}{\partial t} &= - \frac{\partial J_{hi}}{\partial x} \left(\frac{10^6}{\rho} \right) - \lambda_{hi} C_{hi} \\ &= D_{\bar{G}h} \left\{ \frac{d^2 C_{hi}}{dx^2} - \Gamma_h \frac{d C_{hi}}{dx} \right\} - \lambda_{hi} C_{hi} \end{aligned} \quad \{7D\}$$

For the steady state, $\partial C_{hi} / \partial t = 0$

and

$$\frac{d^2 C_{hi}(x)}{dx^2} = \Gamma_h \frac{d C_{hi}(x)}{dx} + \frac{\lambda_{hi}}{D_{\bar{G}h}} C_{hi}(x) \quad \{8D\}$$

To solve this equation we make use of the Laplace transformation.*
The Laplace transforms are

$$\mathcal{L} \frac{d^2 C_{hi}(x)}{dx^2} = s^2 \tilde{C}_{hi}(s) - s C_{hi}(0) - \frac{d C_{hi}(0)}{dx} \quad \{9D\}$$

$$\mathcal{L} \frac{d C_{hi}(x)}{dx} = s \tilde{C}_{hi}(s) - C_{hi}(0) \quad \{10D\}$$

$$\mathcal{L} C_{hi}(x) = \tilde{C}_{hi}(s) = \int_0^{\infty} \exp(-sx) C_{hi}(x) dx \quad \{11D\}$$

Now at the fuel-graphite interface the flux of isotope hi,

$$F_{hi} = - \frac{D_{Goh} \rho}{10^6} \left\{ \frac{d C_{hi}(0)}{dx} - \Gamma_h C_{hi}(0) \right\} \quad \{12D\}$$

Thus

$$\frac{d C_{hi}(0)}{dx} = - \frac{10^6 F_{hi}}{\rho D_{Goh}} + \Gamma_h C_{hi}(0) \quad \{13D\}$$

Now we define

$$\Omega_{hi} \equiv \frac{10^6 F_{hi}}{\rho D_{Goh} C_{hi}(0)} \quad \{14D\}$$

* Thomson, W. T., "Laplace Transformation", Prentice-Hall, Inc., New York (1950).

then

$$\frac{d C_{hi}(0)}{dx} = - C_{hi}(0) (\Omega_{hi} - \Gamma_h) \quad \{15D\}$$

Now we substitute the Laplace transforms {9D}, {10D}, and {11D} into {8D} and use {15D} to express $d C_{hi}(0)/dx$ in terms of $C_{hi}(0)$.

We then have

$$\begin{aligned} s^2 \tilde{C}_{hi}(s) - s C_{hi}(0) + C_{hi}(0) (\Omega_{hi} - \Gamma_h) \\ = \Gamma_h (s \tilde{C}_{hi}(s) - C_{hi}(0)) + \frac{\lambda_{hi}}{D_{Gh}} \tilde{C}_{hi}(s) \end{aligned}$$

or

$$\tilde{C}_{hi}(s) \left[s^2 - \Gamma_h s - \frac{\lambda_{hi}}{D_{Gh}} \right] = C_{hi}(0) s - C_{hi}(0) \Omega_{hi} \quad \{16D\}$$

Let

$$\alpha - \beta = \Gamma_h \quad \{17D\}$$

$$\alpha\beta = \frac{\lambda_{hi}}{D_{Gh}} \quad \{17D'\}$$

Then

$$s^2 - \Gamma_h s - \frac{\lambda_{hi}}{D_{Gh}} = s^2 - (\alpha - \beta)s - \alpha\beta = (s - \alpha)(s - \beta)$$

and

$$\alpha = \sqrt{\left(\frac{\Gamma_h}{2}\right)^2 + \frac{\lambda_{hi}}{D\bar{G}_h}} + \frac{\Gamma_h}{2} \quad \{18D\}$$

$$\beta = \sqrt{\left(\frac{\Gamma_h}{2}\right)^2 + \frac{\lambda_{hi}}{D\bar{G}_h}} - \frac{\Gamma_h}{2} \quad \{18'D\}$$

(strictly speaking α and β should have the subscripts hi but these are omitted for convenience in writing the following)

Equation {16D} can be written

$$\tilde{C}_{hi}(s) [(s - \alpha)(s - \beta)] = C_{hi}(0) [s - \Omega_{hi}]$$

or

$$\tilde{C}_{hi}(s) = C_{hi}(0) \left[\frac{s}{(s - \alpha)(s - \beta)} - \frac{\Omega_{hi}}{(s - \alpha)(s - \beta)} \right] \quad \{17'D\}$$

Performing the inverse transformation of {18B}

$$C_{hi}(x) = \frac{C_{hi}(0)}{\alpha + \beta} \left[\alpha e^{\alpha x} + \beta e^{-\beta x} - \Omega_{hi} (e^{\alpha x} - e^{-\beta x}) \right] \quad \{19D\}$$

Differentiating

$$\frac{d C_{hi}(x)}{dt} = \frac{C_{hi}(0)}{\alpha + \beta} \left[\alpha^2 e^{\alpha x} - \beta^2 e^{-\beta x} - \Omega_{hi} (\alpha e^{\alpha x} + \beta e^{-\beta x}) \right] \quad \{20D\}$$

These equations correspond to {23} and {24} when $\alpha = \beta = \sqrt{\frac{\lambda_{hi}}{D\bar{G}_h}}$

and to {33} and {34} otherwise.

Table D1
DIFFUSION COEFFICIENTS FOR Sr, Ba AND Cs IN
FUEL ELEMENT GRAPHITE

h	$\ln D_{oh} (\text{cm}^2/\text{sec})$	$Q_{dh} (\text{kcal/mole})$	$\delta_h (\text{g}/\mu\text{mole})$
Sr	5.7	60.	0.25
Ba	7.4	70.	0.25
Cs	-1.0	39.	0.20

These constants were obtained from an evaluation of diffusion data obtained at General Atomic (reported in Project Progress Reports, i. e., GA-6113 and by E. Anderson and L. Zumwalt, Trans. Am. Nucl. Soc. 7, No. 2, 451-52 (1964) and by E. Bryant, et al, Nucl. Sci and Eng 15, 288-295 (1963)). They are subject to change as more experimental data becomes available.