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Mathematical Expressions for the Fractional Release of a Fission Product from an HTR Nuclear Fuel Element

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1. Introduction

In a high temperature reactor (HTR), a few amount of fission products (FPs) are released from a coated fuel particle (CFP) which usually consists of a fuel kernel and four surrounding coating layers, through a graphite matrix, and finally into a coolant. The quantity called fractional release (or release fraction) is used as a measure quantifying the FP release in many HTR fuel performance analysis code [1]. The fractional release of FPs under irradiation and simulated accident conditions are compared between codes, and with experimental data.

This study describes the mathematical expressions of the FP releases from an HTR fuel element, and the analytical forms of fractional releases for the radioactive and stable isotopes diffusing out of a spherical particle.

2. Fractional release formulae

Table I and II list the mathematical expressions for the fractional releases of radioactive and stable fission products, respectively [2-5]. The fractional release rate H means the ratio of the total rate at which atoms of an isotope are being released to the total rate at which they are being produced. It cannot be defined in the heating phase because no atoms are produced during heating. The fractional release rate at equilibrium is $\frac{3}{\sqrt{\mu}}\left(coth\sqrt{\mu}-\frac{1}{\sqrt{\mu}}\right)$ for a radioactive isotope or unity for a stable isotope, corresponding to infinite time.

Under irradiation, for a radioactive isotope, the fractional release F is the instantaneous ratio of the amount of external nondecayed atoms to the amount of nondecayed atoms in the system (a nuclear fuel and its exterior). It becomes the ratio of the amount of atoms released to the amount of atoms produced, for a stable isotope. The fractional release at equilibrium is $\frac{3}{\sqrt{\mu}}\left(coth\sqrt{\mu}-\frac{1}{\sqrt{\mu}}\right)$ for a radioactive isotope or unity for a stable isotope, as in the case of the fractional release rate.

Under heating, for a radioactive isotope, the intrinsic fractional release F^{INT} is the instantaneous ratio of the amount of external nondecayed atoms to the amount of atoms that exist in a nuclear fuel at the beginning of heating. It becomes the ratio of the amount of atoms released to the amount of atoms at the beginning of heating, for a stable isotope. The intrinsic fractional release at equilibrium is zero for a radioactive isotope and unity for a stable isotope, respectively. For the heating of an irradiated fuel, it is convenient to, instead of the intrinsic fractional release, use a fractional release which is based on the amount of atoms produced in the irradiation phase [6]:

$$F_{st} = \left[1 - F_{irr}^{(e)}\right] F_{st}^{INT}, \quad (1)$$

where F_{st} = the fractional release in a heating phase, $F_{irr}^{(e)}$ = the fractional release at the end of irradiation, and F_{st}^{INT} = the intrinsic fractional release in a heating phase.

The amounts released and produced, R_V and B_V, are actually computed numerically because of complex geometries, variable birth rates, and variable diffusivities:

$$R_V^{(n)} = R_V^{(n-1)} e^{-\lambda \Delta t^{(n)}} + \frac{\Delta t^{(n)}}{2}\left[\dot{R}_V^{(n-1)} e^{-\lambda \Delta t^{(n)}} + \dot{R}_V^{(n)}\right], \quad (2)$$

$$B_V^{(n)} = B_V^{(n-1)} e^{-\lambda \Delta t^{(n)}} + \frac{\Delta t^{(n)}}{2}\left[\dot{B}_V^{(n-1)} e^{-\lambda \Delta t^{(n)}} + \dot{B}_V^{(n)}\right], \quad (3)$$

where n = the time step and Δt = the time interval.

3. Fractional releases from a UO_2 kernel

The fractional releases of four radioactive isotopes, ^{137}Cs, ^{110m}Ag, ^{90}Sr and ^{85}Kr, from a UO_2 kernel of an HTR were calculated. The kernel was assumed to be a sphere. Table III shows the half-lives and fission yields of the above-mentioned radioactive isotopes. Table IV lists their diffusivities in a UO_2 kernel of an HTR [7].

Table III: Half-lives and fission yields of major radioactive isotopes

Isotope	Half-life	Fission yield (atom/fission)
^{110m}Ag	249.950 d	4.60×10^{-6}
^{137}Cs	30.17 y	9.37×10^{-3}
^{90}Sr	28.79 y	7.64×10^{-3}
^{85}Kr	10.756 y	3.84×10^{-4}

Table IV: Fission product diffusivities in a UO_2 kernel.

$D = D_{0,1} e^{-\frac{Q_1}{RT}} + D_{0,2} e^{-\frac{Q_2}{RT}}$
where R = the gas constant (8.314 J/(mol K)),
T = the temperature (K).

Isotope	D_{01} (m^2/s)	Q_1 (kJ/mol)	D_{02} (m^2/s)	Q_2 (kJ/mol)
^{110m}Ag	6.7×10^{-9}	165	-	-

^{137}Cs	5.6×10^{-8}	209	5.2×10^{-4}	362
^{90}Sr	2.2×10^{-3}	488	-	-
^{85}Kr	1.3×10^{-12}	126	-	-

It was assumed that a spherical UO_2 kernel with a diameter of 800 μm had been irradiated at 1000 ℃ for four years and then it had been heated at 1200 ℃ for 240 hours. The degree of diffusion in a spherical UO_2 kernel is larger in order of ^{110m}Ag, ^{137}Cs, ^{85}Kr and ^{90}Sr. Fig. 1 shows the fractional releases and their rates of the radioactive isotopes ^{137}Cs, ^{110m}Ag, ^{90}Sr and ^{85}Kr from a spherical UO_2 kernel during irradiation. All the fractional release rates are a little bit higher than the fractional releases. At the end of irradiation, the fractional releases are 78.5, 59.4, 0.029, and 17.3 % for ^{137}Cs, ^{110m}Ag, ^{90}Sr and ^{85}Kr, respectively. Fig. 2 shows the fractional releases during a safety test. At the end of heating, the fractional releases are 13.2, 13.7, 0.082, and 4.3 % for ^{137}Cs, ^{110m}Ag, ^{90}Sr and ^{85}Kr, respectively.

4. Summary

The mathematical fractional releases were classified systematically for radioactive and stable isotopes diffusing out of a fuel element under irradiation and accident conditions. Only when the diffusing medium is a single layer, it is possible to get an analytical form of fractional release. In other cases, the fractional releases must be calculated numerically. The choice of either the fractional release or the fractional release rate depends on whether the amount generated or the birth rate on which they are based on is more easily identifiable. According to the fractional releases calculated, the radioactive isotopes ^{110m}Ag, ^{137}Cs, ^{85}Kr and ^{90}Sr release from a spherical UO_2 kernel more in that order. The strontium release is negligible.

ACKNOWLEDGEMENTS

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Table I: Mathematical expressions for the fractional release of a radioactive fission product

C = concentration (atoms/m^3), $\dot{B}$ = volumetric birth rate (atoms/(m^3 s)), λ = decay constant (s^{-1}), D = diffusion coefficient (m^2/s), r = radial coordinate (m), t = time (s), z = 0 for a slab, 1 for a cylinder, 2 for a sphere, J = mass current, (atoms/(m^2 s)), $C^{(0)}$ = initial concentration (atoms/m^3), $C_V = \int_V CdV$ = amount in a volume (atoms), $\dot{R}_V$ = release rate (atoms/s), $\dot{B}_V = \int_V \dot{B}dV$ = birth rate (atoms/s), V = volume (m^3), R_V = amount released (atoms), B_V = amount generated (atoms), A_s = surface area (m^2), $\mu = \lambda a^2/D$, $\tau(t) = \int_0^t [D(r,x)/a^2]dx$, a = radial coordinate of the outermost location (m), $F_{irr}^{(e)}$ = fractional release at the end of irradiation.

Phases	Quantities	Fractional release formulae for a sphere with a constant D and a constant $\dot{B}$
Irradiation phase: $\frac{\partial C}{\partial t} = \dot{B} - \lambda C + \frac{1}{r^z}\frac{\partial}{\partial r}\left(r^z D \frac{\partial C}{\partial r}\right)$ $C(a,t) = C(r,0) = 0$ $J(0,t) = 0$ $\frac{dC_V}{dt} = \dot{B}_V - \lambda C_V - \dot{R}_V$ $B_V = C_V + R_V$	$\dot{R}_V = A_s J(a,t)$ $R_V = e^{-\lambda t}\int_0^t e^{\lambda x}\dot{R}_V dx$ $B_V = e^{-\lambda t}\int_0^t e^{\lambda x}\dot{B}_V dx$	$J(\mathrm{a,t}) = \frac{a}{3}\dot{B}\left[\frac{3}{\sqrt{\mu}}\left(coth\sqrt{\mu} - \frac{1}{\sqrt{\mu}}\right) - 6e^{-\mu\tau}\sum_{n=1}^{\infty}\frac{e^{-n^2\pi^2\tau}}{n^2\pi^2+\mu}\right]$ $H = \dot{R}_V/\dot{B}_V = \frac{\text{total rate at which atoms are being released}}{\text{total rate at which atoms are being produced}}$ $= \frac{3}{\sqrt{\mu}}\left(coth\sqrt{\mu} - \frac{1}{\sqrt{\mu}}\right) - 6e^{-\mu\tau}\sum_{n=1}^{\infty}\frac{e^{-n^2\pi^2\tau}}{n^2\pi^2+\mu}$ $\approx \begin{cases}\frac{3}{\sqrt{\mu}}erf\sqrt{\mu\tau} - \frac{3}{\mu}(1-e^{-\mu\tau}) & ,\tau \le 1/\pi^2 \\ \frac{3}{\sqrt{\mu}}\left(coth\sqrt{\mu} - \frac{1}{\sqrt{\mu}}\right) - \frac{6e^{-(\pi^2+\mu)\tau}}{\pi^2+\mu} & ,\tau > 1/\pi^2\end{cases}$ $F = R_V/B_V = \frac{\text{amout of external nondecayed atoms}}{\text{amount of nondecayed atoms in the system}}$ $= \frac{1}{1-e^{-\mu\tau}}\left[\frac{3}{\sqrt{\mu}}\left(coth\sqrt{\mu} - \frac{1}{\sqrt{\mu}}\right) - e^{-\mu\tau} + 6\mu e^{-\mu\tau}\sum_{n=1}^{\infty}\frac{e^{-n^2\pi^2\tau}}{n^2\pi^2(n^2\pi^2+\mu)}\right]$ $= \frac{1}{1-e^{-\mu\tau}}\begin{cases}\frac{3}{\sqrt{\mu}}\left(erf\sqrt{\mu\tau} - 2\sqrt{\frac{\mu\tau}{\pi}}e^{-\mu\tau}\right) - \frac{3}{\mu}[1-(1+\mu\tau)e^{-\mu\tau}] & ,\tau \le 1/\pi^2 \\ \frac{3}{\sqrt{\mu}}\left(coth\sqrt{\mu} - \frac{1}{\sqrt{\mu}}\right) - e^{-\mu\tau} + \frac{6\mu e^{-(\pi^2+\mu)\tau}}{\pi^2(\pi^2+\mu)} & ,\tau > 1/\pi^2\end{cases}$
Heating phase: $\frac{\partial C}{\partial t} = -\lambda C + \frac{1}{r^z}\frac{\partial}{\partial r}\left(r^z D \frac{\partial C}{\partial r}\right)$ $C(a,t) = 0, C(r,0) = C^{(0)}$ $J(0,t) = 0$ $\frac{dC_V}{dt} = -\lambda C_V - \dot{R}_V$ $0 = C_V - C_V^{(0)}e^{-\lambda t} + R_V$	$\dot{R}_V = A_s J(a,t)$ $R_V = e^{-\lambda t}\int_0^t e^{\lambda x}\dot{R}_V dx$ $C_V^{(0)} = \int_V C^{(0)}dV$	$J(\mathrm{a,t}) = \frac{2C^{(0)}D}{a}\sum_{n=1}^{\infty}e^{-(n^2\pi^2+\mu)\tau}$ $F^{INT} = R_V/C_V^{(0)} = \frac{\text{amout of external nondecayed atoms}}{\text{amount of atoms at the initial time of heating}}$ $= e^{-\mu\tau}\left(1 - 6\sum_{n=1}^{\infty}\frac{e^{-n^2\pi^2\tau}}{n^2\pi^2}\right) \approx e^{-\mu\tau}\begin{cases}6\sqrt{\tau/\pi} - 3\tau & ,\tau \le 1/\pi^2 \\ 1 - \frac{6}{\pi^2}e^{-\pi^2\tau} & ,\tau > 1/\pi^2\end{cases}$ $F = \left[1 - F_{irr}^{(e)}\right]F^{INT} = \frac{\text{amout of external nondecayed atoms}}{\text{amount of nondecayed atoms in the system at the end of irradiation}}$

Table II: Mathematical expressions for the fractional release of a stable fission product

C = concentration (atoms/m^3), $\dot{B}$ = volumetric birth rate (atoms/(m^3 s)), λ = decay constant (s^{-1}), D = diffusion coefficient (m^2/s), r = radial coordinate (m), t = time (s), z = 0 for a slab, 1 for a cylinder, 2 for a sphere, J = mass current, (atoms/(m^2 s)), $C^{(0)}$ = initial concentration (atoms/m^3), $C_V = \int_V CdV$ = amount in a volume (atoms), $\dot{R}_V$ = release rate (atoms/s), $\dot{B}_V = \int_V \dot{B}dV$ = birth rate (atoms/s), V = volume (m^3), R_V = amount released (atoms), B_V = amount generated (atoms), A_s = surface area (m^2), $\mu = \lambda a^2/D$, $\tau(t) = \int_0^t [D(r,x)/a^2]dx$, a = radial coordinate of the outermost location (m), $F_{irr}^{(e)}$ = fractional release at the end of irradiation.

Phases	Quantities	Fractional release formulae for a sphere with a constant D and a constant $\dot{B}$
Irradiation phase: $\frac{\partial C}{\partial t} = \dot{B} + \frac{1}{r^z}\frac{\partial}{\partial r}\left(r^z D \frac{\partial C}{\partial r}\right)$ $C(a,t) = C(r,0) = 0$ $J(0,t) = 0$ $\frac{dC_V}{dt} = \dot{B}_V - \dot{R}_V$ $B_V = C_V + R_V$	$\dot{R}_V = A_s J(a,t)$ $R_V = \int_0^t \dot{R}_V dx$ $B_V = \int_0^t \dot{B}_V dx$	$J(\mathrm{a,t}) = \frac{a}{3}\dot{B}\left(1 - 6\sum_{n=1}^{\infty}\frac{e^{-n^2\pi^2\tau}}{n^2\pi^2}\right)$ $H = \dot{R}_V/\dot{B}_V = \frac{\text{total rate at which atoms are being released}}{\text{total rate at which atoms are being produced}}$ $= 1 - 6\sum_{n=1}^{\infty}\frac{e^{-n^2\pi^2\tau}}{n^2\pi^2} \approx \begin{cases}6\sqrt{\tau/\pi} - 3\tau & ,\tau \le 1/\pi^2 \\ 1 - \frac{6}{\pi^2}e^{-\pi^2\tau} & ,\tau > 1/\pi^2\end{cases}$ $F = R_V/B_V = \frac{\text{amout of atoms released}}{\text{amount of atoms produced in the system}}$ $= 1 - \frac{1}{15\tau} + \frac{6}{\tau}\sum_{n=1}^{\infty}\frac{e^{-n^2\pi^2\tau}}{n^4\pi^4} \approx \begin{cases}4\sqrt{\frac{\tau}{\pi}} - \frac{3}{2}\tau & ,\tau \le 1/\pi^2 \\ 1 - \frac{1}{15\tau} + \frac{6}{\pi^2\tau}e^{-\pi^2\tau} & ,\tau > 1/\pi^2\end{cases}$
Heating phase: $\frac{\partial C}{\partial t} = \frac{1}{r^z}\frac{\partial}{\partial r}\left(r^z D \frac{\partial C}{\partial r}\right)$ $C(a,t) = 0, C(r,0) = C^{(0)}$ $J(0,t) = 0$ $\frac{dC_V}{dt} = -\dot{R}_V$ $0 = C_V - C_V^{(0)} + R_V$	$\dot{R}_V = A_s J(a,t)$ $R_V = \int_0^t \dot{R}_V dx$ $C_V^{(0)} = \int_V C^{(0)}dV$	$J(\mathrm{a,t}) = \frac{2C^{(0)}D}{a}\sum_{n=1}^{\infty}e^{-n^2\pi^2\tau}$ $F^{INT} = R_V/C_V^{(0)} = \frac{\text{amout of atoms released}}{\text{amount of atoms at the initial time of heating}}$ $= 1 - 6\sum_{n=1}^{\infty}\frac{e^{-n^2\pi^2\tau}}{n^2\pi^2} \approx \begin{cases}6\sqrt{\tau/\pi} - 3\tau & ,\tau \le 1/\pi^2 \\ 1 - \frac{6}{\pi^2}e^{-\pi^2\tau} & ,\tau > 1/\pi^2\end{cases}$ $F = \left[1 - F_{irr}^{(e)}\right]F^{INT} = \frac{\text{amout of atoms released}}{\text{amount of atoms produced in the system until the end of irradiation}}$

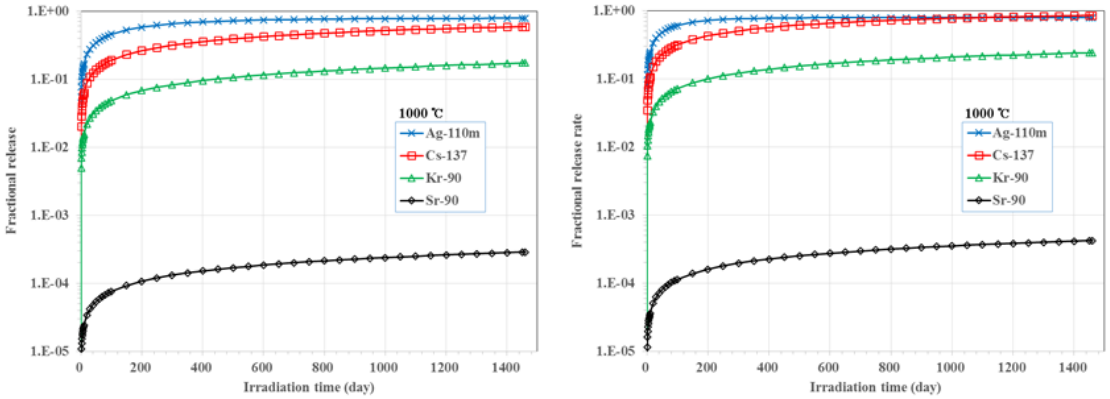

Fig. 1. Fractional release and its rate in a UO_2 kernel during irradiation.

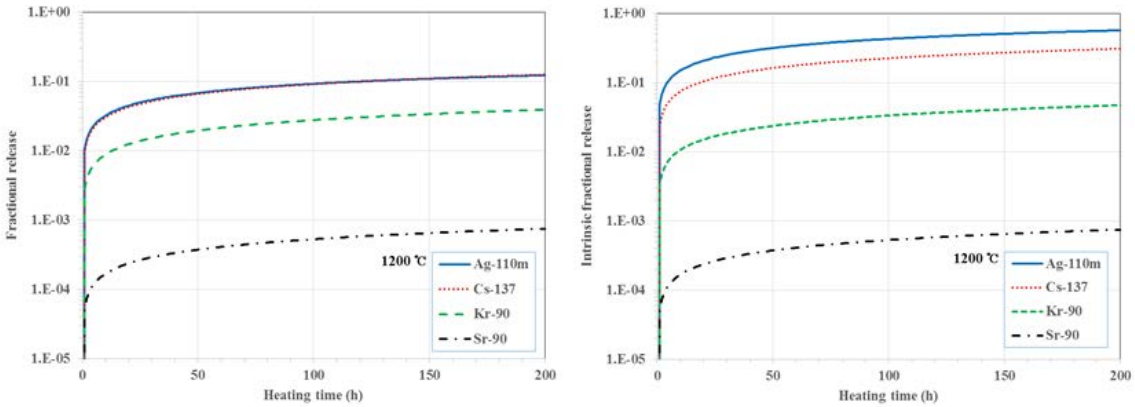

Fig. 2. Fractional releases from a UO_2 kernel during heating.