Technical Report

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Thermal spike theory of athermal diffusion of fission products due to alpha decay of actinides in spent fuel (UO_2)

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Radioactive decay of an actinide produces an alpha particle of typically 5 MeV and a recoil atom of ~100 keV. The alpha particle loses energy primarily by electronic stopping, which is quickly converted to heat. This is the origin of the "thermal spike" discussed by Matzke for fission fragments. Using the TRIM code, a 5 MeV alpha particle produces only 178 defects (vacancy-interstitial pairs), most of which are sufficiently widely separated that recombination is minimal. Figure 1 shows the TRIM output for a 5 MeV alpha particle entering UO_2 from the left. The black dots at the end of the brush-like displacement damage pattern (2000 ions were injected) represent the stopped alpha particles, which are helium atoms. Examination of the tracks on the periphery show widely-spaced dots, each of which represents a nuclear collision of the alpha particle with a U or O ion. The dots merge towards the end of the alpha particle range because the atom-ion cross section increases with decreasing energy.

The recoil heavy element from the decay event produces ~1200 displacements /1/. The range of the recoil is only 25 nm, so most of the vacancies and interstitials recombine. The effect of recoils on the athermal diffusion coefficient is probably small, and is neglected in this analysis. Matzke argues that the enhanced diffusivity due to the fission fragments arises from two sources: the thermal spike and the pressure spike. Displacement damage is a minor contributor.

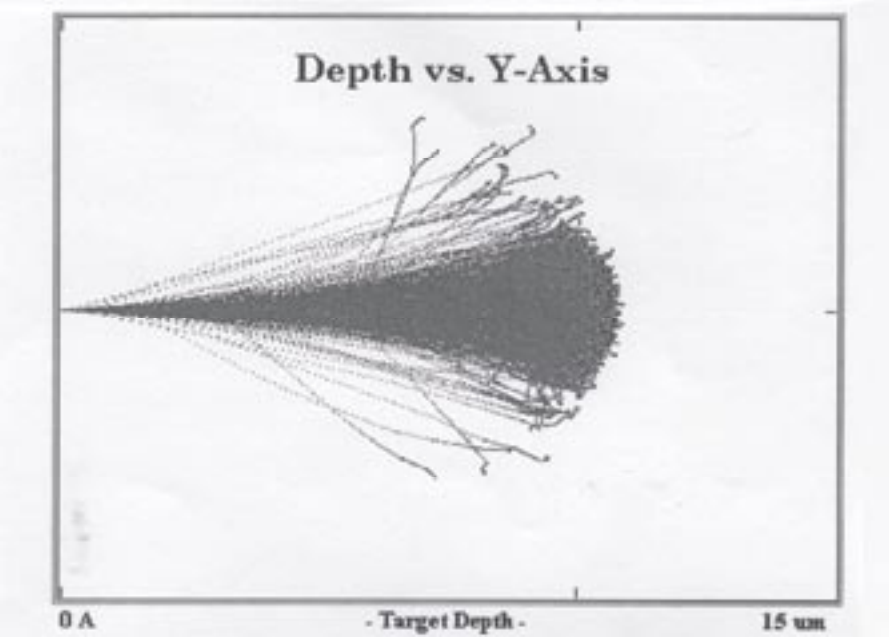

Figure 1. *Damage pattern in UO_2 produced by 5 MeV alpha particles.*

The present work assumes that an alpha particle slowing down in UO_2 possesses a constant electronic stopping power and negligible nuclear stopping (99.7% of the particle's energy is lost by ionization /1/). The slowing down creates a line source of heat equal to 8×10^{-14} J/μm over the 10 μm range of the 5 MeV alpha particle in UO_2.

The trajectories in Figure 1 do not look much like a line because the ordinate scale is much enlarged compared to the abscissa scale. The radial straggling is only ~0.6 μm.

Thermal spike due to 5 MeV Alpha Particle in UO_2

The transient temperature profile from a line source of heat is given by Carslaw and Jaeger /2/:

$$\Delta T = \frac{dE/dx}{4\pi kt}\exp\left(-\frac{r^2}{4\pi t}\right) \qquad (1)$$

where dE/dx is the energy released from the line source per unit length (see above) and k is the thermal conductivity of UO_2 (~0.03 W/cm-K). The radial distance from the line is r and t is the time. Inserting these numbers into Equation (1) yields:

$$\Delta T = \frac{2.12}{t}\exp\left(-280\frac{r^2}{t}\right) \qquad (2)$$

where r is in μm and t is in ns.

Equation (2) is plotted in Figure 2.

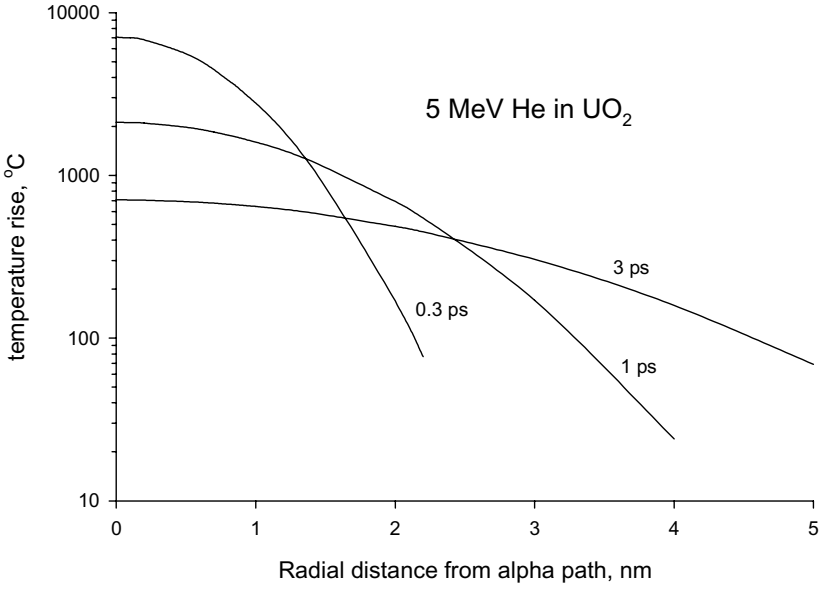

Figure 2. *Temperature rise in UO_2 resulting from passage of a 5 MeV alpha particle.*

Diffusion Coefficient

The "thermal" diffusion coefficient in a solid is given by the Einstein equation:

$$D = \tfrac{1}{6} a^2 \beta \nu \exp(-E/RT) = D_o \exp(-E/RT) \qquad (3)$$

where

a = jump distance

β = number of sites surrounding equilibrium site

ν = vibration frequency of atom in equilibrium site

E = activation energy for diffusion

R = gas constant

βν exp(–E/RT) = jump frequency per atom

The approach to athermal diffusion is to first determine the jumps of a solute atom (e.g., Xe) due to the alpha particle track from a single decay event. The jumps are caused by the momentary increase in temperature, as given by Equation (2). It is assumed that the base temperature of the spent fuel is low enough that the jump frequency, and hence the solute diffusivity, is negligible. A differential cylindrical volume $2\pi r dr L$, where L is the range of the alpha particle, contains $2\pi r dr L \rho_i$ atoms of species i, where ρ_i is the concentration of this species (atoms of i per unit volume). In this volume element and in time dt, the number of jumps of species i is:

$$dj_i = 2\pi r dr L \rho_i \, dt \nu_i \exp\left[-\frac{E_i / R}{\Delta T(r,t)} \right]$$

Integrating over r and t yields the jumps of species i per alpha particle:

$$j_i = \pi L \rho_i \nu_i \int_0^\infty dr^2 \int_0^\infty dt \exp\left[-\frac{E_i / R}{\Delta T(r^2,t)} \right] \qquad (4)$$

The spatial variable of integration has been switched from r to r^2 because the square appears in Equation (2).

The rate of disintegration of actinide n per unit volume is $\lambda_n N_n$, where λ_n is the decay constant and N_n is the atom density of the actinide. The number of jumps of fission product i per unit volume and per unit time due to decays of actinide n is $\lambda_n N_n j_i$. Dividing by the density of species i, ρ_i, gives the frequency of jumping of an atom of i, which is the equivalent of βνexp(–E/RT) in Equation (3). Multiplying Equation (4) by $a^2\beta/6$ yields the athermal diffusivity of species i due to alpha particles from actinide n:

$$D^*_{i,n} = (\tfrac{1}{6} a^2 \beta \nu_i) \pi L \lambda_n N_n \int_0^\infty dr^2 \int_0^\infty dt \exp\left[-\frac{E_i / R}{\Delta T(r^2,t)} \right]$$

From Equation (3), the terms in the parentheses in the above equation constitutes the pre-exponential factor of the Arrhenius form of the diffusivity of i, so the athermal diffusivity of fission product i due to actinide n is:

$$D^*_{i,n} = \pi D_{o,i} L \lambda_n N_n \int_0^\infty dr^2 \int_0^\infty dt \exp\left[-\frac{E_i / R}{\Delta T(r^2,t)} \right] \qquad (5)$$

For i = Xe, E_{Xe} = 293 kJ/mole /3/ and so the double integral in Equation (5) is equal to 6.54×10^{-12} μm²-ns, or 6.54×10^{-29} cm²-s. The pre-exponential factor is $D_{o,Xe} = 7.6\times10^{-6}$ cm²/s and the alpha particle range is $L = 10^{-3}$ cm. The athermal diffusivity of species i in the spent fuel is the sum of Equation (5) over all actinides:

$$D^*_{Xe} = \sum D^*_{Xe,n} = 1.6\times10^{-36}\sum_n \lambda_n N_n \qquad (6)$$

where λ_n is in s^{-1} and N_n is in atoms/cm³. Table 1 gives the components of the sum for PWR fuel with a burnup of 60 MWd/kgU after 20 years of cooling.

Table 1. Alpha activity in spent fuel (only nuclides with activities >10^7 dis/s-cm³ included).

Nuclide	Dis/s-cm³ × 10^{-8}
Pu 238	25.0
Pu 239	1.3
Pu 240	1.9
Am 241	22.8
Am 243	0.4
Cm 243	0.1
Cm 244	20.9
Total	72.4

Inserting the total alpha activity from Table 1 into Equation (6) gives an athermal diffusivity for Xe of:

$D^*_{Xe} = 1.2 \times 10^{-26}$ cm²/s

This value is five orders of magnitude smaller than the value estimated by Poinssot et al /4/.

References

/1/ H J Matzke, Solid State Phenomena, 30 (1993) 355

/2/ Carslaw and Jaeger, “Conduction of Heat in Solids”, p 258 Oxford Press (1959)

/3/ J A Turnbull and E Kolstad, Inter. Seminar on Fission Gas Behavior in Water Reactor Fuels, Paper No. 3.4, Cadarache. France (2000)

/4/ C Poinssot et al, Mat. Res. Soc. Symp. Proc. 713 (2002) 615

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