

3. Pore Migration Via Thermomigration-Driven Surface Diffusion

3.1 Introduction

Postirradiation examinations from experiments on annular, U-10Mo, helium-bonded, 55 percent smear density fuel [1] conducted by the Advanced Fuels Campaign [2] often show substantially greater porosity near the center of the fuel than near the fuel periphery; the degree of pore redistribution appears to increase with increasing temperatures (see Figure 3.1). These observations suggest that temperature gradient-driven material transport, commonly referred to as thermomigration, may contribute to the observed redistribution of porosity.

The hypothesis considered here is that pores migrate through the fuel by thermomigration along pore surfaces. The objectives of the present work are to demonstrate the capability to model this process explicitly, compare finite element predictions of pore migration with a corresponding analytic solution, and provide a basis for developing an engineering-scale representation of the same transport mechanism.

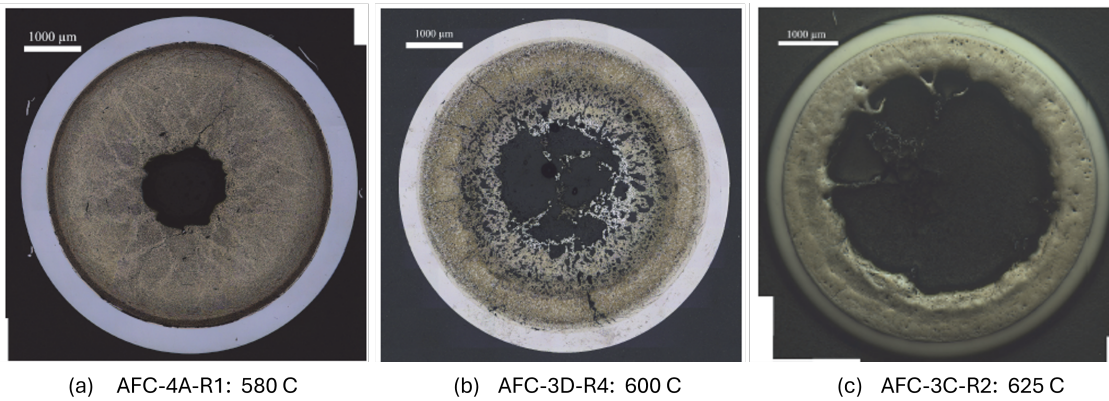


Figure 3.1: Post-irradiation metallography of annular metallic fuel at inner cladding temperatures of about (a) 580 °C; (b) 600 °C; and (c) 625 °C [1]. Note that with increasing temperatures, the mode of deformation appears to change from swelling dominated behavior where the central hole closes with increasing burnup to a mode dominated by mass transport towards the fuel periphery and a corresponding migration of porosity to the fuel interior [1].

3.2 Governing Equations

Surface diffusion is driven by gradients in chemical potential along a surface. The contributions to the chemical potential considered in the present formulation can be written as

$$\mu = \mu_0 + \mu_\psi + \mu_{\sigma_{nn}} + \mu_{TM}, \quad (3.1)$$

where

$$\mu_\psi = \Omega\psi, \quad \mu_{\sigma_{nn}} = -\Omega\sigma_{nn}, \quad (3.2)$$

and

$$\mu_{\text{TM}} = Q^* \ln \left(\frac{T}{T_{\text{ref}}} \right). \quad (3.3)$$

Here, Ω is the atomic volume, ψ is the elastic strain energy density, σ_{nn} is the normal stress acting on the diffusing surface, and Q^* is the heat of transport on a per atom basis. Note that the normal stress acting on the diffusing surface may include stresses produced by externally applied normal tractions as well as capillary stresses associated with surface energy and surface curvature. The thermomigration term is treated here as a pseudo-chemical potential; it provides a convenient scalar representation of the thermomigration driving force and allows the different transport mechanisms to be treated within the same framework.

Given expressions for the chemical potential, the surface flux is given by

$$\mathbf{J}_s = -M_s \nabla_s \mu, \quad (3.4)$$

where M_s is the surface mobility and ∇_s denotes the surface gradient. Conservation of material gives the normal rate of motion of the surface, v_n , as

$$v_n = -\Omega \nabla_s \cdot \mathbf{J}_s. \quad (3.5)$$

During the development of these capabilities in HeavyMetal, the finite element predictions were tested against classic analytic solutions by Mullins [3], by Asaro and Tiller [4], and by Grinfeld [5] (ATG). The Mullins problem isolates capillarity-driven surface diffusion and predicts smoothing of an initially perturbed surface, while the ATG solution adds the elastic strain energy contribution and predicts unstable surface growth when the elastic driving force is sufficiently large to overcome the stabilizing effect of surface energy. HeavyMetal reproduced the analytic surface evolution behavior for both problems, providing independent verification of the corresponding terms in equation (3.1).

3.3 Benchmark Unit Cell Model for a Translating Pore

The finite element benchmark considers a circular pore of radius a in a representative unit cell, as shown in Figure 3.2. Temperatures are prescribed on the left and right boundaries of the unit cell, while the transverse boundaries and the pore surface are thermally insulated. In the pore migration simulation, the normal stress and thermomigration terms in Equation (3.1) are retained. The normal stress term provides the capillary contribution that resists changes in pore shape, while thermomigration drives pore translation.

The approach taken for this benchmark is to estimate pore velocities both numerically and analytically, with both calculations using pore surface temperatures obtained from the finite element thermal solution. In this manner, the benchmark specifically tests the implementation of the surface diffusion equations in HeavyMetal; the heat conduction solution is not independently tested by this comparison. As will become evident, it will be helpful to write the pore surface temperatures in terms of a Fourier series, using cosine functions because of the

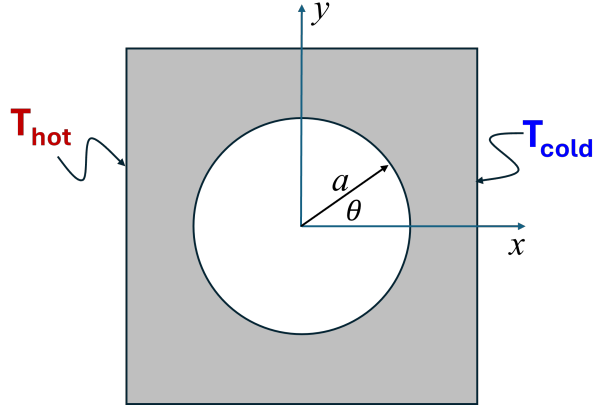


Figure 3.2: Schematic of the finite representative-cell model used to evaluate pore migration caused by thermomigration along the pore surface.

symmetry about the x-axis, i.e.,

$$T_s(\theta) = T_0 + \sum_{n=1}^{\infty} A_n \cos(n\theta), \quad (3.6)$$

For the relatively small temperature variations used in the benchmark, Equations (3.3)–(3.5), together with $s = a\theta$, give

$$v_n(\theta) = -\frac{K_T}{a^2} \sum_{n=1}^{\infty} n^2 A_n \cos(n\theta), \quad (3.7)$$

where, for convenience, the combination of material parameters appearing in the thermomigration term is written as

$$K_T = \Omega M_s \frac{Q^*}{T_{\text{ave}}}. \quad (3.8)$$

and where T_{ave} is taken to be the average of the temperatures imposed on the left and right sides of the unit cell.

The first Fourier mode in equation (3.7) represents rigid translation of a circular pore; higher modes produce changes in pore shape. Pore migration with velocity V_{pore} gives $v_n = -V_{\text{pore}} \cos \theta$, so the analytic translation velocity associated with the first temperature mode is

$$V_{\text{analytic}} = K_T \frac{A_1}{a^2}. \quad (3.9)$$

The coefficient A_1 is obtained directly from the predicted pore surface temperatures from the finite element model using

$$A_1 = \frac{2}{\pi} \int_0^{\pi} T_s(\theta) \cos \theta d\theta. \quad (3.10)$$

For a consistent comparison, the finite element normal velocities are projected onto the same mode,

$$V_{\text{FE}} = -\frac{2}{\pi} \int_0^{\pi} v_n^{\text{FE}}(\theta) \cos \theta d\theta. \quad (3.11)$$

Figure 3.3 shows results from a representative calculation. With the sign convention used here, a positive value of Q^* corresponds to migration of matter toward the cold side of the domain and, consequently, pore migration toward the hot side. As shown in the figure, the pore translates toward the hot side of the cell while remaining nearly circular, and the temperature field evolves with the changing pore position. When estimating pore velocities however, the pore was located in the center of the unit cell and only small translations were required to estimate pore velocities.

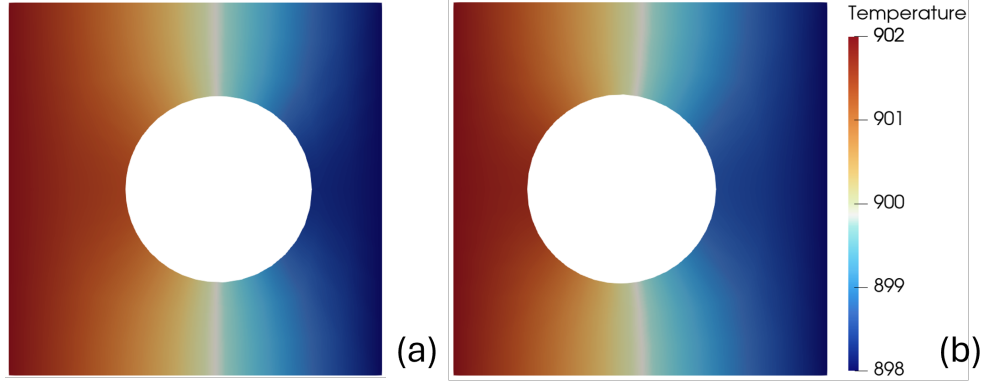


Figure 3.3: Representative thermomigration calculation showing the temperature field and pore position at (a) the beginning of the calculation; and (b) after appreciable pore translation. The pore migrates toward the hot side of the cell while remaining circular because of the effect of surface energy.

Calculations were performed for pore radii from 0.1 to 100 μm to estimate the effect of pore size on migration rates. As shown in Figure 3.4, the finite element and analytic first-mode translation velocities are nearly indistinguishable over the range of pore sizes considered and reproduce the expected inverse dependence of migration velocity on pore radius. The comparison provides a direct check of the conversion of the calculated pore surface temperature variation into thermomigration surface flux, material deposition and removal, and pore translation.

3.4 Homogenized Model for Engineering-Scale Analysis

Explicitly resolving individual pores is not practical in an engineering-scale fuel calculation. The microstructural-scale migration model can instead be homogenized by converting the motion of a population of pores into an equivalent volumetric flux of fuel alloy. Let $n(a) da$ denote the number of isolated pores per unit bulk volume having characteristic size between a and $a + da$, let $\mathcal{V}_p(a)$ denote the volume of one such pore, and let $\mathbf{V}_p(a)$ denote its migration velocity as obtained from the pore-scale model. The corresponding pore volume flux is

$$\mathbf{J}_p = \int n(a) \mathcal{V}_p(a) \mathbf{V}_p(a) da. \quad (3.12)$$

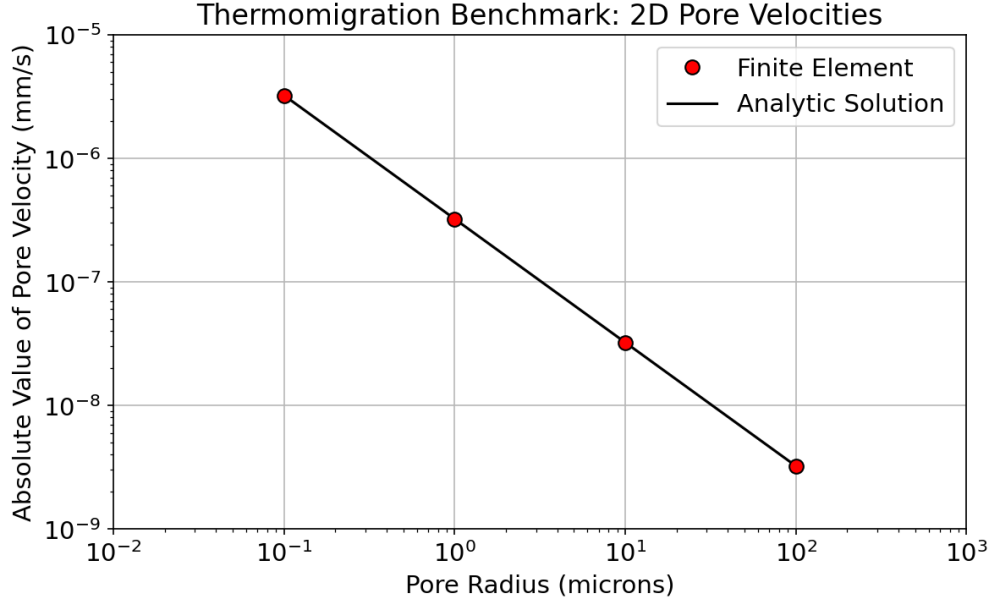


Figure 3.4: Comparison of finite element and analytic pore translation velocities for pore radii from 0.1 to 100 μm . The analytic velocity is based on the first Fourier mode of the finite element pore-surface temperature distribution.

Migration of pore volume in one direction corresponds to transport of an equal volume of fuel alloy in the opposite direction. The homogenized fuel alloy volume flux is therefore

$$\mathbf{J} = -\mathbf{J}_p = - \int n(a) \mathcal{V}_p(a) \mathbf{V}_p(a) da, \quad (3.13)$$

where $\mathbf{J}$ has units of volume of fuel alloy transported per unit bulk area per unit time. For a monodisperse population of pores, Equation (3.13) reduces to

$$\mathbf{J} = -v_f \mathbf{V}_p. \quad (3.14)$$

The corresponding atomic flux is $\mathbf{J}/\Omega$.

Equations (3.12)–(3.14) presently describe transport by isolated pores. The effects of pore interactions and pore interconnectivity are left for future work. Interconnected porosity could produce a larger effective volumetric flux because a connected pore network is more likely to contain transport pathways favorably aligned with the local temperature gradient; the magnitude of this effect will depend on network connectivity and tortuosity.

Once the pore-scale transport has been expressed as a macroscopic fuel alloy volume flux, its effect can be incorporated directly into expressions of conservation of volume. In the homogenized description, $\dot{\epsilon}_V$ will denote the volumetric strain rate of the fuel alloy/porosity composite, whereas $\dot{\epsilon}_V^{\text{solid}}$ will represent a kinematically effective volumetric strain rate of the solid fuel alloy caused by the fissioning of uranium and from local accumulation or depletion of fuel alloy caused by material transport. A review paper on electromigration [6] (mass transport driven

by high current densities) specifically identified the formulations of Povirk [7] and Rzepka et al. [8] as early approaches in which material accumulation or depletion was related to an inelastic strain rate through the divergence of material flux.

$$\dot{\epsilon}_V^{\text{solid}} = \dot{\epsilon}_V^{\text{solid}}(\dot{F}) - \frac{1}{1 - v_f} \nabla \cdot \mathbf{J}, \quad (3.15)$$

where $\dot{\epsilon}_V^{\text{solid}}(\dot{F})$ is the volumetric strain rate of the fuel alloy caused by the fissioning process, $\dot{F}$ is the fission density rate, and $\mathbf{J}$ is the fuel alloy volume flux defined above. The porosity evolution follows from the relative volumetric strain rates of the composite and the fuel alloy,

$$\dot{v}_f = (1 - v_f) (\dot{\epsilon}_V - \dot{\epsilon}_V^{\text{solid}}). \quad (3.16)$$

In the absence of fuel alloy swelling and material transport, Equation (3.16) reduces to the familiar evolution equation for pore volume fraction used by Tvergaard and Needleman [9]. Combining Equations (3.15) and (3.16) gives

$$\dot{v}_f = (1 - v_f) \left[\dot{\epsilon}_V - \dot{\epsilon}_V^{\text{solid}}(\dot{F}) \right] + \nabla \cdot \mathbf{J}. \quad (3.17)$$

Equation (3.17) provides the connection between the pore-scale thermomigration calculation and an engineering-scale fuel model. Fissioning changes the volume of the fuel alloy, deformation changes the volume of the fuel-alloy/porosity composite, and divergence of the homogenized fuel-alloy flux redistributes fuel alloy and therefore porosity. The present formulation has not yet been implemented in HeavyMetal; implementation, extension to interconnected porosity, and integration with the fission-gas swelling and tearing-induced cavitation models described elsewhere in this proposal are planned future work.

References

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