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Modeling of Irradiation Temperatures and Constituent Redistribution in U-10Zr Metallic Fuel

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Abstract

The computational scheme on a irradiation temperature of U-10Zr fuel was established considering porosity formation, bond sodium infiltration and constituent redistribution. Thermotransport theory was adapted to model the redistribution phenomenon. As a results, the bond sodium seems to be logged in the outer region of fuel slug. The main driving force for constituent redistribution appears to be the Zr solubility change along to radial position of the fuel. It is evident that the heat of transport also has some contribution to the redistribution.

I. INTRODUCTION

Recently the potential use of metallic fuel for liquid metal reactors has enhanced owing to the discovery of its merits in safety, high burnup achievement and fabrication by extensive investigation in Argonne National Laboratory(ANL). The Korea Atomic Research Institute(KAERI) has been developing the Korea Advanced Liquid Metal Reactor(KALIMER), and its start-up fuel was potentially chosen as sodium bonded U-10Zr alloys. As one of the KALIMER activities, the metallic fuel performance code MACSIS has been developing to specify design limits and to assure reliable performance of the fuel pin.

The prediction of the fuel temperature distribution is one of the most important factors in a fuel performance code since fuel temperature affects almost all of the fuel element behaviors. It is known that the key factors which may affect the temperature distribution of metallic fuel are porosity formation, bond sodium infiltration into the porosity, and variation of heat generation rate resulted from fuel constituent migration. The fuel temperature calculation scheme was established and implemented into the MACSIS code to incorporate the major temperature controlling factors as mentioned above. This paper presents temperature calculation and constituent redistribution model, and the computational results are compared with experimental data.

II. ANALYSIS

II.1 Fuel Constituent Redistribution

The radial fuel constituent migration related to the formation of three distinct phasal zones is a general phenomenon in the irradiated U-Pu-Zr and U-Zr alloy[1,2]. This phenomenon may affect the in-reactor performance of metallic fuel rods such as melting temperature, thermal conductivity, power generation rate, phase boundaries and porosity distribution of the fuel slug.

Recently several authors[3~5] have attempted to model and explain the phenomenon, solving the diffusion equations governed by chemical potential and thermotransport (Soret effect) effects. The model developed by Hofman et al.[5] predicted that the $\gamma \leftrightarrow (\beta+\gamma)$ and $\gamma \leftrightarrow (\alpha+\gamma)$ phase boundaries occurred at r/R = 0.46, and 0.72, respectively. These values do not correspond closely with measured data which are 0.39 and 0.65, respectively. In this paper, the constituent redistribution of U-10Zr binary alloy was calculated with using more elaborate prediction of the pin temperature. The redistribution model adapted in this work was basically identical with Hofman's model.

According to Shewmon[6] the flux of component 1, J_1, in binary system which is placed in a temperature gradient, can be expressed as,

$$J_1 = -\tilde{D}_1\left(\nabla C_1 + \frac{Q_1^* C_1}{RT^2}\nabla T\right) \quad \text{---------- (1)}$$

where, D_1 is the inter-diffusion coefficient, ∇C_1 is the concentration gradient and Q^*_1 is the heat of transport, R is the universal gas constant and ∇T is the temperature gradient. The equation (1) is used to calculate the Zr flux in the single phase region.

Thermotransport behavior in the two-phase region of several binary alloy systems have been studied to model the dilute interstitial atom migration[7,8], and their theory is adapted in this modeling. According to previous studies of thermotransport, the flux in a two-phase region can be expressed as

$$J_1 = -\tilde{D}_1 C_{S1} V_f\left(\frac{\Delta H_{S1} + Q_1^*}{RT^2}\right)\nabla T \quad \text{---------- (2)}$$

where C_S is the saturation concentration, ΔHs is the partial molar enthalpy of solution, and V_f is the volume fraction of matrix phase which can be determined by the lever rule from the phase diagram. The ΔHs can be determined by the following relations.

$$C_S = C_0 \exp\left(-\frac{\Delta H_S}{RT}\right) \quad \text{---------- (3)}$$

$$\Delta H_S = \frac{RT^2}{C_S}\frac{\partial C_S}{\partial T} \quad \text{---------- (4)}$$

To determine the Zr flux between single and two phase region, Marino's method[9] is applied as follows,

$$J_1 = -\tilde{D}_1\left(\frac{2(C_1 - C_{S1})}{\Delta r} + \frac{Q_1^* C_1}{RT^2}\nabla T\right) \quad \text{---------- (5)}$$

After calculation of the Zr flux from above equations (1), (2) and (5), the Zr concentration change with irradiation time is calculated from continuity equation in cylindrical geometry,

$$C_i^t = C_i^{t-\Delta t} + 2\Delta t\left(\frac{r_i J_i - r_{i+1} J_{i+1}}{r_{i+1}^2 - r_i^2}\right) \quad \text{---------- (6)}$$

where t is time step and i is the annulus node.

A subroutine program has been made and installed into the MACSIS code to simulate constituent

redistribution. This subroutine determines the phase field at each node from the temperature profile and composition concentrations. The polynomial equations have been made to determine the phase field from the U-Zr phase diagram. After then, it evaluates the material properties such as molar enthalpy, volume fraction and diffusion coefficient, and compute the Zr concentration at each node.

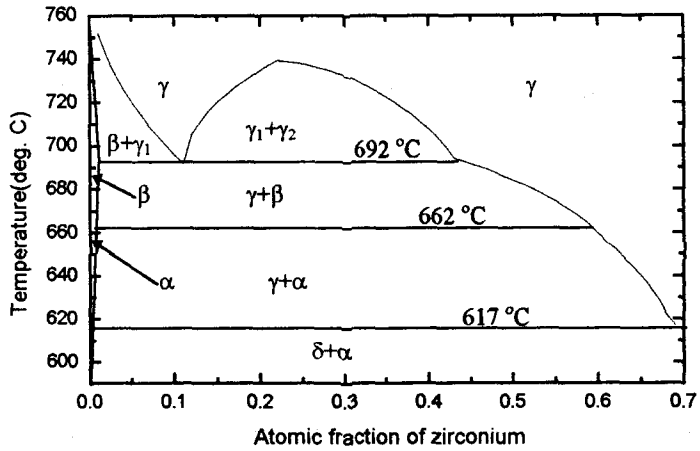

Figure 1. U-Zr phase diagram used in the redistribution calculations.

II.2 Thermal Conductivity Correlation

The thermal conductivity of the unirradiated U-Zr alloy, k_0 (W/mK), can be expressed as a function of temperature and alloy composition[10].

$$k_0 = 17.5\frac{(1-2.23Wz)}{(1+1.61Wz)} + 1.54\times10^{-2}\frac{(1+0.061Wz)}{(1+1.61Wz)}T + 9.38\times10^{-6}T^2 \quad \text{----------(7)}$$

where, T is the temperature in kelvin and Wz is the weight fraction of zirconium.

The porosities and sodium infiltration effect should be considered in calculating the thermal conductivity of irradiated metallic fuel. These effects can be determined by porosity correction factor, P_f, which was derived by Bauer and Holland[11],

$$P_f = \left\{1-3\frac{P_{Na}}{(1-P_g)}\frac{\left(1-\frac{k_{Na}}{k_0}\right)}{\left[\frac{2}{\varepsilon}+\left(3-\frac{2}{\varepsilon}\right)\left(\frac{k_{Na}}{k_0}\right)\right]}\right\}\times(1-P_g)^{3/2} \quad \text{----------(8)}$$

where, k_{Na} is the thermal conductivity of logged sodium, k_0 is unirradiated fuel thermal conductivity, P_{Na} is the sodium-filled porosity fraction, P_g is the gas filled porosity fraction and ε is 1.72.

II.3 Temperature Calculation

The one dimensional steady state heat conduction equation in cylindrical coordinate is described as,

$$rk\frac{\partial T}{\partial r} + \int_0^r rq'''dr = 0 \quad \text{----------(9)}$$

where r is the radial position, T is the temperature, k is the thermal conductivity and q''' is the volumetric heat

generation rate. Equation (9) can be written in finite difference form with assuming parabolic temperature distribution.

$$r_i k_i \frac{2r_i(T_{i+1}-T_i)}{r_{i+1}^2-r_i^2} + \Delta r \sum_{j=1}^{i}(r_j q_j''') = 0 \quad \text{---------- (10)}$$

$$T_{i+1} = T_i + \frac{1}{2k_i}\left[1-\left(\frac{r_{i+1}}{r_i}\right)^2\right]\Delta r \sum_{j=1}^{i}(r_j q_j''') \quad \text{---------- (11)}$$

Equation (11) enables the temperature calculation of fuel pin in which heat generation rate is non-uniform in radially.

III RESULTS AND DISCUSSION

The DP-11 fuel element(U-10Zr) was irradiated at EBR-II by ANL. The temperature profile of this pin could be inferred by measurement of radial phase boundaries[12], and it exhibited significant constituent migration[5]. In this work, the calculation models for fuel temperature profile and redistribution phenomenon were compared with the PIE results of DP-11 element. The major irradiation conditions and PIE results of this pin are summarized in the table 1.

Table 1. Major irradiation conditions and PIE results of the DP-11 fuel pin.

Fuel Length	34.3 cm	Burnup	7.7 at%
Fuel Slug Radius			
Unirradiated	0.217 cm	Linear Power	7.38kW/ft
Irradiated	0.259 cm		
Effective Full Power Days (EFPD)	619	Average Fuel Surface Temperature	622 ℃
Porosity Fraction(EOL)		Phase Boundary Position (r/R)	
Outer zone	0.3	$\gamma \leftrightarrow (\beta+\gamma)$ transition	0.39
Middle zone	0.35	$\gamma \leftrightarrow (\alpha+\gamma)$ transition	0.65
Inner zone	0.31		

III.1 Constituent Redistribution

The diffusion coefficients used in this subroutine was the same with Hofman's model[5] which assumed tenfold increment of all ex-reactor diffusion coefficients due to irradiation effects. However, the diffusion coefficient of ($\alpha+\gamma$) phase region was increased only threefold to simulate closely fit with PIE results.

The driving forces acting on the Zr migration are consist of molar enthalpy of solution ($\triangle Hs$) term due to Zr solubility change with radial temperature profile, heat of transport (Q^*) term due to temperature gradient, and concentration gradient term which is always opposite direction against the ($\triangle Hs+Q^*$) term. As shown in figure 2, significant amount of Zr is depleted in the middle zone in case of no Soret effect assumed, e.g., no heat of transport term acting on the redistribution. However, $\triangle Hs$ term alone does not give the best estimation compared with measured data. Therefore it is assumed the heat of transport (Q^*) as -150 kJ/mole and then, the

calculated Zr concentration is well agreed with experimental data.

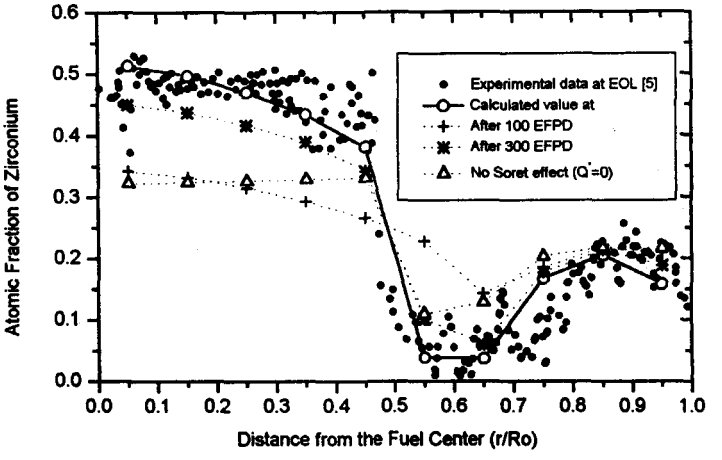

Figure 2. Radial redistribution of zirconium as a function of elapsed time for the pin DP-11.

As shown in figure 2, the Zr concentration after 300 EFPD with Q^* = -150 kJ/mole gives very similar trend with calculated value at EOL (619 EFPD). This indicates that significant amount of Zr is depleted in the middle region and enriched in the central region at the early irradiation

III.2 Temperature Prediction

In the calculation of the fuel temperature distribution, it was assumed that the heat generation rate at each annulus is linearly proportional to the uranium concentration which can be varied with burnup due to the fuel constituent redistribution. A simple correlation was implemented to the MACSIS code to simulate the fuel swelling.

As shown in figure 3, MACSIS code overestimates the fuel temperature when sodium logging effect is not considered. Pahl et al[13] reported that U-10Zr fuel had about 34% of its fission gas bubble volume filled with sodium at 2.9 at% peak burnup. It is not reasonable to assume the logged sodium is homogeneously distributed within the fuel slug. It is assumed that there physically would be some repulsive force against the infiltrating sodium because of the buildup of fission gas pressure and its relaxation by means of the release to the plenum. In addition to this point, it is apparent that all of the porosities up to the innermost zone of the fuel cannot be interconnected each other. The temperature profiles between MACSIS calculation and Yacout's data[12] shows good agreement under assumption that only the outermost ($\alpha+\gamma$) region is logged by bond sodium.

The temperature profile, which is not consider the Zr migration, exhibits a little bit lower estimation than those of measurements. The temperature difference between the calculated with no migration model and the measured are mostly occurred at the ($\beta+\gamma$) phase region. According to the equation (7), the temperature difference will be decreased because of the thermal conductivity is increasing as Zr depleting. On the contrary, the increment of the heat generation rate at the Zr depleted zone is fully compensated for the thermal conductivity decrement and result in the overall temperature increment.

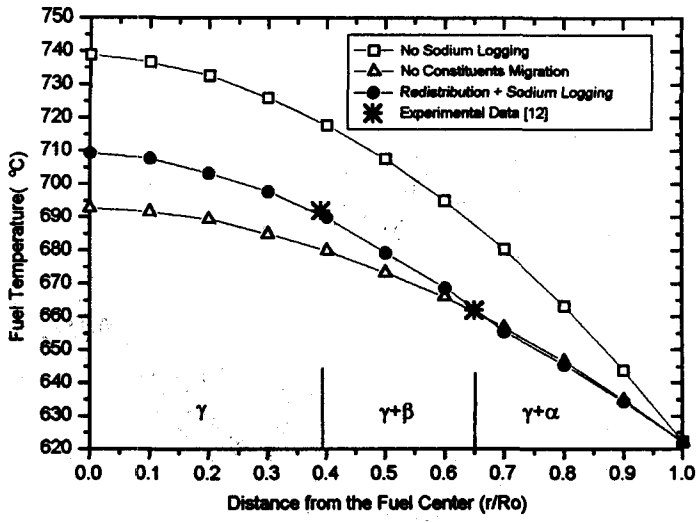

Figure 3. Fuel temperature distributions at EOL of the pin DP-11.

IV CONCLUSIONS

A temperature calculation scheme of metallic fuel slug has been developed, which covers the effects of constituent redistribution and sodium logging. It is apparent that the main driving force for zirconium migration is radial solubility change of zirconium. The heat of transport also plays additional role in the redistribution. The predicted temperatures are overestimated if no sodium infiltration were incorporated, and are underestimated when no constituent migration model is considered. From the computational results, it seems that the bond sodium would not homogeneously distributed but concentrated in the outer region of the fuel slug. The calculated zirconium migration and temperature distribution were generally good agreement with experimental data.

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