Multicomponent Modeling of Radiation-Induced Segregation in Commercial 316 Stainless Steel and Evaluation in Molten Salt Reactor Environments

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1. Introduction

Radiation-induced segregation (RIS) is a phenomenon where the composition of an alloy changes at interfaces or surfaces due to irradiation [1]. Stainless steel, known for its excellent mechanical properties and radiation resistance, is widely used as a internal structural material in pressurized water reactors (PWRs) and is now also being considered for use as vessel material in molten salt reactors (MSRs) [2]. In PWR environments, extensive fundamental research has been conducted due to the potential association of RIS with irradiation-assisted stress corrosion cracking (IASCC). Various rate theory-based or computational models have been developed to quantitatively understand and predict RIS in these environments [3-5]. Compared to PWRs, MSR environments operate at relatively higher temperatures and have different irradiation rates and total irradiation doses. Consequently, there has been limited quantitative analysis of RIS under these conditions.

In this study, we quantitatively evaluate RIS in commercial stainless steels by implementing a multicomponent model that extends the commonly known Fe-Cr-Ni system to the Fe-Cr-Ni-Mo-Si system. The developed model is used to assess and compare RIS behavior in various environments include MSRs.

2. Modeling

The Fe-Cr-Ni-Mo-Si system was employed to simulate radiation-induced segregation (RIS) in SS316. We extended the ternary model implemented by Perks et al. [2] to our quinary system based on Fukuya's research [5]. The model is based on the inverse Kirkendall effect, specifically the vacancy mechanism. Given that Si is a smaller species compared to Fe, we consider the coupling parameter, which reflects the fraction of each solute forming the mixed dumbbell. The differential rate equations for point defects and solutes are fundamentally described below:

$$\frac{\partial C_{v,i}}{\partial t} = -\nabla J_{v,i} + G_{v,i} - R_{v,i} - S_{v,i}$$

$$\frac{\partial C_k}{\partial t} = -\nabla J_k \quad (k = \text{Fe, Cr, Ni, Mo, Si})$$

$$J_v = -D_v \nabla C_v + C_v \sum_k d_k^v \nabla C_k$$

$$J_i = -D_i \nabla C_i - C_i \sum_k d_k^i \nabla C_k$$

$$J_k = -D_k \nabla C_k + C_k \left(d_k^v \nabla C_v - d_k^i \beta_k \nabla C_i \right)$$

$$D_v = \sum_k d_k^v C_k$$

$$D_i = \sum_k d_k^i \beta_k C_k$$

$$D_k = d_k^v C_v + d_k^i \beta_k C_i$$

$$d_k^{v,i} = \nu_{v,i} \omega_{k-v,i} a_0^2 \exp\left(-\frac{E_{k-v,i}^m}{kT} \right)$$

The most critical term is $\mathrm{d}_\mathrm{k}^\mathrm{v}$, which represents the diffusivity correlation factor of atom-defect relationships. As this factor increases, atoms move more easily with vacancies. The coupling parameter (β_k) is described as follows:

$$\beta_k = \frac{\exp\left(\frac{E_{k-i}^b}{kT} \right)}{\sum_k C_k \exp\left(\frac{E_{k-i}^b}{kT} \right)}$$

The detailed equations are provided elsewhere [5]. To account for the grain boundary effect, the grain boundary sink strength S_{gb} was introduced [6]. There were one or two boundaries with S_{gb} at the central region of the grain. Both ends of the system boundary were fixed as deep boundary conditions, which represented an infinitely large grain.

The parameters of the model were derived from the values provided in Fukaya's paper [5] and those from Perks' publication [3]. The calculated composition was convoluted with the X-ray generation profile having a standard deviation of 1 nm to compare with real experimental data.

3. Results

Perks reported various results on the RIS of the Fe-Cr-Ni ternary model [3]. Using the developed model and Perks' parameters, we were able to reproduce the results of Perks. Fig. 1 shows the composition distribution in the Fe-Cr-Ni ternary system under neutron irradiation in

PWR conditions. In this case, the material composition was Fe-20Cr-25Ni, irradiated to 1 dpa at 2e-8 dpa/s and 450°C. As illustrated in the figure, depletion of Cr and enrichment of Ni occurred at the boundary.

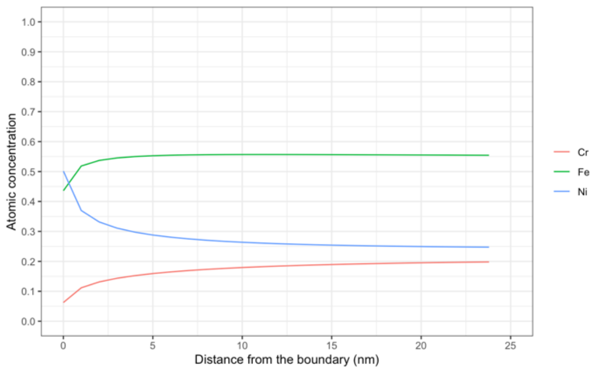

Fig. 1. Calculated atomic concentration profiles of Cr, Fe, and Ni as a function of the distance from the boundary.

The most significant difference between PWR conditions and MSR conditions is the operating temperature. The operating temperature of MSRs is expected to be around 650°C. Over a 20-year operation period, the total dpa of the MSR vessel is approximately 72, so the dpa/s is about 1.14e-7, which is quite similar to the irradiation doses experienced by reactor internals of PWRs. Fig. 2 shows the difference in RIS behavior with temperature under these conditions. As seen in the figure, RIS decreases sharply with increasing temperature. Particularly, Cr concentration, which is a crucial factor determining the overall corrosion characteristics, significantly decreases with higher operating temperatures.

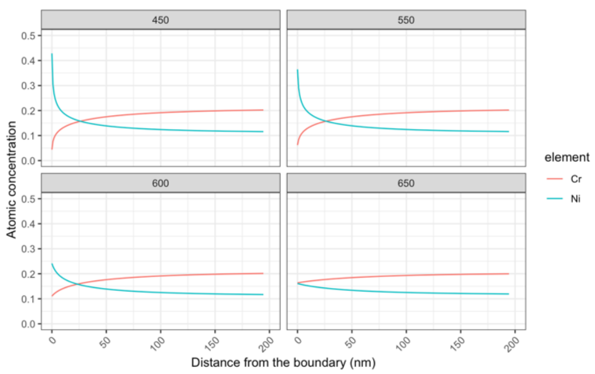

Fig. 2. Calculated atomic concentration profiles of Cr and Ni as a function of the distance from the boundary at different temperatures: 450°C, 550°C, 600°C, and 650°C. The material composition is Fe-20Cr-25Ni. The strips represent the temperatures.

4. Summary

The RIS model for the Fe-Cr-Ni-Mo-Si system was developed to predict grain boundary compositional changes in commercial stainless steel under neutron irradiation. Using the optimized parameters for PWR environments, RIS was evaluated under MSR conditions. The calculated results were compared with values from the literature, and it was specifically observed that RIS significantly decreases compared to PWR conditions as the temperature increases. This modeling can be useful in predicting RIS behavior in SS316 under MSR conditions, and we are considering adapting this model for other candidate materials for MSR vessels.

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