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Simulation of the High Burnup Structure of UO_2 under a Thermal Gradient using the Hybrid Phase-Field Potts Kinetic Monte Carlo Model

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1. Introduction

As nuclear fuel, mainly UO_2, is irradiated in reactors during operation, the microstructure experiences severe irradiation damage. When the fuel exceeds a certain burnup, the damage is increased significantly [1]. This damage is not uniformly distributed along the fuel as the outer side of the fuel experiences significantly higher damage compared to the center of the fuel as a result of excessive fission damage and growth of fission gas bubbles. This phenomenon leads to the creation of a unique structure in the rim region of the fuel that restructures the grains. This region is referred to as the rim structure or the High burnup Structure (HBS) [1-5] region.

The mechanism of the HBS formation during irradiation is not fully understood, due to the lack of in-situ microstructural characterization techniques during the reactor operation. However, the characteristics of the resulting structure are well-known [1, 3, 4]. It has also been observed that the temperature and the local burnup thresholds for the formation of the HBS range between 1000-1200 °C and 60-80 MWD/kgU, respectively [1, 6]. In addition, it is agreed [2, 5, 7, 8] that the fission gas release in UO_2 does not occur in the HBS region and the intergranular bubbles formed by the fission gases in that region is a result of the retention of these gases under the effect of the temperature gradient present in the fuel. Therefore, the pores and bubbles diffusion and movement present in UO_2 during operation under the thermal gradient is of significance.

Computational and numerical modelling has played a significant role in revealing the mechanism of several phenomena that occur in the microstructural level in the material. Kinetic Monte Carlo (kMC) [1, 2, 9, 10] and Phase-Field [11-13] simulations have been separately applied to understand the formation mechanism of the HBS of UO_2. However, in this study, a hybrid model that combines elements of the phase-field model as well as the kMC Potts model is used to simulate the fission gas diffusion under the thermal gradient that the UO_2 nuclear fuel experiences under operation in order to provide an insight on the behaviour of fission gas bubbles in the HBS of UO_2.

2. Model Description

The hybrid phase-field Potts model is developed by Homer et al. [14]. The kinetics are simulated by a combination of Monte Carlo Potts methods and solving the Cahn–Hilliard equation of the phase-field model. To apply this model to the HBS, The grains are assigned to one phase while the pores and bubbles are assigned as a second phase. In addition, the simulations represent the recrystallization phenomena that resemble the evolution of HBS in nuclear fuels. Thus, the HBS is evolved if the system energy of the HBS is lower than that of the original structure in an irradiated UO_2. Therefore, the total system energy, E, is defined as the following equation [1, 2]:

$$E = \sum_{i}^{M}\left[Hf(S_i) + \frac{J}{2}\sum_{j}^{n}\left(1-\delta_{S_iS_j}\right)\right] \quad \dots (1)$$

where J is the boundary energy, δ_{SiSj} is the Kronecker delta function, M the total number of lattice sites, S_i the site, n is the number of neighboring sites and is equal to 8 for 2D simulations, and S_j the nearest neighboring site.

Then, the standard Metropolis algorithm is applied to determine the probability of accepting the event based on the energy calculated in Equation 1. The probability is calculated by the following equation [1, 9, 14]:

$$P = \begin{cases} 1, & \Delta E \leq 0 \\ e^{\left(-\frac{\Delta E}{k_B T}\right)}, & \Delta E > 0 \end{cases} \quad \dots (2)$$

where k_B is the Boltzmann constant and T the simulation temperature.

The relationship between burnup and porosity (P) in a HBS is reported in [1, 2] and it is obtained by a linear fitting from the experimental data in [5] as follows:

$$P(\%) = \begin{cases} 0.06 \cdot Bu, & Bu \leq 60 \\ -6.6 + 0.17 \cdot Bu, & 60 < Bu \leq 100 \\ 4.4 + 0.06 \cdot Bu, & Bu > 100 \end{cases}$$

$$\dots (3)$$

where P(%) is the porosity and Bu is the burnup in MWd/kgU. More details about the model are reported in [1, 2, 14]. It is important to mention that the aim of using this model is to generate the amount of fission gas bubbles that corresponds to certain burnup, as reported in [1, 2].

After modifying the model, a temperature gradient has been applied to the hybrid phase-field Potts model to simulate the grain behaviour and the fission bubbles diffusion in the HBS. This behaviour represents that fission gas bubbles sweep the recrystallized grains. Furthermore, all the simulations have been done using the open-source Stochastic Parallel PARticle Kinetic Simulator (SPPARKS) package developed by Sandia National Laboratories [15].

3. Results

Fig. 1 shows the grains and the fission gas bubbles at a burnup of approximately 95 MWd/kgU without the influence of the temperature gradient. It is shown in Fig. 1 that the grains and the bubbles match the results reported in [1, 2]. However, since a radial temperature gradient that is in the range of 100 °C/mm [16] is present in the UO_2 fuel pellet in the real reactor operation scenario, that thermal gradient is applied in the model to simulate the grains and fission gas bubbles behaviour under the real operation conditions.

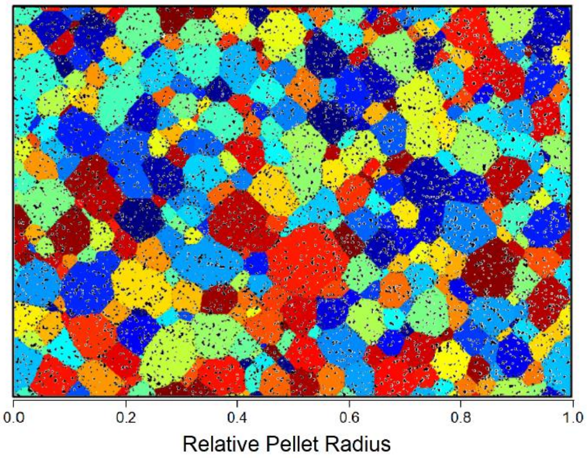

Fig. 1. Grains and fission gas bubbles in irradiated UO_2 without the influence of the radial temperature gradient

In that sense, Fig. 2 shows the effect of the applied temperature gradient on the grain and fission gas bubbles of irradiated UO_2 at the same burnup (95 MWd/kgU). It is clear that the temperature gradient induced grain growth and recrystallization while the fission gas bubbles are accumulated near the edge of the fuel. In addition, the formation of fine grains in the low temperature side is also noted and it matches the experimental observations reported in [3, 17].

However, in the current simulation results, improvements are necessary to be applied to take into account the power peaking that occurs in the rim region of the fuel. This phenomena increases the burnup in the rim region and leads to the production of large amounts of fission gas bubbles. This behavior can be applied in the simulations by implementing a burnup profile instead of a constant value across the whole microstructure. In addition, quantification of the HBS percentage to the overall structure should also be obtained.

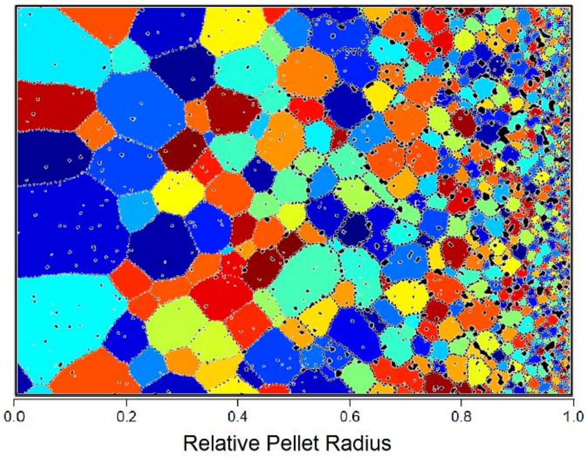

Fig. 2. Grains and fission gas bubbles in irradiated UO_2 under the influence of the radial temperature gradient.

4. Conclusions

A 2D hybrid model that utilizes the phase-field model alongside kMC Potts model has been used to simulate the high burnup structure of irradiated UO_2 at a burnup of approximately 95 MWd/kgU. The results show that the presence of a temperature gradient induces grain growth and recrystallization in addition to fission gas bubbles accumulation near the edge of the fuel. The results of these simulations show good agreement with other simulation results [1, 2] as well as experimental measurements [3, 17]. These simulation methods allows for the prediction of the mechanisms that occur under irradiation and that are complex, if not impossible, to be observed experimentally.

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