

# SIMULATION OF HIGH BURNUP STRUCTURE IN $\text{UO}_2$ USING POTTS MODEL

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The evolution of a high burnup structure (HBS) in a light water reactor (LWR)  $\text{UO}_2$  fuel was simulated using the Potts model. A simulation system for the Potts model was defined as a two-dimensional triangular lattice, for which the stored energy was calculated from both the irradiation damage of the  $\text{UO}_2$  matrix and the formation of a grain boundary in the newly recrystallized small HBS grains. In the simulation, the evolution probability of the HBS is calculated by the system energy difference between before and after the Monte Carlo simulation step. The simulated local threshold burnup for the HBS formation was 62 MWd/kgU, consistent with the observed threshold burnup range of 60-80 MWd/kgU. The simulation revealed that the HBS was heterogeneously nucleated on the intergranular bubbles in the proximity of the threshold burnup and then additionally on the intragranular bubbles for a burnup above 86 MWd/kgU. In addition, the simulation carried out under a condition of no bubbles indicated that the bubbles played an important role in lowering the threshold burnup for the HBS formation, thereby enabling the HBS to be observed in the burnup range of conventional high burnup fuels.

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**KEYWORDS :** High Burnup Structure, HBS, Rim Structure,  $\text{UO}_2$ , Nuclear Fuel, Potts Model, Monte Carlo Simulation

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## 1. INTRODUCTION

When a nuclear fuel burnup exceeds a certain burnup, a high burnup structure (HBS), also referred to as a rim structure, is formed at a  $\text{UO}_2$  pellet periphery as a result of excessive fission damage and growth of fission gas bubbles [1]. Although its formation mechanism is not fully understood, its characteristics are well recognized: The HBS microstructure consists of very small sub-mm sized grains and large  $1\mu\text{m}$  sized bubbles instead of the original  $10\mu\text{m}$  sized  $\text{UO}_2$  grains [2,3]. The threshold burnup for a HBS formation has been observed at a local burnup between 60-80 MWd/kgU [1,4,5], and a threshold temperature between 1000-1200°C [6].

Several models have been proposed for investigating the HBS characteristics and formation mechanism [7-9]. These complex mathematical models were based on the assumption that the HBS formation began on or around the grain boundaries. Since they assumed that gas precipitations and grain subdivision propagations occurred simultaneously, they did not consider the effect of the bubbles on the HBS evolution. However, it was found that the gas precipitation and the grain subdivision propagation are two distinct phenomena [10].

Since the recrystallization phenomena in materials

resemble the HBS evolution in  $\text{UO}_2$  fuel from the aspect of forming new small defect-free grains, some HBS models adopted a basic concept that the driving force for the HBS evolution is the stored energy in the  $\text{UO}_2$  grains due to an irradiation [8,11]. In this paper, the Potts model, based on the Monte Carlo method, was implemented for simulating the HBS evolution instead of an analytical solution. Although it was originally developed for magnetic domain evolutions, it has also been applied to grain growth in polycrystals in that there are similarities in the structures between the Potts domains and grains [12]. It also has been implemented to model various microstructural phenomena, such as grain growths [13,14], recrystallization [15,16], and sintering [17]. Here, we adopt the Potts model for the HBS evolution from simulating a recrystallization phenomenon [16]. The Potts model in this paper does not consider a detailed HBS evolution mechanism, but only considers a total system energy change due to the formation of new small defect-free grains. The HBS may be evolved if the system energy of the HBS is lower than that of the original structure in an irradiated  $\text{UO}_2$ . The advantage of the Potts model over an analytical solution is that it can generate snapshots of the evolving microstructure with time [12], which improves the understanding of the HBS evolution phenomena.

## 2. DESCRIPTION OF THE POTTS MODEL FOR A HBS FORMATION

In this paper, the Potts model defines a microstructure as a two-dimensional triangular lattice system with a periodic boundary condition. A site in the system has six nearest neighboring sites as shown in Fig. 1. The system has 1024 × 1024-sized triangular lattice sites and the distance between sites is equal to the size of a site, which is a basic element in the Potts model. As the Potts model cannot treat an element smaller than a site, the smallest element described in the HBS simulation should be evaluated. Since a nano-bubble in UO<sub>2</sub> is much smaller than a grain, we consider that a nano-bubble is the smallest element in the HBS simulation and consequently assume that the distance between sites is 40 nm, which is the size of the stabilized coalescent nano-bubbles range [10].

Each site has an integer value,  $S_i$ , representing a site state and a grain orientation.  $S_i = 0$  means that the site is a bubble. If a site is an original UO<sub>2</sub> grain structure, it has a  $S_i$  between 1, 2, ..., Q. When a site is a HBS grain, it has a  $S_i$  between Q+1, Q+2, ..., 2Q. We choose Q=100 which is large enough to guarantee a Q independence for the simulation results [13]. We assume that the neighboring sites with the same state are the same grain, and the perimeters of the sites with different states are grain boundaries as shown in Fig. 1.

The HBS evolution makes new small dislocation-free (or reduced dislocation density) HBS grains [18], which results in an increased grain boundary area and decreased stored energy. Therefore, the total system energy for the Potts model,  $G$ , should take this energy change into account as

$$G = \sum_i^M \left[ Hf(S_i) + \frac{J}{2} \sum_j^{nm} (1 - \delta_{S_i S_j}) \right] \quad (1)$$

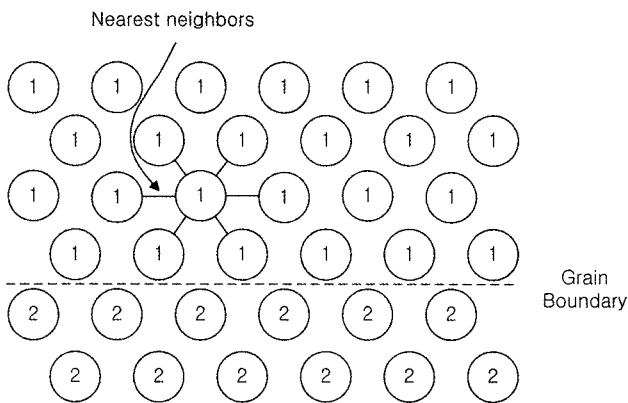


Fig. 1. Schematic Diagram of a Two-dimensional Triangular Lattice in the Potts Model

where  $J$  is a constant related to a boundary energy,  $\delta_{S_i S_j}$  the Kronecker delta function,  $M$  the total number of lattice sites,  $S_i$  the chosen site, and  $S_j$  the nearest neighboring site [15]. For a two-dimensional triangular lattice,  $nm$  is 6 and  $H$  is the stored energy accumulated per site due to irradiation. The function  $f(S_i)$  has a value of one for the original grain sites and zero for the HBS grain sites.

The HBS evolution is simulated as follows: For the grain step, a grain site is chosen and a new state at that site is assigned randomly. The energy change,  $\Delta G$ , is calculated by equation (1), and then the state change probability,  $W$ , is calculated by

$$W = \begin{cases} \exp\left(-\frac{\Delta G}{k_B T}\right) & \text{for } \Delta G > 0 \\ 1 & \text{for } \Delta G \leq 0 \end{cases} \quad (2)$$

where  $k_B$  is the Boltzmann constant and  $T$  the simulation temperature (K) [14]. In this paper, the temperature is fixed at 923 K, which is a typical temperature for the HBS region in UO<sub>2</sub> fuel. For the simulation of a bubble migration, a bubble site and a neighboring site which is not a bubble are chosen randomly and exchanged. Then, the neighboring site's state is determined by randomly selecting a neighbor's state at a new site and the state change probability is calculated. The total number of bubble sites and grain sites remains constant during the simulation of a bubble migration.

The time unit is represented as a Monte Carlo Step (MCS). One MCS corresponds to  $N$  re-orientation attempts, where  $N$  is the total number of sites in the system [13]. We assume that the time for 1 MWd/kgU burnup corresponds to 100 MCS. To simulate faster bubble mobility than grain boundary mobility, more attempts for a bubble migration should be carried out than for a grain step. We assume that 10 bubble migration steps are attempted for each grain step.

The initial microstructure for this study is also obtained

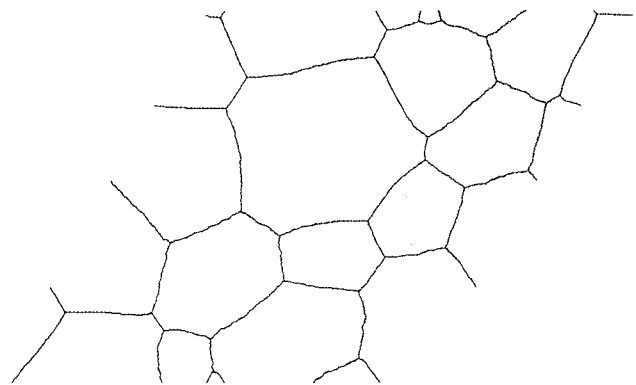


Fig. 2. Original Microstructure for the Potts Model

by the Potts model described above. We assign a random number between 1 and 100 for each site, and then simulate a grain growth. Fig. 2 shows the initial microstructure with an average of 9.62  $\mu\text{m}$  grains, which is a typical grain size for a PWR nuclear fuel. The sizes of grains and bubbles are calculated by converting them into an equivalent area of a circle and statistically averaged sizes are calculated.

### 3. PARAMETERS FOR THE POTTS MODEL

The parameters for equations (1) and (2) can be obtained as follows:  $J$  depends on the boundary types as

$$J = \begin{cases} \gamma_{GB}S & \text{for grain boundary} \\ \gamma_s S & \text{for pore surface} \end{cases} \quad (3)$$

where  $\gamma_{GB}$  is the grain boundary energy,  $S$  is the contact length between sites, and  $\gamma_s$  is the surface energy ( $\text{J/m}^2$ ) [19]. The grain boundary energy,  $\gamma_{GB}$  [9], is

$$\gamma_{GB} = \sqrt{2}\gamma_s b_V \sqrt{\rho_N} \quad (4)$$

where  $b_V$  is the magnitude of the Burgers vector [9] and  $\rho_N$  the dislocation density ( $\text{m}^{-2}$ ) [19],

$$\gamma_s = 0.85 - 1.4 \times 10^{-4} T \quad (5)$$

$$\rho_N = \begin{cases} 10^{0.022 Bu + 13.8} & \text{for } Bu \leq 44 \\ 5.86 \times 10^{14} & \text{for } Bu > 44 \end{cases} \quad (6)$$

where  $Bu$  is the local burnup ( $\text{MWd/kgU}$ ).

If the stored energy due to an irradiation is assumed to be equal to the strain energy, the stored energy per site is

$$H = \Delta U \cdot A \quad (7)$$

where  $\Delta U$  is the strain energy, and  $A$  is the site area. The strain energy,  $\Delta U$  [9], is

$$\Delta U = \frac{1}{2} \left( \frac{\Delta a}{a} \right) E \quad (8)$$

where  $\left( \frac{\Delta a}{a} \right)$  is the lattice parameter expansion [20], and  $E$  the elastic modulus of  $\text{UO}_2$  ( $\text{N/m}^2$ ) [9] as follows:

$$\left( \frac{\Delta a}{a} \right) = 1.57 \times 10^{-5} \cdot Bu \quad (9)$$

$$E = 2 \times 10^{11} (1 - 1.09154 \times 10^{-4} T) \quad (10)$$

The relationship between burnup and porosity ( $P$ ) in a HBS can be obtained by a linear fitting from the experimental data [21] as follows:

$$P(\%) = \begin{cases} 0.06 \cdot Bu & \text{for } Bu \leq 60 \\ -6.6 + 0.17 \cdot Bu & \text{for } 60 < Bu \leq 100 \\ 4.4 + 0.06 \cdot Bu & \text{for } Bu > 100 \end{cases} \quad (11)$$

After a difference between the porosity in the simulation system and the increased porosity in equation (11) with the burnup is evaluated every 200 MCS, the number of bubbles corresponding to the porosity difference is introduced into the system. The bubble introduction is carried out by randomly choosing a grain site and changing it into a bubble site.

## 4. RESULTS AND DISCUSSION

### 4.1 Microstructure and Threshold Burnup for a HBS Formation

Fig. 3 shows the evolution of the simulated HBS fraction, which is defined as an area ratio of the HBS grains to the total grains. If the threshold burnup for the HBS formation is defined as the burnup where the slope of the HBS fraction changes abruptly, the threshold burnup is estimated to be 62  $\text{MWd/kgU}$ . It is consistent with the observed results, 60-80  $\text{MWd/kgU}$  [3-5]. The slope of the HBS fraction increases abruptly again at 86  $\text{MWd/kgU}$ . The reasons why the threshold burnup and a second change of the HBS fraction occur were investigated by analyzing snapshots of the evolving microstructures.

Fig. 4 demonstrates the simulated microstructure at 80  $\text{MWd/kgU}$ , which has a 9.4% HBS fraction. As the grain boundaries are too thin to be shown in a reduced scale system figure, clusters of small HBS grains are shown as dispersed grey parts in Fig. 4. The black spots in the figure are bubbles. In Fig. 4, the intergranular bubbles are formed by migrations into the grain boundaries and coalescences of the intragranular bubbles located around the grain boundaries, whereas the intragranular bubbles coalesce with adjacent

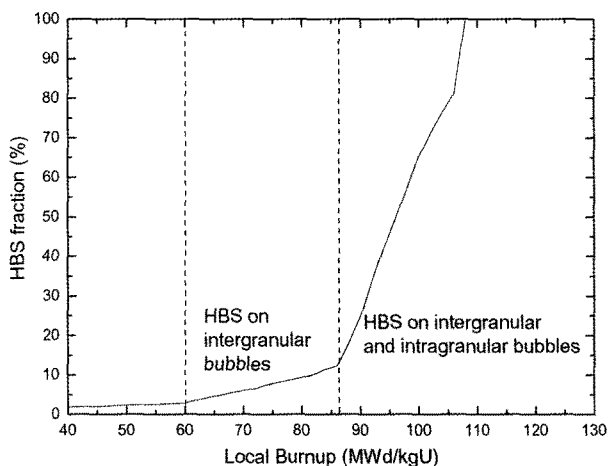


Fig. 3. Evolution of the HBS Fraction with Local Burnup

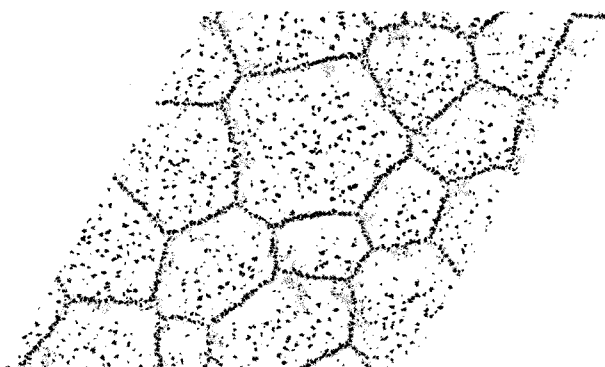


Fig. 4. Simulated Microstructure at a Local Burnup of 80 MWd/kgU

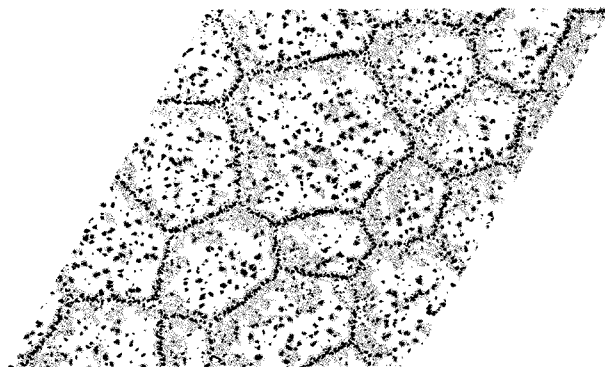


Fig. 5. Simulated Microstructure at a Local Burnup of 94 MWd/kgU

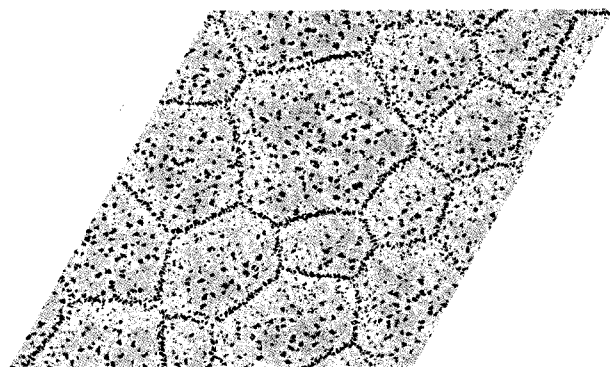


Fig. 6. Simulated Fully-developed HBS Microstructure at a Local Burnup of 110 MWd/kgU

small bubbles. The bubble growth, however, is restricted due to its low mobility at a low temperature. The original UO<sub>2</sub> grains mostly remain unchanged and small HBS grains are nucleated heterogeneously on the intergranular bubbles and begin to develop into the inside of the grains. The Potts model accepts a site change on the condition that the system energy after a site status change is lower than that of a previous status. It means that high energy interfaces, i.e., a grain boundary and a bubble surface, are favorable sites for a HBS formation. The bubble surface has a higher energy than a grain boundary [19], and consequently is the most favorable site for a HBS formation. Since the stored energy of the original UO<sub>2</sub> grains for the burnup range from 62 to 86 MWd/kgU is not sufficiently high for a HBS nucleation only at the bubble surface, the contribution of other sites with a high energy, i.e., a grain boundary, is essential for the HBS formation. Therefore, small HBS grains are nucleated heterogeneously on the intergranular

bubbles at this burnup range. The role of the bubble surface in the new small grain formation can be indirectly confirmed by the observation of the formation of round subgrains near the pores [22].

Fig. 5 shows the simulated microstructure at 94 MWd/kgU, whose HBS fraction reaches 42%. It reveals that the small HBS grains are nucleated not only on the intergranular bubbles but also on the intragranular bubbles because the original UO<sub>2</sub> grains accumulate sufficient stored energy over a burnup of 86 MWd/kgU. This addition to the HBS nucleation mechanism makes the HBS evolution faster, and it also explains the steep slope change of the HBS fraction at a burnup of 86 MWd/kgU in Fig. 3.

Fig. 6 shows the fully developed HBS microstructure at 110 MWd/kgU. The original UO<sub>2</sub> grain structure does not exist any longer but the bubbles located at the original grain boundaries maintain their position. The average HBS grain size is 0.18 μm, which is a little smaller than the experimental data [1,10]. However, the average bubble size is 0.28 μm, which is much smaller than the about 1 μm

bubbles in the experimental data [1]. This discrepancy could result from an incomplete simulation process: The migration and coalescence behaviors of the bubbles were simulated in this paper but a bubble growth due to internal gas pressurization [10] was not considered because of its difficult techniques for programming. This bubble growth will be simulated in a further study.

#### 4.2 No Bubble Condition

One of the advantages of a computer simulation is the ability to observe a behavior under a complex condition. To more clearly estimate the effects of the bubbles on the HBS formation, we excluded bubbles from the simulation and observed a role of only grain boundaries in the HBS formation. Fig. 7 shows the simulation results under the condition that no bubbles existed in the system. Although

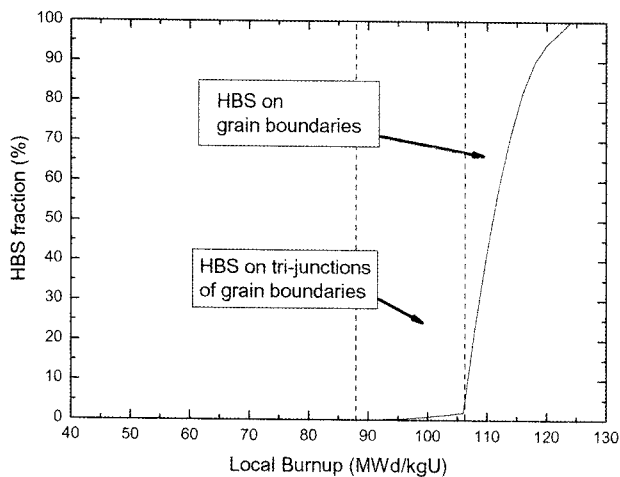


Fig. 7. Evolution of the HBS Fraction with a Local Burnup under a No Bubbles Condition

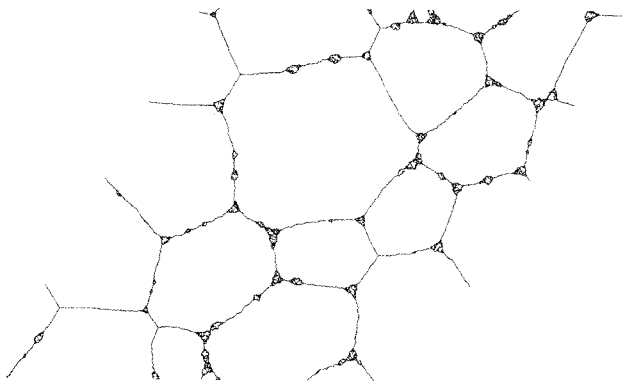


Fig. 8. Simulated Microstructure at a Local Burnup of 106 MWd/kgU under a No Bubbles Condition

the HBS begins to form at 88 MWd/kgU, the HBS fraction is negligible. Since this HBS fraction is too low to be observed by the experimental methods, it is not reasonable that 88 MWd/kgU is defined as the local threshold burnup for the HBS formation. Thus we defined 106 MWd/kgU as the local threshold burnup for forming a sufficient HBS where the HBS fraction increased sharply.

Fig. 8 demonstrates the simulated microstructure at 106 MWd/kgU. In the burnup from 88 to 106 MWd/kgU, the HBS is mostly evolved at the tri-junctions of the grain boundaries. If intergranular bubbles existed in the system, they would be the most favorable sites for HBS formation as mentioned in section 4.1. The most favorable site for HBS formation under a no bubbles condition, however, becomes the tri-junctions of the grain boundaries due to their relatively high energy, which is lower than the bubble surface energy. The HBS fraction for this case, however, is negligible because the fraction of the tri-junctions of the grain boundaries is very low in the resultant microstructure. As the burnup increases to over 106 MWd/kgU, the amount of stored energy increases sufficiently for a HBS to evolve on all the normal grain boundaries as shown in Fig. 9. Since the HBS grain boundaries also act as a nucleation site, the original UO<sub>2</sub> grains are rapidly transformed into a HBS structure.

The simulation under a condition of no bubbles reveals that a HBS can be evolved without bubbles, but that bubbles play an important role in lowering the threshold burnup for the HBS formation and enabling the HBS to be observed in the burnup range of conventional high burnup fuels.

The threshold burnup values derived in this paper slightly fluctuated depending on the adopted assumptions and the system. If another system or different assumptions were implemented for the simulation instead of the two-dimensional triangular system in this paper, the general trend of the results would be maintained but the threshold burnup values would be shifted a little due to a change in the obtained system energy.

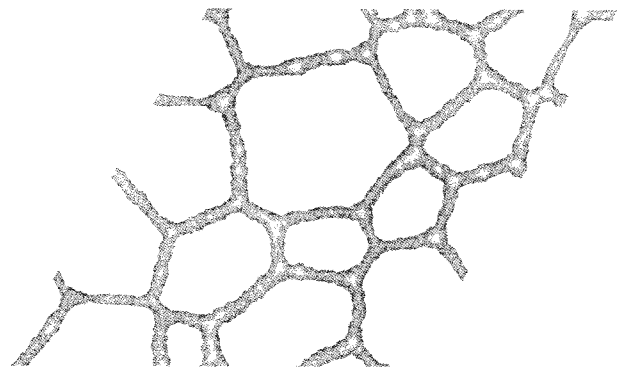


Fig. 9. Simulated Microstructure at a Local Burnup of 108 MWd/kgU under a No Bubbles Condition

## 5. CONCLUSIONS

A two-dimensional Potts model, based on system energy stability, was implemented for simulating the HBS formation. The simulated threshold burnup for the HBS formation was 62 MWd/kgU, which lies within the range of experimental observations. The simulation revealed that the HBS was heterogeneously nucleated on the intergranular bubbles in the proximity of the threshold burnup and then additionally on the intragranular bubbles for a burnup above 86 MWd/kgU. The simulation results under a condition of no bubbles indicated that a HBS can be evolved without bubbles but that the bubbles played an important role in lowering the threshold burnup for a HBS, thereby enabling a HBS to be observed in the burnup range of conventional high burnup fuels.

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