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Kinetic Monte Carlo Potts Model for Simulating a High Burnup Structure in UO_2

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1. Introduction

A Potts model, based on the kinetic Monte Carlo method, was originally developed for magnetic domain evolutions, but it was also proposed as a model for a grain growth in polycrystals due to similarities between Potts domain structures and grain structures [1]. It has modeled various microstructural phenomena such as grain growths [2,3], a recrystallization [4,5], a sintering [6], and so on.

A high burnup structure (HBS) is observed in the periphery of a high burnup UO_2 fuel. Although its formation mechanism is not clearly understood yet, its characteristics are well recognized: The HBS microstructure consists of very small grains and large bubbles instead of original as-sintered grains [7,8]. A threshold burnup for the HBS is observed at a local burnup 60-80 Gwd/tM [9,10], and the threshold temperature is 1000-1200^{o}C [11].

Concerning a energy stability, the HBS can be created if the system energy of the HBS is lower than that of the original structure in an irradiated UO_2. In this paper, a Potts model was implemented for simulating the HBS by calculating system energies, and the simulation results were compared with the HBS characteristics mentioned above.

2. Potts model

The Potts model in this paper defines a microstructure as two-dimensional triangular lattices with a periodic boundary condition. A distance between sites was assumed to be 40 nm. Fig.1. shows an as-sintered microstructure for a Potts model, whose average grain size is 7.4 um.

Each site at a lattice has a state S_i, representing a site state and a grain orientation. Si = 0 means that the site is a pore. If a site has a Si between = 1, 2, …, Q, the site is an original as-sintered structure. If a site has a Si between = Q+1, Q+2, …, 2Q, the site is a HBS. We chose Q=100 which was large enough to guarantee a Q independence for the simulation results [2]. We assumed that the neighboring sites with the same state were the same grain, and the perimeters of the sites with different states were a grain boundary.

The total energy of the system , G, was calculated by

$$G = \sum_{i}^{M}\left[Hf(S_i) + \frac{J}{2}\sum_{j}^{nm}(1-\delta_{S_iS_j}) \right] \qquad (1)$$

where J was a constant related to a grain boundary energy, $\delta_{S_iS_j}$ the Kronecker delta function, S_i the chosen site, and S_j the nearest neighboring site [4]. For the two-dimensional triangular lattice, nm is 6. H is the stored energy per site due to an irradiation. The function $f(S)$ is one for original as-sintered sites and zero for HBS sites.

The HBS was simulated as follows: For the grain growth step, a grain site was chosen and a new state at that site was randomly chosen. The energy change, ΔG, was used for the state change probability, W, which was determined from

$$W = \begin{cases} \exp(-\dfrac{\Delta G}{k_B T}) & for \quad \Delta E > 0 \\ 1 \quad for & \Delta E \le 0 \end{cases} \qquad (2)$$

where k_B is the Boltzmann constant, T the simulation temperature [3]. For the pore migration step, a pore site and a neighbor site, which was not a pore, were randomly chosen and were exchanged. The neighbor site's state was determined from randomly selecting a neighbor's state at a new site. Then, the state change probability was calculated.

The time unit was represented as a Monte Carlo step (MCS). 1 MCS corresponds to N re-orientation attempts, where N is the total number of sites in a system [2]. For the pore sites we attempted 5 pore migration steps for each grain growth step.

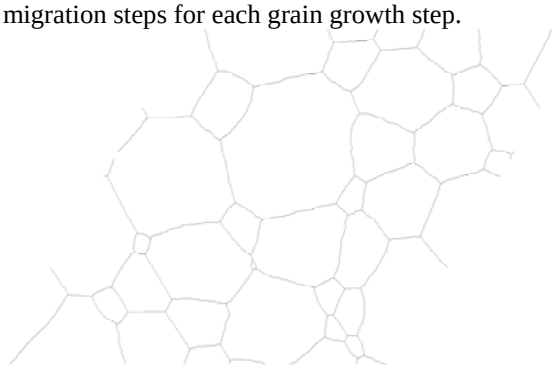

Fig. 1. As-sintered microstructure for a Potts model

3. Parameter for Simulation

Parameters for eq. (1) and (2) can be obtained as follows. If the distance between sites is 40 nm, the contact length, S, is 23nm. Then J is

$$J = \gamma_{GB} S \qquad (3)$$

where γ_{GB} is the grain boundary energy and S is the contact length between sites. The each parameter was obtained from literatures [12,13,14].

The stored energy per site due to an irradiation is

$$H = \Delta U \cdot A \qquad (4)$$

where ΔU is a strain energy, and A is a site area. The strain energy, ΔU is [12]

$$\Delta U = \frac{1}{2}\left(\frac{\Delta a}{a}\right)E \qquad (5)$$

where $\left(\frac{\Delta a}{a}\right)$ is the lattice parameter expansion [15], and E the elastic modulus of the material [12]. The porosity in the HBS was obtained in the literature [16].

4. Results and Discussions

Fig.2. shows the simulated HBS fraction at 923K. If the burnup threshold for HBS is defined as the burnup at which the HBS fraction sharply increases, the local burnup threshold for HBS is 60 GWd/tM. It agreed well with the observed results [9,10]. Due to rough assumptions and estimations of the parameters, the HBS fraction is not zero below the threshold burnup.

The simulation shows that the formation mechanism of the HBS is a heterogeneous nucleation at the grain boundaries. The combination of grain boundaries and the pores near them induces a high energy state, which makes the HBS formation easy.

Fig.3 shows the simulated HBS microstructure at 100GWd/tM and 923K. The average pore size was 0.206 um and the average grain size was 0.120 um. The average pore size is smaller than expected, but some large pores near 1 um size exist.

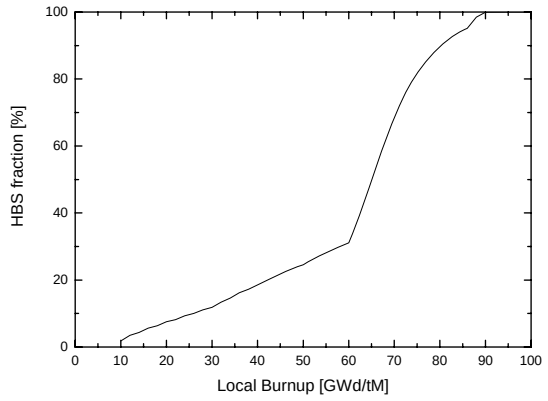

Fig. 2. HBS fraction as function of local burnup at 923K

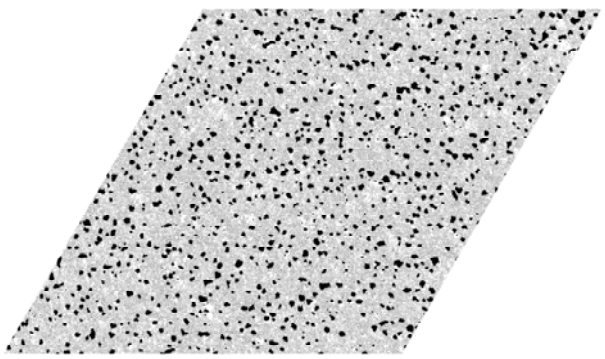

Fig. 3. Simulated HBS microstructure at 100GWd/tM and 923K

5. Conclusion

A two-dimensional Potts model, based on the system energy stability, was implemented for simulating the HBS. The simulated burnup threshold was 60 GWd/tM, which agreed well with the observed results. The simulation of the microstructure showed that the formation mechanism of the HBS is a heterogeneous nucleation at the grain boundaries and pores.

Acknowledgments

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REFERENCES

[1] E.A. Holm and C.C. Battaile, JOM, Vol. 53, No. 9, pp. 20-23, 2007.
[2] M.P. Anderson, D.J. Srolovitz, G.S. Grest, and P.S. Sahni, Acta metal., Vol. 32, No. 5, pp. 783-791, 1984.
[3] V. Tikare and E.A. Holm, J. Am. Ceram. Soc. Vol. 83, No. 3, pp. 480-484, 1998.
[4] D.J. Srolovitz, G.S. Grest, M.P. Anderson, and A.D. Rollett, Acta metal., Vol. 36, No. 8, pp. 2115-2128, 1988.
[5] A.D. Rollett, D.J. Srolovitz, M.P. Anderson, and R.D. Doherty, Acta metal., Vol. 40, No. 12, pp. 3475-3495, 1992.
[6] M.R. Dudek, J.-F. Gouyet, and M. Kolb, Surface Science, Vol. 401, pp. 220-226, 1998.
[7] J. Spino, K. Vennix, and M. Coquerelle, J. Nucl. Mater., Vol. 231, pp. 179-190, 1996.
[8] Y.H. Koo, J.Y. Oh, B.H. Lee, J.S. Cheon, H.K. Joo, and D.S. Sohn, Journal of Korean Nuclear Society, Vol. 35, No. 1, pp. 55-63, 2003.
[9] M.E. Cunningham, M.D. Freshley and D.D. Lanning, J. Nucl. Mater., Vol. 188, pp. 19-27, 1992.
[10] C.T. Walker, T. Kameyama, S. Kitajima, and M. Kinoshita, J. Nucl. Mater., Vol. 188, pp. 73-79 ,1992
[11] T. Sonoda, M. Kinoshita, I. L. F. Ray, T. Wiss, H. Thiele, D. Pellottiero, V. V. Rondinella, and Hj. Matzke, Nucl. Inst. And Meth. in Phys. Res. B,, Vol. 191, pp. 622-628, 2002.
[12] J.Rest, J. Nucl. Mater., Vol. 349, pp.150-159, 2006.
[13] R.O.A. Hall, M.J. Mortimer, and D.A. Mortimer, J. Nucl. Mater., Vol. 148, pp.237-256, 1987.
[14] K. Nogita and K. Une, Nucl. Inst. And Meth. in Phys. Res. B, Vol. 91, pp 301-306, 1994.
[15] J. Spino, and D. Papaioannou, J. Nucl. Mater., Vol. 281, pp. 146-162, 2000.
[16] J. Spino, A.D. Stalios, H. Santa Cruz, and D. Baron, J. Nucl. Mater., Vol. 354, pp. 66-84, 2006.