

Computer Simulation of Sintering and Grain Growth

Hideaki Matsubara

*Fine Ceramics Research Association Japan Fine Ceramics Center,
2-4-1 Mutsumo Atsuta-ku Nagoya, 456, Japan*

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Abstract This paper is aimed to study the computer simulation of sintering process for ceramics by Monte Carlo and molecular dynamics methods. Plural mechanisms of mass transfer were designed in the MC simulation of sintering process for micron size particles; the transfer of pore lattices for shrinkage and the transfer of solid lattices for grain growth ran in the calculation arrays. The MD simulation was performed in the case of nano size particles of ionic ceramics and showed the characteristic features in sintering process at atomic levels. The MC and MD simulations for sintering process are useful for microstructural design for ceramics.

1. Introduction

Many studies on sintering have been conducted by theoretical or experimental techniques.^{1,2)} The classical theories had treated the simplified behaviors of sintering, so that there was a big gap between the theories and the experiments. The studies using the modern techniques such as mapping and computer simulation can give us not only basic information of sintering but also more realistic understanding of sintering to narrow the gap.³⁻⁵⁾ Especially, the computational techniques could be a promising method to provide the future and important directions for the material design; for example, designing new fabrication processes, complicated microstructures, synergistic effects, etc.

The aim of this paper is to study the computer simulation of sintering process for ceramics by Monte Carlo and molecular dynamics methods and then discuss the possibilities of the simulations for microstructure design in ceramics.

2. Simulation method

2.1. MC simulation

The Monte Carlo (MC) simulation for grain growth^{6,7)}

is a probability type method in which sequences proceed to the direction of decreasing the total grain boundary energy. The MC simulation can be applied into other processes of microstructural development in ceramics such as grain growth through liquid (Ostwald ripening), sintering of solid state and sintering under the existence of liquid (liquid phase sintering).⁸⁻¹⁰⁾

The array of MC simulation in this study is the two dimensional and triangular lattices which consist of a main solid phase with different crystal orientations, a second solid phase and a pore (or void) phase. There are different energy values depending on grain boundaries, interfaces and surfaces. When a solid lattice is selected in the array, it tries the mass transfer through grain boundaries. When a pore lattice is selected, the pore lattice moves due to the four diffusion routes; volume, grain boundary, pore-surface and outside-surface routes. If the total energy change ≤ 0 , this try is in practice.

2.2. MD simulation

The software of MASPHYC (Fujitsu Co.) is used to perform the molecular dynamics (MD) simulations in the three dimensional array which consist of the total number of 4000 atoms with the cyclic boun-

dary condition. Cubic Zirconia-Yttria is the target material for MD simulation of grain boundary and surface structures. The simulations of grain boundary and surface were carried out under the ensembles of NTP and NTV, respectively. The formations of grain boundary and surface were simulated as a function of starting structure, temperature and time to examine the structural changes and to calculate the energy change in the formations.

3. Results and discussion

3.1. MC simulation

Fig. 1 shows MC simulation results for grain growth at solid state. The simulation of (a) is the most simple case single phase polycrystalline with random configuration and size distributions. The result of (b) shows the grain growth in the microstructure involving randomly dispersed second par-

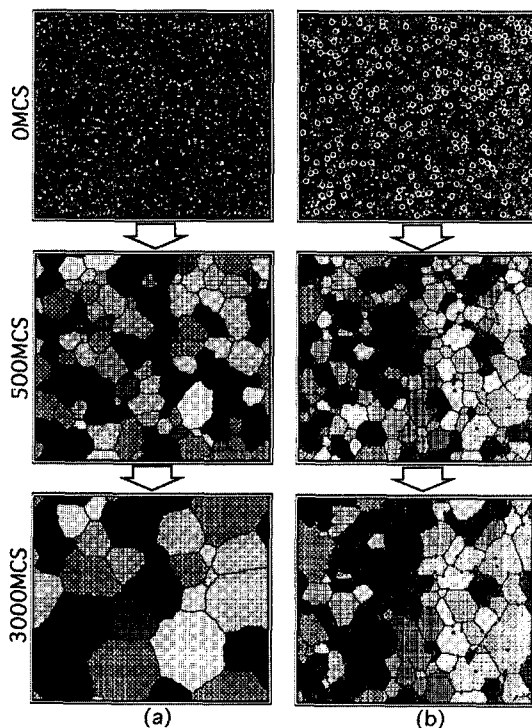


Fig. 1. Grain growth simulations at solid state by MC method. (a) single phase polycrystalline, (b) involving randomly dispersed second particles.

ticles which suppress the grain growth of matrix phase by pinning effect.

The three cases of modified simulation for grain growth under the presence of a liquid phase are shown in Fig. 2. These results are simulated commonly at a constant fraction of a liquid phase, 20%, to examine the influence of the interfacial energy between solid and liquid (γ_{SL}). The simulation can indicate the two different mass transfer mechanisms; the movement of solid boundary (grain growth at solid state) and that of solid/liquid interface (solution/precipitation through a liquid phase, Ostwald ripening). The result of (a) is the case of high γ_{SL} which demonstrates the dominant mechanism of solid state growth in the microstructure with isolated liquid phase. The result of (b) is the simulation with low γ_{SL} indicating the activated movement of solid/liquid interface and thin film formation of liquid phase. The case of (c) shows the effect of anisotropy of γ_{SL} .

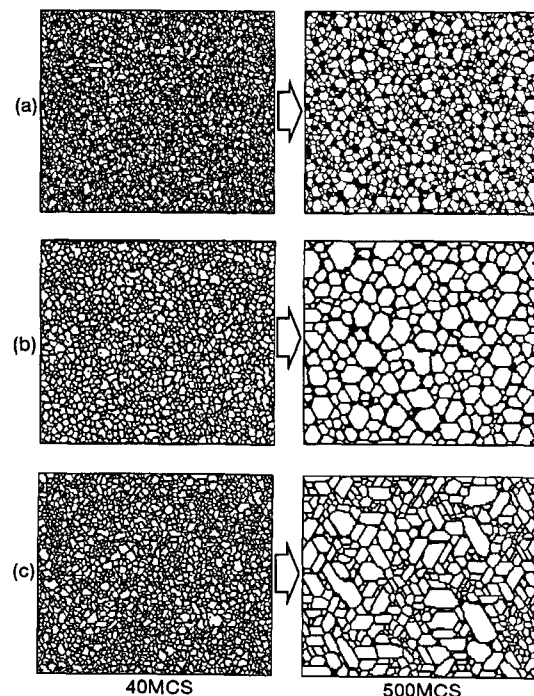


Fig. 2. MC simulations of grain growth under the presence of a liquid phase affected by the interface energy between solid and liquid (γ_{sl}). (a) high γ_{sl} , (b) low γ_{sl} , (c) anisotropic γ_{sl} .

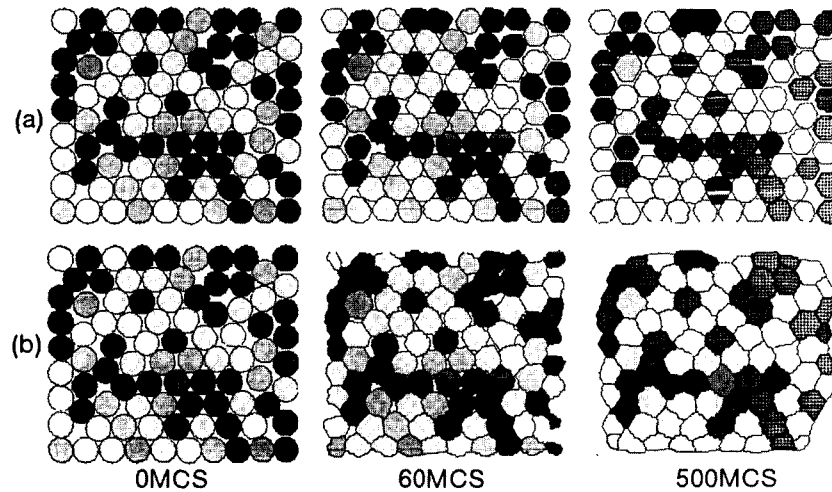


Fig. 3. Exmample of sintering simulation of solid particles. γ_{sv}/γ_{ss} ratio is 2 and 0.5 in (a) and (b), respectively.

by which directional growth behavior or unique microstructure with texture can be designed in such systems as silicon nitride base ceramics.

Fig. 3 shows a couple of MC simulation results for sintering of solid particles as a function of calculation steps (MCS). Both cases of (a) and (b) have commonly the initial configuration of circle particles with the same diameter. The two cases have different conditions of energy ratio. The ratio of surface energy (γ_{sv}) to grain boundary energy (γ_{ss}) is 2 and 0.5 in (a) and (b), respectively. The simulation of (a) shows a feature of non-shrinkage

in a compact because the shrinkage never bring an energy decrease in this condition. The simulation of (b) indicates clearly such a shrinkage behavior as neck growth is followed by pore elimination and also illustrates that grain growth slightly occurs due to homogeneous size of particles.

Fig. 4 shows two more results of MC sintering simulation. In both cases, γ_{sv}/γ_{ss} is 0.5. The case (a) in Fig. 4 has the initial configuration of single phase particles with different sizes. The simulation shows distinctly the behavior of shrinkage accompanied by grain growth. The results simulated for 10000MCS

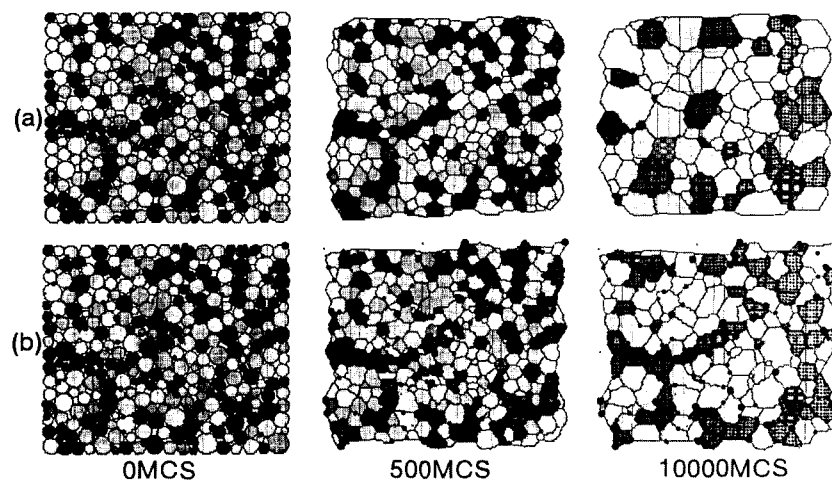


Fig. 4. MC sintering simulations for solid particles. (a) different sizes, (b) second phase particles.

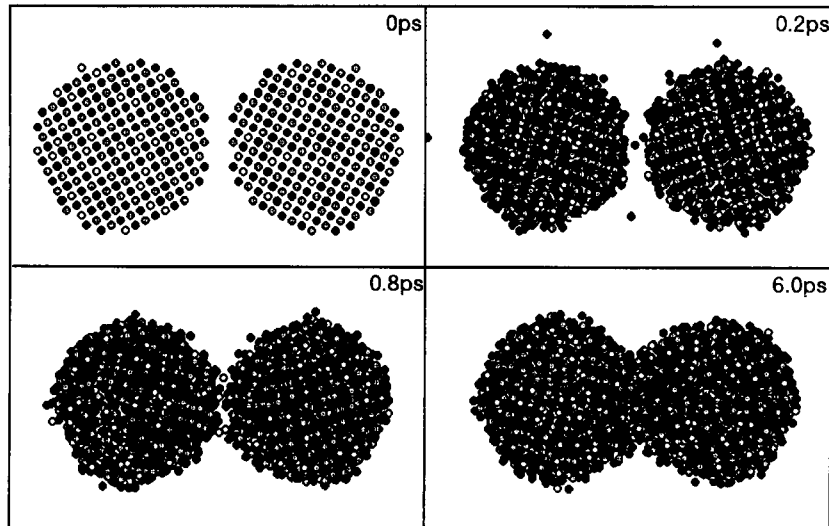


Fig. 5. MD simulation results of sintering of super-fine particles of cubic $ZrO_2-8 \text{ mol}\% Y_2O_3$.

is the microstructure including coarse grains and a residual pore caused by grain growth during sintering. Fig. 4(b) is the simulation of the initial configuration that second phase particles are distributed in the same initial configuration of matrix phase particles as (a). This simulation can treat such a more complicated feature of microstructure development under the interaction of three phases. The second phase particles have not only the inhibiting effect of grain growth but also delaying effect of shrinkage. The final simulated structure illustrates finer grain structure and a little more residual pores comparing the case of the pure material shown in Fig. 4(a). The MC simulation possesses many other possibilities of the design for complex microstructures.

3.2. MD simulation

Fig. 5 shows an example of MD simulation results for cubic Zirconia-Yttria. The starting configuration is that the two super fine grains with 20 nm diameter and cylinder shape have a space of one atomic length and the orientation relationship of $\Sigma 5$ between two grains. The material consists of approximately 4000 atoms of Zr, Y and O ions of $ZrO_2-8 \text{ mol}\% Y_2O_3$ composition and involves the amount of O ion vacancies corresponding to the composi-

tion. Temperature is 1300K and time is up to 6 pico second. The two fine particles join immediately. At the same time, the crystal structure remarkably deforms into amorphous-like structure. Such a behavior is considered to be due to a extremely strong attractive force between super fine particles.

It is possible to calculate grain boundary energies by the MD simulations, which are useful data for MC sintering and grain growth simulations. Such a treatment is available in the systems including inclusion or sintering additive. Anyway, the MD simulations can be a very interesting method for microstructural design at atomic level.

4. Conclusion

The computer simulations of sintering process for ceramics were studied by the methods of Monte Carlo and molecular dynamics.

1. The two mechanisms of mass transfer, the transfer of pore lattices for shrinkage and the transfer of solid lattices for grain growth, were designed in the MC simulation of sintering process at micron size particle.

2. The MC sintering simulations provided the microstructural design for the effects of energy balance,

particle size and second particle inclusion in the sintering process of solid state.

3. The MD simulation was performed in the case of nano size particles of ionic ceramic and showed the characteristic features in sintering process at atomic levels.

4. The MC and MD simulaitons for sintering process are useful for microstructural design for ceramics.

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References

1. R. L. Coble and J. E. Burke: *Progress Ceramics Science* **3** (1963) 197.
2. D. L. Johnson and I. B. Cutler: *J. Am. Ceram. Soc.*, **46** (1963) 541.
3. R. J. Brook: *J. Am. Ceram. Soc.*, **52** (1969) 56.
4. M. P. Harmer: *Advances in Ceramics* 10, *Am. Ceram. Soc.*, (1983) 679.
5. R. J. Brook: *Bulletin. Ceramic Society of Japan*, **31** (1996) 135.
6. M. P. Anderson, D. J. Srolovitz, G. S. Grest and P. S. Sahni: *Acta Metall.*, **32** (1984) 783.
7. G. N. Hassold, I. W. Chen and D. J. Srolovitz: *J. Am. Ceram. Soc.*, **73**, (1990) 2857.
8. H. Matsubara and R. J. Brook: *Sintering Technology* (ed. R. M. German, G. L. Messing, R. G. Cornwall), p. 415, Marcel Dekker, Inc. (1996).
9. H. Matsubara and R. J. Brook: *Fourth Euro Ceramics 3* (ed. S. Meriani, V. Sergo), p. 597, Gruppo Editoriale Faenza Editrice S.p.A. (1995).
10. H. Matsubara and R. J. Brook: *Ceramic Transaction* 71(ed. K. Koumoto), p. 403, The American Ceramic Society (1996).