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Computer simulation of ceramic sintering processes

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In this research, to see the effects of temperature on the grain growth during sintering in the spark plasma sintering process, the grain growth associated with sintering temperatures are simulated by the Monte Carlo Method (MCM) and the Finite Element Method (FEM) for the ceramic material, Al_2O_3.The spark plasma sintering process produces ceramics that are sintered at high temperatures with high energy and cooled rapidly in a short time. The grain growth at the center of the sintered body is different from that in the boundary between the carbon die and sintered body. This phenomenon is explained by the fact that the grain growth during the sintering is mainly affected by heat conduction in the sintered body. For verifying the dependency on heat conduction, the temperature distribution in the sintered body was first found. Based on the temperature distribution, the grain growth in Al_2O_3 in the interior and at the exterior interface with the die at sintering temperatures of 1450°C and 1650°C are simulated and compared with those observed in SEM micrographs. The grain growth results numerically simulated by MCM seem to agree well with those experimentally observed by SEM.

Key words: Ceramic Sintering Process, Al_2O_3, Finite Element Method, Monte Carlo Method, Grain Growth, Computer Simulation.

Introduction

Interest in simulation which replaces difficult, expensive and time consuming experiments has recently increased. The simulation experiment, meaning an extended concept of computer simulation, easily provides much of the information obtained in a real experiment and eventually improves the experimental results through more exact, efficient, and varied functions.

By virtue of the simulation, we can predict and verify material behavior that is hard to obtain by experiment.. Also, using simulation we can get virtual results otherwise obtained from experiments carried out at high temperature. With the rapid development of computer-aided design technology, large and complicated engineering problems, such as those relating to fluid mechanics and structure mechanics, can be solved in a short time using computer analysis methods. Such methods include graphic modeling techniques, Finite Element Method (FEM), Finite Difference Method (FDM), Finite Volume Method (FVM), etc. Large amounts of data from computer analyses can be handled and visually examined to evaluate the analysis using computer graphic visualization techniques.

As precise technology becomes more and more crucial in engineering analysis, the computer-aided simulation can approximate physical behavior in engineering problems by performing engineering calculations and estimations on a numerical model that represents a real problem. Computeraided modeling, visualization and analytical techniques closely approach classical physics.

Spark Plasma Sintering (SPS) processes have been rapidly introduced recently to improve the quality and productivity in ceramic products. Because the spark plasma sintering process is simpler and produces better product quality than the conventional sintering processes, research on grain growth in the spark plasma sintering process is becoming more active. Simulation technology consists of modeling, calculation, analysis, and estimation in that order. Among them, modeling and calculation are the most important in the simulation experiment. Good experimental results can be derived through computer simulation by defining the model size, element components and calculation procedures.

The sintering temperature is known to be the most important factor highly affecting the quality in the spark plasma sintering process. In this study, the temperature distribution of a sintered body was first found by the finite element method. The grain growth associated with the temperature distribution in a sintered body and the relative density were then simulated using the Monte Carlo Method (MCM). The difference in the grain growth between interior and exterior was examined with respect to the effect of low outside temperatures during rapid cooling. The grain growth in the interior is less than that within a certain distance from the surrounding die.

Theory

In order to know the temperature distribution of a

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green ceramic during the sintering process and the temperature dependence of the grain structure, the finite element method is first employed for finding the temperature distribution inside the ceramic and then the Monte Carlo Method is used for visualizing the grain growth.

Temperature distribution by FEM

During the spark plasma sintering process, a high voltage current flows though a carbon die and discharges to generate bulk heat within a short time, so that the temperature of the die becomes very high. In order to sinter the green ceramic, the high temperature heat is transmitted mainly from the carbon die to the center. The sintering temperature is assumed to be the temperature of a thermocouple inserted into the carbon die and the temperature distribution near the center is found by calculating heat conduction from the die.

Looking at formulation processes for FEM analysis, the minimization of the following function in a steady state enables the derivation of the heat conduction equation:

$$\pi_C = U + \Omega_Q + \Omega_q \tag{1}$$

where

$$U = \frac{1}{2}\iiint_V \left[k_x\left(\frac{\partial T}{\partial x}\right)^2\right]dV,\ \Omega_Q = \iiint_V QT dV,$$
$$\Omega_q = -\iint_{S_2} q^* T dS \tag{2}$$

In Eq. (2), S_2 is a surface area over which heat flux q^* is specified. The derived heat equation is expressed as follows:

$$\frac{\partial}{\partial x}\left(k_x\frac{\partial T}{\partial x}\right) + Q = \rho c\frac{\partial T}{\partial t} \tag{3}$$

Temperature in an element {T} is defined with a shape function [N] and a nodal temperature {t}:

$$\{T\} = [N]\{t\} \tag{4}$$

where

$$\{t\} = \begin{Bmatrix} t_1 \\ t_2 \\ \vdots \\ t_i \end{Bmatrix} \tag{5}$$

The temperature gradient matrix {g} is given by

$$\{g\} = \begin{Bmatrix} \frac{\partial T}{\partial x} \end{Bmatrix} \tag{6}$$

Heat flux gradient is expressed as follows:

$$\{q_x\} = -[D]\{g\}$$

where the material property matrix [D] is given by

$$[D] = \begin{bmatrix} k_x \\ 0 \end{bmatrix} \tag{7}$$

Substituting Eq. (4) to Eq. (7) into Eq. (2), one can write Eq. (1) in matrix form as

$$\pi_C = \frac{1}{2}\iiint_V [\{g\}^T[D]\{g\}]dV - \iiint_V \{t\}^T[N]^T Q dV - \iint_{S_2} \{t\}^T[N]^T q^* dS \tag{8}$$

Substituting Eq. (4) into Eq. (6),

$$\{g\} = \begin{bmatrix} \frac{\partial N_1}{\partial x} & \frac{\partial N_2}{\partial x} & \cdots & \frac{\partial N_i}{\partial x} \end{bmatrix}\begin{Bmatrix} t_1 \\ t_2 \\ \vdots \\ t_i \end{Bmatrix} \tag{9}$$

Simply writing Eq. (9) in matrix form,

$$\{g\} = [B]\{t\} \tag{10}$$

Substituting Eq. (9) into Eq. (8) and factoring out the nodal temperature matrix {t} due to the independence, Eq. (11) is derivable:

$$\pi_C = \frac{1}{2}\{t\}^T\iiint_V [B]^T[D][B]dV\{t\} - \{t\}^T\iiint_V [N]^T Q dV - \{t\}^T\iint_{S_2} [N]^T q^* dS \tag{11}$$

Eq. (12) can be obtained by minimizing the functional with respect to the nodal temperature matrix {t}:

$$\frac{\partial \pi_C}{\partial \{t\}} = \iiint_V [B]^T[D][B]dV\{t\} - \iiint_V [N]^T Q dV - \iint_{S_2} [N]^T q^* dS = 0 \tag{12}$$

Shortly expressing Eq. (12),

$$\left[\iiint_V [B]^T[D][B]dV\right]\{t\} = \{f_Q\} + \{f_q\} \tag{13}$$

where force matrices are defined like follows:

$$\{f_Q\} = \iiint_V [N]^T Q dV,\ \{f_q\} = \iint_{S_2} [N]^T q^* dS \tag{14}$$

Defining an element conduction matrix as follows:

$$[k] = \iiint_V [B]^T[D][B]dV \tag{15}$$

by virtue of Eq. (15), Eq. (13) gives an equation for a heat transfer problem:

$$\{f\}=[k]\{t\} \tag{16}$$

Now, the global conduction matrix and force matrix are the sum of all local conduction matrices and force matrices, respectively and are given by

$$[K]=\sum_{e=1}^{N}[k^{(e)}] \tag{17}$$

$$\{F\}=\sum_{e=1}^{N}[f^{(e)}] \tag{18}$$

The global equation is then

$$\{F\}=[K]\{t\} \tag{19}$$

The amount of energy, given as i^2Rt, is a heat energy generated by energy transition rate, $dU/dt=iV$ during the current flow t.

An electromotive force doing work at electric charge can be defined as Eq. (20)

$$dW=\varepsilon dq=\varepsilon i dt \tag{20}$$

By the principle of the conservation of energy, the work done by electromotive force is the same as the heat energy generated:

$$dQ=\varepsilon i dt=i^2Rdt \tag{21}$$

where U is transition energy, ε is electromotive force, q is electric charge quantity, i is effective current, R is resistance and t is time.

The numerical analysis procedure for finding the temperature distribution, based on the FEM formulation stated above, is listed as follows:

1) Kind of elements to be used for the analysis and material properties, heat transfer coefficient, heat capacity, etc. are defined.

2) After generating a circular model with a radius of 10mm, construct a finite element mesh for a quarter circle.

3) Symmetric boundary conditions are imposed on the symmetric lines. In order to impose temperature boundary conditions on the die, the sintering temperature is applied to the outermost boundary nodes.

4) Finite element analysis is performed with user supplied input data.

5) The temperature distribution is obtained and the post-processor visualizes the analysis results.

Grain Growth MCM

Grain growth modeling utilizes the deterministic method calculating the progress of grain growth by moving grain boundaries and peak points of a fine cell structure plotted virtually on the computer according to a method determined in advance. The reasons why the MCM based on lattices is employed to simulate grain growth and the development of fine cell structure in a ceramic compact are explained below.

Firstly, with an experimental method only, it is not easy to control even a few variables that effect grain growth. For example, the estimation of grain growth velocity is not possible because a small number of almost uncontrollable impurities have a big effect on the grain boundary movement . Because the effects of very small factors are so big, it is hard to understand the effect of one factor in the case where many factors are mutually involved.

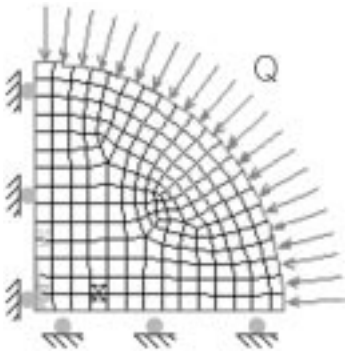

Fig. 1. Finite element model for temperature distribution analysis.

Secondly, the MCM requires assumptions on the shape and distribution of the grains. As the assumption requires many parameters in defining irregularly shaped fine cells, it is very difficult to handle the factors and to compare analytical and experimental results. After matching individual properties to lattice points regularly arranged in a space, the interactions between two lattice points are estimated.

Because of these characteristics, the MCM can overcome the limit on calculation capacity or memory required in the molecular dynamics approach which considers the information of all lattice points at the same time. Therefore, the Monte Carlo system consists of the ensemble of lattice points and its size depends on calculation capacity. Furthermore, because crystalline materials consist of periodic arrangements of cubic crystal lattices, and their properties are determined by the interaction of the atoms located on these cubic lattices, the MCM can be a useful tool for the research on material properties. The 400×400 triangle lattice used in this research has the infinite boundary condition that all ends are assumed to be recycled.

All grain boundaries in the original Voronoi Mosaic method are lines as it has the characteristic of vertically bisecting between the seeds in a continuous space. However, in this study, grain boundaries are irregular as the lattice arrangement has the characteristic to be discontinuous in a given space. The grain boundary creation with irregular forms contributes to the unity of particles when the seeds with the same bearing adjoin one another. This is the reason why such an irregular grain boundary appears.

The grain growth simulation was carried out by the MCM. As seen in Fig. 2, the grain seeds were randomly generated in the lattice and the conversion

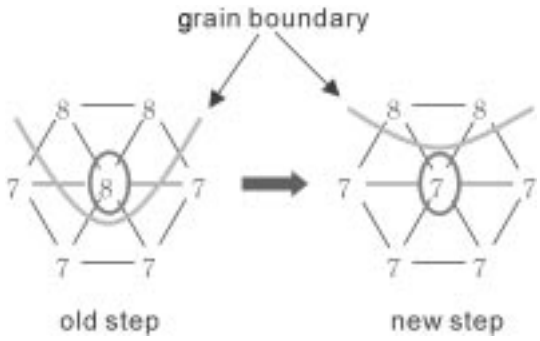

Fig. 2. Monte Carlo step procedure.

energy $\Delta E=E_{new}-E_{old}$ is calculated so that the probability P can be computed. $P=1$ $\Delta E \leq 0$ if and $P=\exp(-\Delta E/k_BT)$ if $\Delta E>0$. According to the value of P, the grain is grown as seen in Fig. 2.

The Monte Carlo simulation procedure for grain growth is listed below:

1) As many grain seeds are generated as needed and randomly spread in a lattice disposition.

2) A lattice point is selected according to the sequence of a lattice disposition and investigated whether or not it is a seed number. If the chosen lattice point is not a seed number, the position of the nearest seed number is sought.

3) The chosen lattice point has the same seed direction number as the nearest seed. When two or more seeds exist at an equal distance, the direction number of the lattice point is fixed with the lattice disposition number investigated in advance.

4) In order to determine a disposition number at a lattice point, the same procedures stated above are repeated.

Simulation Results and Discussion

The temperature distribution in a sintered body which is known to have multi-layers is analyzed using the finite element method. For the sintered body with a diameter of 20 mm, the heat conductivity associated with sintering temperatures is considered in the analysis. The heat generated at the carbon die was calculated from the intensity of electric current, the electric resistance of the carbon die and the time of the flowing current.

Figures 3 and 4 show the internal temperature distributions in Al_2O_3 at the respective sintering temperatures of 1450ºC and 1650ºC, as analyzed by FEM.

A sintered body with a low thermal conductivity, like Al_2O_3, has heat generation by the discharge occurring at the carbon die and the generated heat transfers to the interior by means of heat conduction. The thermal conductivity of Al_2O_3 decreases rapidly as the temperature increases and then decreases gradually. Above 2000 K it increases gradually. Consequently, the temperature

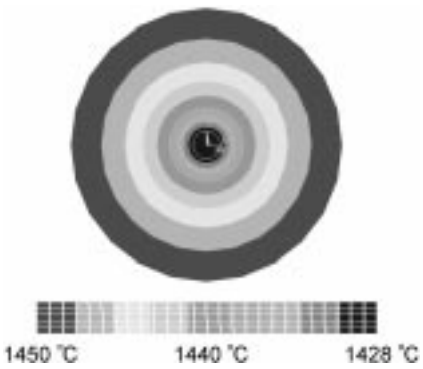

Fig. 3. Temperature distribution of Al_2O_3 at sintering temperature 1450ºC.

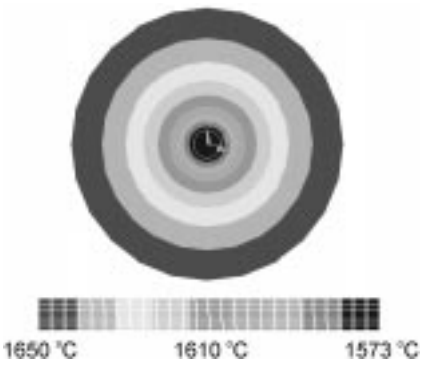

Fig. 4. Temperature distribution of Al_2O_3 at sintering temperature 1650ºC.

distributions between interior and exterior are very similar at sintering temperatures from 1700 K to 2000 K because the difference in thermal conductivity is very small. The temperature at the outer boundary of the sintered body is equal to the sintering temperature and, in the interior, the temperature distribution is established by heat conduction.

Next, the grain growth associated with sintering temperatures were simulated using the MCM. After randomly creating grain seeds inside the 400×400 triangle lattices, the sintering process is carried out by calculating the sintering energy in the neighboring grids around seeds and the simulation results are visualized.

Figure 5 shows the simulation result of grain growth analyzed by the MCM. Figures 6 and 7 show comparisons of simulated grain growth and SEM images for Al_2O_3 sintered by spark plasma sintering process at 1450ºC and 1650ºC. The simulated grain growth at sintering temperatures 1450ºC and 1650ºC were for 200 and 1000 steps, respectively.

Conclusions

The temperature distribution in a sintered body during the sintering process, caused by the heat transfer generated by the discharge of current at the carbon die, was analyzed by the finite element method, considering

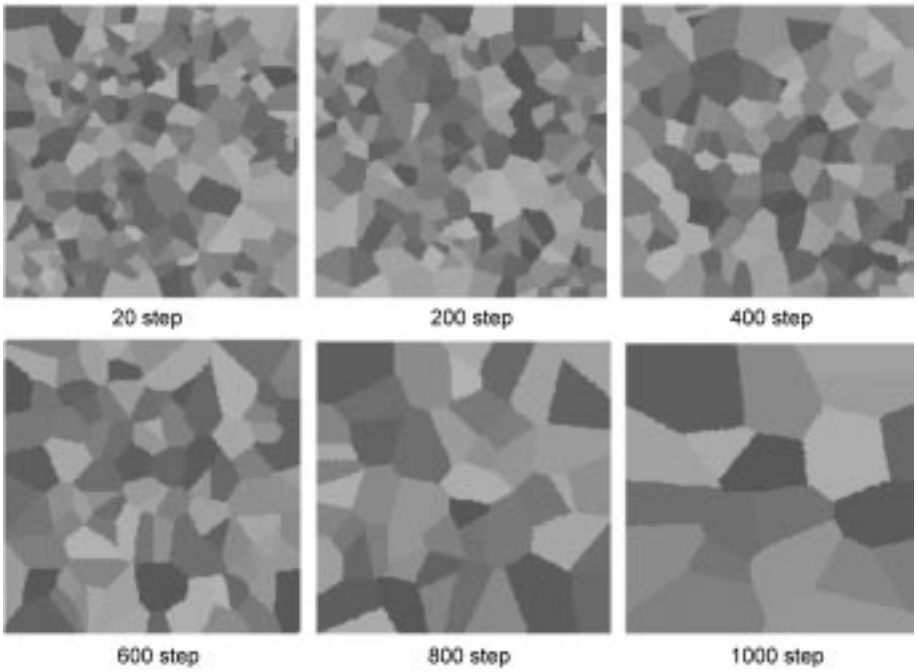

Fig. 5. Shows simulation result of grain growths in various Monte Carlo Steps.

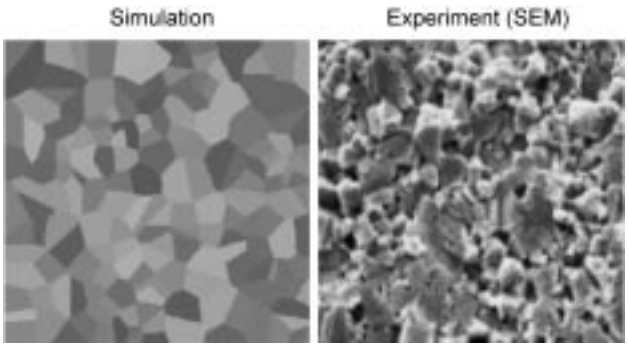

Fig. 6. Comparison of grain growth of Al_2O_3 between SEM and simulation at the sintering temperature 1450°C.

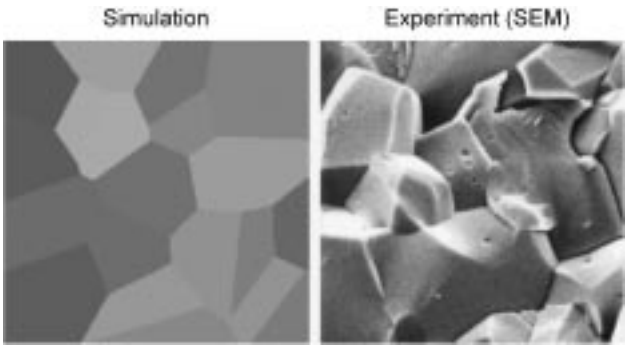

Fig. 7. Comparison of grain growth of Al_2O_3 between SEM and simulation at the sintering temperature 1650°C.

the thermal conductivity of Al_2O_3 associated with the sintering temperatures. The grain growth at sintering temperatures of 1450ºC and 1650ºC were simulated by the Monte Carlo method. As a result of the simulation, the larger grain growth and higher density are verified as the sintering temperature of the body is increased.

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