Computational Materials Science 90 (2014) 16–22

Contents lists available at ScienceDirect

Computational Materials Science

journal homepage: www.elsevier.com/locate/commatsci

The effect of sintering additives on ceramic material sintering densification process based on cellular automata model

Wang min [a,*], Ai xing [b], Zhao jun [b]

[a] College of Mechanical and Electronic Engineering, China University of Petroleum, Qingdao 266580, China
[b] School of Mechanical Engineering, Shandong University, Ji'nan 250061, China

ARTICLE INFO

Article history:
Received 16 December 2013
Received in revised form 24 February 2014
Accepted 27 March 2014
Available online 23 April 2014

Keywords:
Ceramic material
Cellular automata
Sintering additives
Sintering densification process

ABSTRACT

A cellular automata model of composite ceramic material is established which containing sintering additives and pores based on grain boundary energy theory and grain growth boundary curvature driving force theory; then the sintering densification process of ceramic material is simulated. The results show that the density of ceramic material containing sintering additives is higher than the ceramic material without sintering additives, and the grain growth index is lower in the condition of containing sintering additives than without sintering additives. The sintering additives have obvious roles to improve the ceramic material density and get smaller grains. The simulation results are very close to the microstructure of Al_2O_3/TiN ceramic material, the results show that the cellular automata model is applicable to the research of ceramic materials sintering densification process containing sintering additives.

0. Introduction

Ceramic material has high hardness, good wear resistance and high temperature stability, and it has been widely used in machinery, materials, aerospace, electronics, etc. Ceramic material is made by high temperature sintering process from the raw powder, and there is a close link between the microstructure mechanical properties of ceramic material. In the load process of raw powders, there are number of pores between powders. The reduction process of pores is the major process in ceramic material sintering densification process.

With the development of computational materials science, there are some research of materials sintering process uses computer simulation [1]. The research methods include phase-field method [2,3], Monte Carlo method (MC) [4–8], Cellular Automata method (CA) [9–11], etc. The sintering process is a very complex process, and most models are simplified models of process, and computer simulations are based on these simplified models mostly. The most widely used model of ceramic material sintering process is MC method. Hassold et al. [12,13] simulated solid phase sintering process of ceramic material using MC Potts model. Tikare [14,15] simulated sintering densification process of ceramic material based on MC Potts model and vacancy annihilation algorithm. Qin et al. [16] improved MC Potts model and applied it to the three-dimensional simulation of ceramic material sintering process. Wenzhong et al. [17] improved MC Potts model and applied it to two-dimensional simulation of ceramic material sintering process. But the application of CA method on simulation of ceramic material sintering process and sintering densification process is rarely at present.

In this paper, a CA model for composite ceramic material containing pores is established based on grain boundary energy theory and grain growth boundary curvature driving force theory, then the ceramic material sintering densification process is simulated, and the hinder mechanism of sintering additives on grain growth is analyzed. The CA model for ceramic material sintering densification process containing sintering additives is established, and the sintering densification process is simulated. The ceramic material sintering densification process is compared in the condition of containing sintering additives and in the condition of without sintering additives, then the effect of sintering additives on sintering densification process is analyzed. The simulation is contrast with Al_2O_3/TiN ceramic material, and the results show that the model can simulate the ceramic material sintering densification process well; it can be used to reproduce the ceramic material sintering densification process.

1. Modeling

1.1. Cellular automata model

Cellular automata is a stochastic mathematical model, and it is proposed by Von Neumann firstly [18]. It decomposes the system into number of cells, and decomposes time into interval time step

* Corresponding author. Tel.: +86 15266235028.
E-mail address: sdwangmin@163.com (W. min).

http://dx.doi.org/10.1016/j.commatsci.2014.03.059
0927-0256/

(CAS). Each cell indicates its status by orientation (Q), and all cells transit according to the transition rules at each CAS. The states of each cell affect by the state of adjacent cells, and it also affect the state of adjacent cells.

In the grain growth process, the different orientation characterizes different grain [19], One grain including number of adjacent cells that has same orientation values, and there exists grain boundary between the adjacent cells that have different orientation values [20]. In this paper, the two-dimensional quadrilateral cells space is used in simulation area, and Moore type relationship is used in adjacent relationship.

1.2. Physics sense of the model

The driving force of grain growth is grain boundary energy; and it is closely linked to grain boundary curvature. The grain boundary motion is determined by changing of grain boundary energy and grain boundary curvature, if grain boundary is isotropic, grain boundary energy E_i of cell i can be expressed as [21]:

$$E_i = J_i^k \sum_{k}^{nn} (1 - \delta_{S_i} \delta_{S_k}) \tag{1}$$

where J_i^k is unit grain boundary energy, J is 1 in Ref. [21]; nn is adjacent number of cell i, nn is 8 in Ref. [21]; δ is Kronecher symbol; S_i is orientation value of cell i; S_k is orientation value of cell k, and k is a adjacent cell of cell i. If the orientation value of cell i changes into the orientation value of cell j, then the change can be denoted by $\Delta E_{i,j}$ by grain boundary energy [21]:

$$\Delta E_{i,j} = E_j - E_i \tag{2}$$

Sintering additives may have effect of reducing grain boundary mobility, improving surface diffusion rate, promoting lattice and grain boundary diffusion, weakening grain boundaries unevenness and reducing grain surface energy in sintering process [8,22]. Bateman et al. [23] found that MgO does not affect the characteristics of Al_2O_3 grains in sintering process, but it can inhibit Al_2O_3 growth grains significantly. Bennison and Harmer [24] and Wang et al. [25] found that adding a small content of MgO in Al_2O_3 will inhibit grains growth and improve the material density significantly. Coble [22] and Rittidech et al. [26] found that Al_2O_3 grains average grains size decreases with the increase of MgO content, but the effect reduces when MgO content is greater than 0.1 wt%. Jialiang et al. [27], Ben et al. [28], Haitao et al. [29] studied the effect of sintering additives on ceramic materials.

In grains growth process, grain boundary migration driving force comes from the reduction of grain boundary energy. When sintering additives are located on grain boundaries, they occupy part area of the grain boundaries, and grain boundary energy reduces. If grain boundaries want to get rid of sintering additives, grain boundary energy will be increased. When grain boundary migration driving force is insufficient to separate grain boundary and sintering additives, the grain boundary is pinned, and grain growth process will stop [20,30]. It can be obtained by formula (1) that when cell i in grain boundary, if $\Delta E_{i,j} \geqslant 0$, grain boundary will be pinned by sintering additives and cannot migrate; if $\Delta E_{i,j} < 0$, grain boundary cannot pinned by sintering additives and it will migrate out of the grain boundary.

In grain growth process, sintering additives may be in grain boundaries or may be not in grain boundaries. If sintering additives are in grain boundaries, they will reduce grain boundary energy and reduce grain boundary migration drive force; if sintering additives do not in grain boundaries, they have no effect on grain boundary energy. The grain boundary energy of cell i can be expressed as [20]:

$$E_i = J_i^j \sum_{j=1}^{nn} [(1 - \delta_{S_i S_j})(1 - f(j))] \tag{3}$$

where J_i^j is unit grain boundary energy between cell i and its adjacent cell j; nn and δ are same as formula (1); $f(j)$ is a function about sintering additives. If cell j is sintering additive and occupy grain boundary, $f(j) = 1$; if cell j is sintering additive and do not occupy grain boundary, or cell j is not sintering additive, $f(j) = 0$.

In multiphase ceramic material, the total grain boundary energy consists of 5 parts [8,31]:

$$E_{tol} = E_{S_1S_1} + E_{S_2S_2} + E_{S_1S_2} + E_{S_1V} + E_{S_2V} \tag{4}$$

where E_{tol} is total grain boundary energy, $E_{S_1S_1}$ is grain boundary energy between adjacent solid phases 1, $E_{S_2S_2}$ is grain boundary energy between adjacent solid phases 2, $E_{S_1S_2}$ is grain boundary energy between adjacent solid phase 1 and solid phase 2, E_{S_1V} is grain boundary energy between adjacent solid phase 1 and pore, E_{S_2V} is grain boundary energy between adjacent solid phase 2 and pore, there are no boundary between adjacent pores. Grain boundary energy between adjacent solid phases 1 can be expressed as [21]:

$$E_{S_1S_1} = J_{S_1S_1} \sum_{k}^{nn} [1 - \delta(S_i, S_k)] \tag{5}$$

The remaining part of the grain boundary energy can be expressed with formula (5) similarly, and unit grain boundary energy can be expressed as $J_{S_2S_2}, J_{S_1S_2}, J_{S_1V}, J_{S_2V}$.

1.3. Initial conditions of the simulation

The Al_2O_3/TiN ceramic material sintering densification process containing 30% TiN and 2% sintering additives is simulated. In simulation area, 400 × 400 quadrilateral mesh structure and periodic boundary conditions are used; the actual length of each cell's side is 0.1 μm. Before start of simulation, each cell is given a random orientation S_i. The orientation range of Al_2O_3 is 500 and the orientation range of TiN is 500 too [32]; the orientation of Al_2O_3 is in range of $501 \leqslant S_i^{S2} \leqslant 1000$, and the orientation of TiN is in range of $1 \leqslant S_i^{S1} \leqslant 500$; the orientation of sintering additives is $S_i^3 = 1001$, It is assumed that sintering additives neither migrate nor react and diffuse with other composition during the sintering densification process [33]. The TiN particles and sintering additives distribute randomly in simulation area by program. The orientation of pores is $S_i^V = 1002$. Such a definition of each cell's orientation makes that there are grain boundaries between adjacent solids, and there are grain boundaries between adjacent solid and pore. There are no grain boundaries between adjacent pores, which show that adjacent pores are interconnected.

1.4. Determination of the unit grain boundary energy

In densification model of simple cubic accumulation two balls, the relationship between dihedral angles and grain boundary energy is [12]:

$$J_{SS} = 2J_{SV} \cos\frac{\phi}{2} \tag{6}$$

where J_{SS} is grain boundary energy between two solid phases, J_{SV} is grain boundary energy between solid phase and pore; ϕ is dihedral angle. In initial stage of sintering, two balls are not throughout mutually, and dihedral angle $\phi = 0$, $J_{SS}/J_{SV} = 2$, the ratio of unit grain boundary energy is $J_{S_1S_1} : J_{S_2S_2} : J_{S_1S_2} : J_{S_1V} : J_{S_2V} = 2 : 2 : 2 : 1 : 1$.