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Computer simulation of anisotropic grain growth

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Abstract

The process of anisotropic grain growth driven by anisotropic grain boundary energies is investigated by Monte Carlo simulations. A new model is developed for describing the anisotropy of grain boundary energies. This model divides the microstructure into minimum elements which are labeled by different orientation numbers. The surfaces of each minimum element are associated with surface energies for a given orientation and are determined from a Wulff plot. The grain boundary energy is then calculated from the energies of the two surfaces meeting at the boundary and the binding energy of the grain boundary. The possibility of anisotropic grain growth is explored for different shapes of Wulff plot.

Keywords: Grain growth; Anisotropy; Computer simulation

1. Introduction

Grain growth has been the subject of active research for many years due to its importance in controlling the material microstructure, and thus its mechanical and physical properties. However, most of the previous theoretical and experimental investigations were concerned with normal grain growth during which the average grain size increases while the shape of the size distribution remains constant [1]. One of the main objectives of grain growth studies has been to find ways to obtain equiaxed, fine-grain microstructures by preventing abnormal grain growth, a process in which a few grains grow much larger than the rest of the grains.

There is increasing evidence that certain properties of a material can be improved by deliberately introducing anisotropic grains in a fine-grain matrix. For example, fracture toughness of a material with anisotropic grain morphologies can increase as a result of crack deflection and crack bridging behind the crack tip [2]. Electrical and magnetic properties can also be improved with anisotropic grains. Lath-like grains result in very low clamping voltages in ZnO varistors [3].

Anisotropic grain growth is a very complicated process. Many factors can affect the microstructure anisotropy, such as differences in grain boundary energy and mobility, segregation of solutes on different boundaries [4], the presence of a liquid phase [5], interface growth velocity differences during phase transformation [6] and the anisotropy of the interfacial energy between two phases [7,8]. However, there is little fundamental understanding about how these factors affect the evolution of an anisotropic microstructure.

The driving force for grain growth is the reduction in the total grain boundary energy. Therefore, it is natural that one of the most important factors for controlling anisotropic grain growth is the grain boundary energy anisotropy. However, it is almost impossible to quantitatively describe the grain boundary energy anisotropy since a grain boundary has nine degrees of freedom: three for the crystal misorientation, three for the position of the grain boundary plane, and three for the rigid-body translation vector.

Recently, several Monte Carlo simulations have been proposed to determine whether grain boundary energy anisotropy leads to anisotropic grain growth in single-phase materials. Grest et al. performed Monte Carlo simulations employing the Read–Shockley dislocation model for describing the grain boundary energy variation with the misorientation angle, suitable for a low-angle grain boundary [9]. In this model, while the microstructure displays broader grain size distributions than obtained with isotropic grain boundary energies, no anisotropic grain growth was observed. To model anisotropic grain growth, Kunaver and Kolar

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simply separated the grains into two groups: anisotropic grains and isotropic grains. They assumed that the grain boundary energies between isotropic grains were isotropic while the grain boundary energies between anisotropic and isotropic grains were higher in one direction and lower in the perpendicular direction. If the high grain boundary energy is larger than the isotropic grain boundary energy and the low grain boundary energy is smaller than the isotropic grain boundary energy, then anisotropic grains were observed to grow into the isotropic matrix in a Monte Carlo simulation [10]. Such a model, however, is more appropriate for describing the anisotropic growth of one phase with anisotropic grain boundary energy from another phase with isotropic grain boundary energy, e.g. the growth of anisotropic β-Si_3N_4 from the isotropic α-Si_3N_4 matrix, than for the anisotropic grain growth in single-phase materials. More recently, Cai and Welch proposed the grain formation energy concept to explain the evolution of anisotropic microstructure in $YBa_2Cu_3O_{7-\delta}$ ceramics [11]. They emphasized that the anisotropy of bonding energies along different directions in a single crystal control anisotropic grain growth. However, a serious artifact of their model is that the bulk energy of a grain is not rotationally invariant, which leads to abnormal growth of grains having lower bulk energies than the rest of the grains.

In this paper, a new model is proposed for determining the effect of grain boundary energy anisotropy on anisotropic grain growth. The grain boundary energy is determined from the surface energies of two grains meeting at a boundary, and the binding energy of the grain boundary. For simplicity, the binding energy is assumed to be a constant which is equivalent to assuming that the fracture strength of the grain boundary is independent of the orientation of the grain boundary. The surface energy of a grain in a given plane is obtained from the corresponding Wulff plot. The anisotropy of the grain boundary energy is then introduced through the surface energy anisotropy. A two-dimensional model was employed and a variety of Wulff plots were examined to determine the Wulff plot shapes that generate anisotropic grain microstructure.

2. The model

"Minimum elements" are used to construct an arbitrary microstructure. Two-dimensional space can be completely filled only by hexagons, squares, triangles or rectangles. Because previous Monte Carlo simulations have shown that hexagons introduce less artifacts due to discretization of a microstructure [9], hexagons were employed as minimum elements. A minimum element is much smaller than a typical grain in a given simulation, but is large enough that the bulk energies of all minimum elements are the same.

Each minimum element is assigned a number q, which corresponds to its crystallographic orientation. Even though in real materials the possible number of orientations of a grain is infinite, only a finite number of orientations Q are allowed for the computer simulation.

A hexagonal element has six surfaces with two along the orientation $\theta = 0$, two along $\theta = \pi/3$ and two along $\theta = 2\pi/3$. It is assumed that the surface energies of the parallel surfaces are equal. The values of the surface energies of the minimum elements with different crystallographic orientations (different q) were determined from the Wulff plot.

A Wulff plot is a polar plot of surface energy $\gamma(\boldsymbol{n})$, where $\boldsymbol{n}$ is the unit vector normal to a surface, with all vectors $[\gamma(\boldsymbol{n})\boldsymbol{n}]$ from a common origin. The distance from the origin to the Wulff plot envelope represents the energy per unit area of the surface of that orientation. For example, the Wulff plot of a crystal with rectangular equilibrium shape is shown in Fig. 1.

Using minimum elements of different orientations, a microstructure can be constructed. When two minimum elements with the same orientation number are brought together, two free surfaces are eliminated to become a single grain (Fig. 2(a)). The energy decreases in an amount equal to the sum of the surface energies of the two surfaces which were eliminated. When two minimum elements of different orientation numbers are brought together, two surfaces are eliminated but a grain boundary is formed (Fig. 2(b)). The energy change for this process is equal to the difference between the grain boundary energy and the total surface energy of the two surfaces meeting at the grain boundary which equals the binding energy of the grain boundary. Thus, a single grain is represented by a combination of minimum elements with the same orientation number. Grain boundaries are defined by adjacent minimum elements of different orientation numbers.

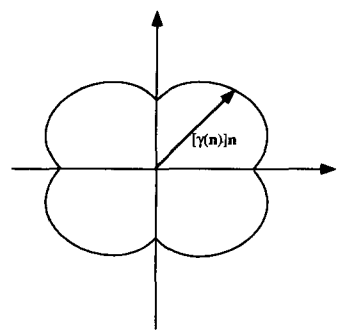

Fig. 1. An example of a Wulff plot.

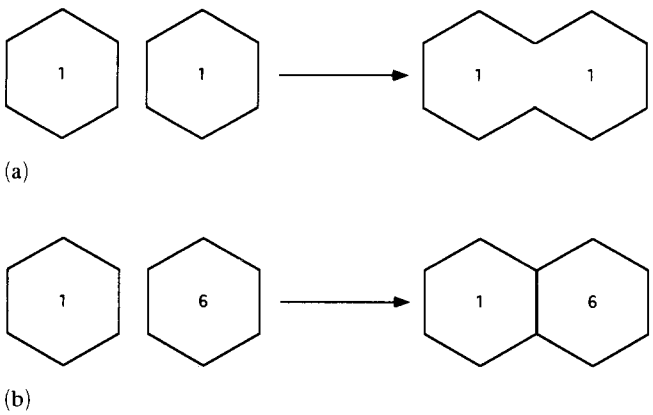

Fig. 2. (a) Combination of two elementary particles to form a single grain. (b) Combination of two elementary particles to form a grain boundary.

Assuming all minimum elements have the same bulk energy, 0, the total energy of a microstructure E_{tot} can be expressed as

$$E_{tot} = E_{surf}^{tot} - E_{surf}^{elim} - E_{gb}^{b} \quad (1)$$

where E_{surf}^{tot} is the total surface energy of all minimum elements comprising the microstructure, E_{surf}^{elim} is the total surface energy eliminated due to the minimum elements having the same orientation number as neighbors, and E_{gb}^{b} is the total binding energy of all grain boundaries in the microstructure. In this model, the interaction energy between two arbitrary neighboring minimum elements can be expressed as

$$E_\theta(q_1, q_2) = J_\theta(q_1) + J_\theta(q_2) - (1 - \delta_{q_1 q_2})J_b - \delta_{q_1 q_2}[J_\theta(q_1) + J_\theta(q_2)] \quad (2)$$

where $J_\theta(q_1)$ is the surface energy of a minimum element q_1 at the plane normal to the axis θ, where θ could be 0, $\pi/3$, $2\pi/3$; $J_\theta(q_2)$ is the surface energy of a minimum element q_2 at the plane normal to the axis θ; J_b is the binding energy of the grain boundary; and $\delta_{q_1 q_2}$ is the Kronecker δ function

$$\delta_{q_1 q_2} = \begin{cases} 1 & \text{if } q_1 = q_2 \\ 0 & \text{if } q_1 \neq q_2 \end{cases} \quad (3)$$

Therefore

$$E_\theta(q_1, q_2) = \begin{cases} 0 & \text{if } q_1 = q_2 \\ J_\theta(q_1) + J_\theta(q_2) - J_b & \text{if } q_1 \neq q_2 \end{cases} \quad (4)$$

The total energy is calculated as the sum over all nearest-neighbor minimum elements

$$E = \frac{1}{2} \sum_{q_1} \sum_{q_2} E_\theta(q_1, q_2) \quad (5)$$

The grain boundary motion, and thus the kinetics of grain growth, can be viewed as a continuous orientation change of minimum elements which can be simulated by using the Monte Carlo technique [9]. In a Monte Carlo simulation, a minimum element is randomly selected, and its orientation is changed at random to one of the other $Q-1$ possible orientation numbers. Then, the change in the system's energy caused by the change in orientation is calculated. Finally, the probability W for changing the minimum element's orientation is determined by the energy change:

$$W = \begin{cases} \exp(-\Delta E/k_B T) & \Delta E > 0 \\ 1 & \Delta E \leqslant 0 \end{cases} \quad (6)$$

where ΔE is the change in energy associated with the reorientation, k_B is the Boltzmann constant and T is temperature.

3. Computer simulation results

In all computer simulations presented below, a microstructure consists of an array of 256×256 hexagonal minimum elements. Periodic boundary conditions are applied to eliminate the finite size effect. The number of allowed orientations Q is equal to 60. Simulation time is measured in terms of Monte Carlo steps (MCS), one MCS corresponding to 256×256 reorientation attempts. The initial microstructure is generated by using a normal grain growth Monte Carlo model in which all grain boundary energies are equal. In order to decrease the computation time, zero-temperature Monte Carlo simulations were performed.

The method for determining the surface energies of a given minimum element from a Wulff plot is illustrated in Fig. 3. Suppose that the minimum element

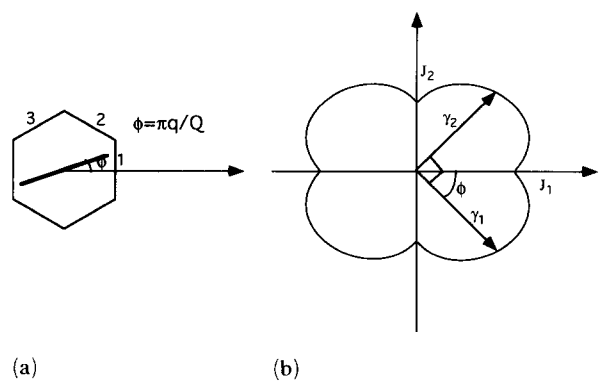

Fig. 3. Determination of the surface energies of an elementary particle oriented along $\phi = \pi q/Q$ from the Wulff plot.

shown in Fig. 3(a) is oriented along $\phi = \pi q/Q$, where q is one of the Q possible orientations. The orientation vector of the minimum element corresponds to the direction along the x axis in the Wulff plot of Fig. 3(b). The surface energy of the plane normal to that orientation is defined as J_1. The angle between the orientation vector and vector normal to surface 1 is equal to $\phi = \pi q/Q$. Thus, the surface energy of surface 1 is γ_1. Since the Wulff plot has a twofold rotation symmetry, the surface energy of surface 2 γ_2 corresponds to a surface orientation which is 90° (instead of 60°) from that of surface 1 in the Wulff plot. The surface energy of surface 3 is set equal to that of surface 1 or 2, whichever is lower. Therefore, grains with orientation number from 1 to 20 have higher surface energies along the $\theta = 0$ direction than the other two directions; grains with orientation number from 21 to 40 have higher surface energies along the $\theta = \pi/3$ orientation; and grains with orientation number from 41 to 60 have higher surface energies along the $\theta = 2\pi/3$ direction.

3.1. Isotropic surface energy

As a first test, the model was examined by assuming that the surface energy of the crystal is independent of orientation. In this case, the Wulff plot is a circle and, by definition, the grain boundary energy is isotropic. Therefore, the model is reduced to a normal grain growth model in which the grain boundaries have the same energy. The temporal microstructure evolution is shown in Fig. 4. The microstructure is isotropic and it has been shown previously that the kinetics of grain growth follows that of the normal grain growth in which grains continue to grow but the grain size distribution does not change with time [9].

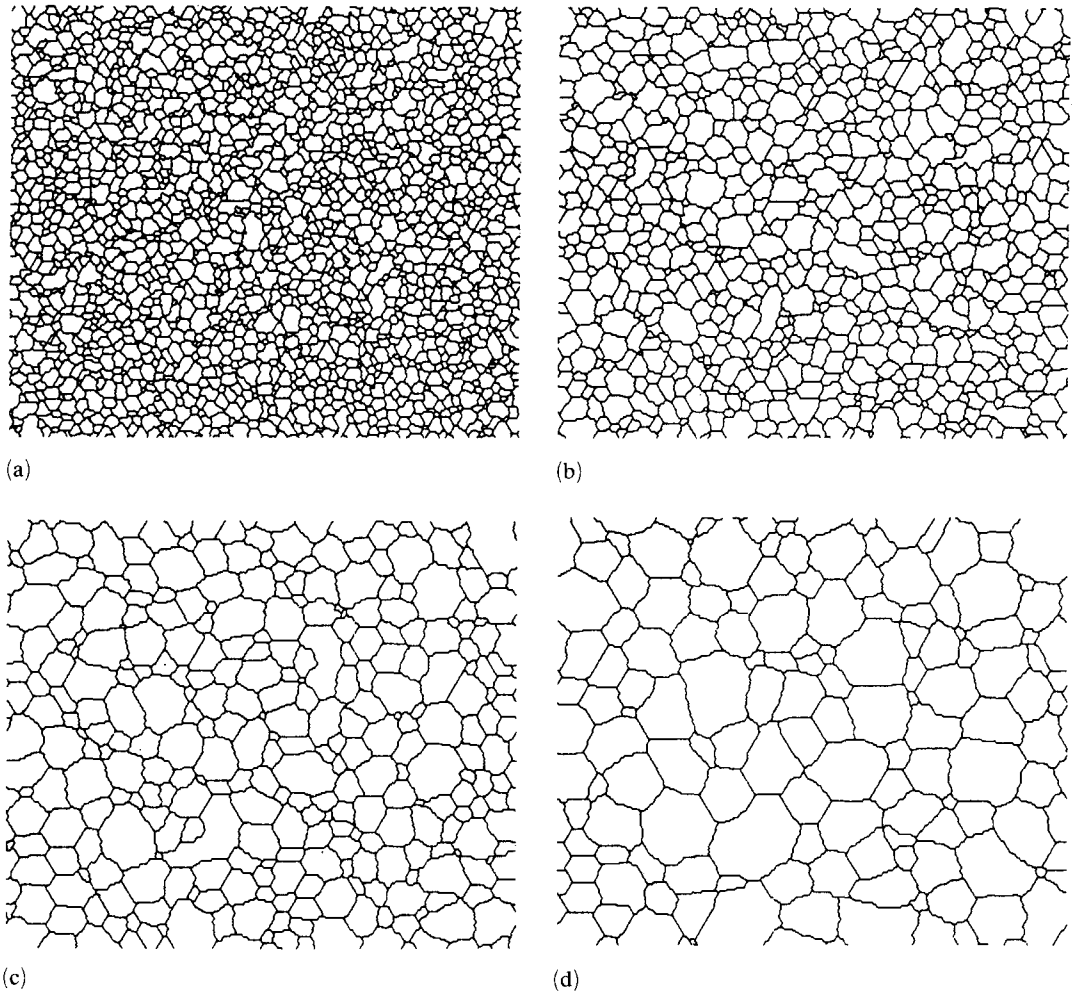

Fig. 4. The temporal microstructure evolution with circular Wulff plot for the case of $Q = 60$: (a) initial stage; (b) 2000 MCSs; (c) 8000 MCSs; (d) 20 000 MCSs.

3.2. Ellipsoidal Wulff shape

One of the simplest anisotropic Wulff plots is an ellipsoid (Fig. 5). For this simulation, it was assumed that the maximum surface energy J_1 is 1.6 and the lowest surface energy J_2 is 0.8. The grain boundary binding energy was assumed to be constant, $J_b = 0.3$. Since the surface energy is anisotropic, the corresponding grain boundary energy is also anisotropic. Therefore, unlike the normal grain growth model, the energy of a grain boundary is a function of the orientations of grains on either side of the grain boundary. The temporal microstructure evolution is illustrated in Fig. 6 for 0, 2000, 5000 and 20 000 MCs. In the initial stage, the grains are very small and isotropic with a narrow size distribution (Fig. 6(a)). With time some grains grow anisotropically and become elongated, while others remain isotropic. The elongated grains grow much faster than the isotropic grains by consuming the isotropic grains. At a later stage of grain growth, the boundaries become straight and the elongated grains impinge upon each other (Fig. 6(d)). The finer grains between the anisotropic grains coarsen.

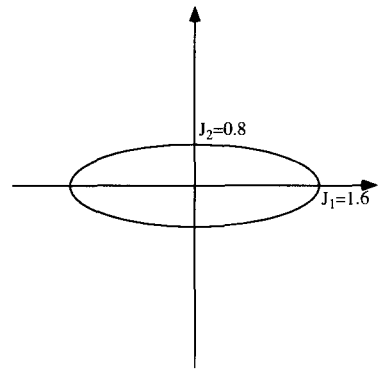

Fig. 5. The ellipsoidal Wulff plot.

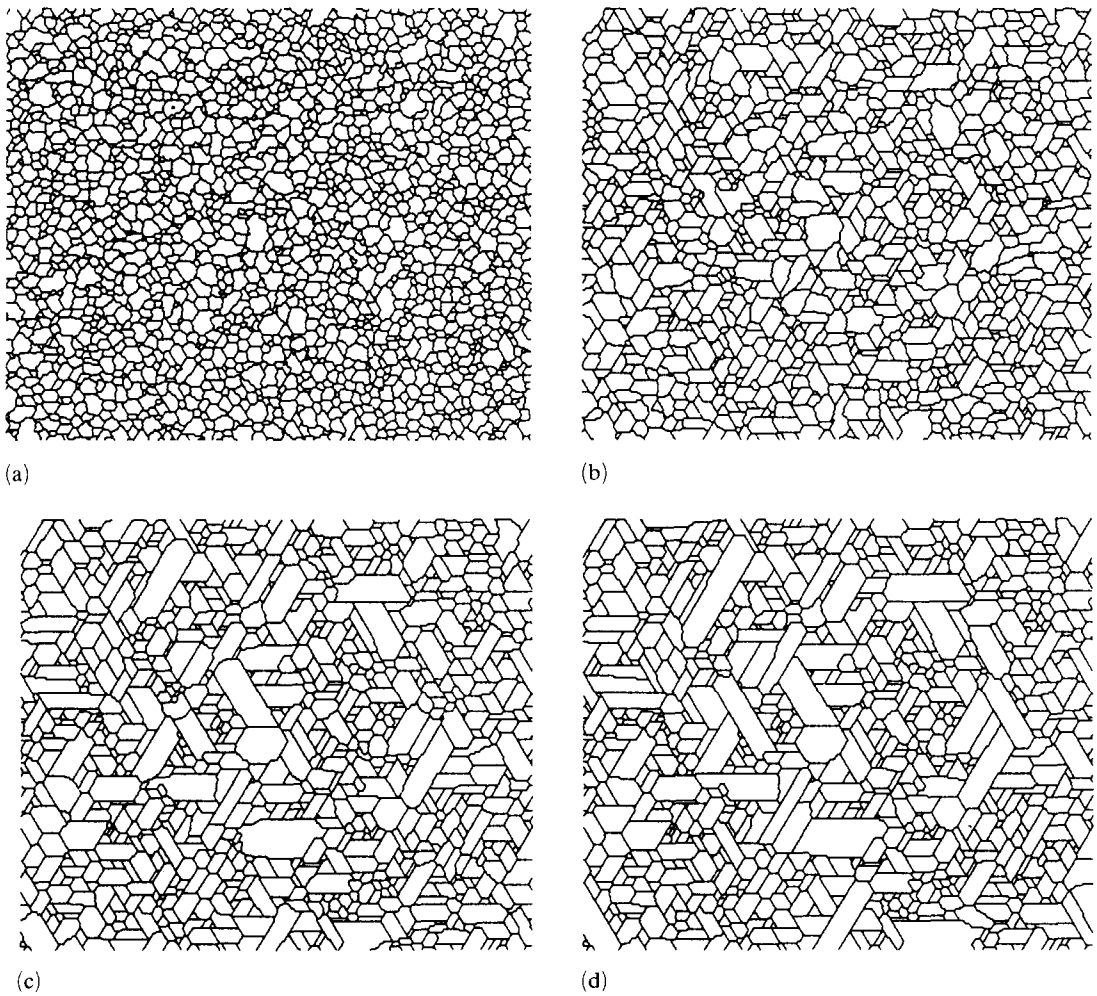

Fig. 6. The temporal microstructure evolution with the ellipsoidal Wulff plot (long axis = 1.6, short axis = 0.8, binding energy = 0.3): (a) initial stage; (b) 2000 MCSs; (c) 8000 MCSs; (d) 20 000 MCSs.

3.3. Ellipsoidal Wulff shape with one cusp

The ellipsoidal Wulff plot with one cusp is shown in Fig. 7 and the corresponding temporal microstructure evolution is displayed in Fig. 8. A comparison of Fig. 8 with Fig. 6 indicates that the overall temporal microstructure evolution in these two cases is very similar. The only difference seems to be that the aspect ratio of the anisotropic grains in Fig. 8 is larger than that in Fig. 6.

3.4. Wulff shape with two cusps

Fig. 9 shows a Wulff plot with two cusps for equivalent surface energies to the system above; but the microstructure generated by this Wulff plot is more like abnormal grain growth and is significantly different from the two previous cases discussed above (Fig. 10). Some grains grow extremely fast at the expense of smaller grains. The microstructure also shows a certain degree of anisotropy due to the surface energy anisotropy, but the aspect ratio is considerably less than the two previous cases.

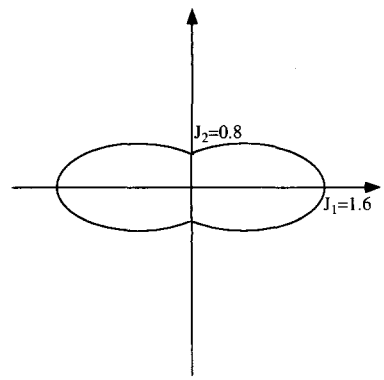

Fig. 7. The ellipsoidal Wulff plot with one cusp.

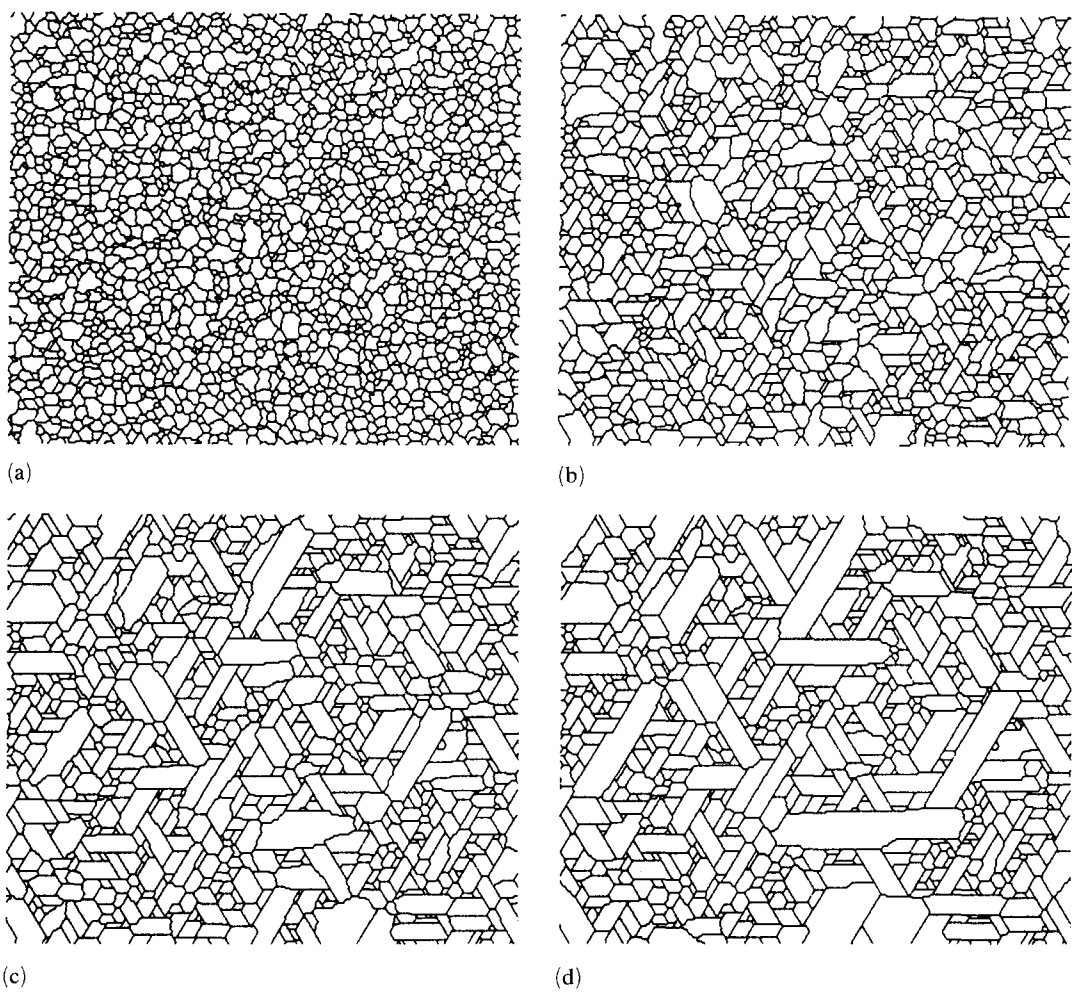

Fig. 8. The temporal microstructure evolution for the ellipsoidal Wulff plot with one cusp (long axis = 1.6, short axis = 0.8, binding energy = 0.3): (a) initial stage; (b) 2000 MCSs; (c) 8000 MCSs; (d) 20 000 MCSs.

4. Discussion

From the above computer simulations, it is clear that the shape of the Wulff plot is a critical factor in the development of an anisotropic grain microstructure. For grains with anisotropic grain boundary energies, the thermodynamic driving force for grain growth depends on the product of two factors, the grain boundary energy per unit area and the grain boundary area. For materials with a circular Wulff plot, the grain boundary energy per unit length (area in three dimensions) is the same in all directions which causes the grains to grow isotropically. However, for materials with a non-circular Wulff plot, the driving force for migration is larger for high energy grain boundaries than low energy boundaries which results in the growth of anisotropic grains. In the initial stage, the grains are small, have a large curvature and grain growth is mainly due to the decrease of the grain curvature. As the grains grow, the grain boundaries become less curved; but the anisotropy in grain boundary energy allows high energy grain boundaries to move faster than low energy grain boundaries, such that the fraction of low energy boundaries increases.

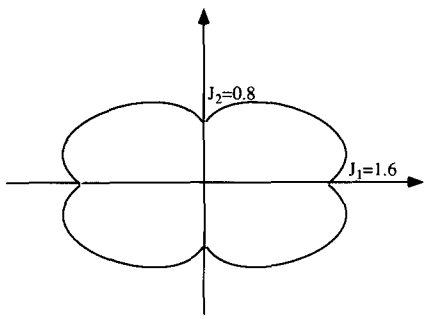

Fig. 9. The Wulff plot with two cusps.

By examining various shapes of the Wulff plot, we found that, to develop highly anisotropic grain structures, the Wulff plot must have a maximum surface

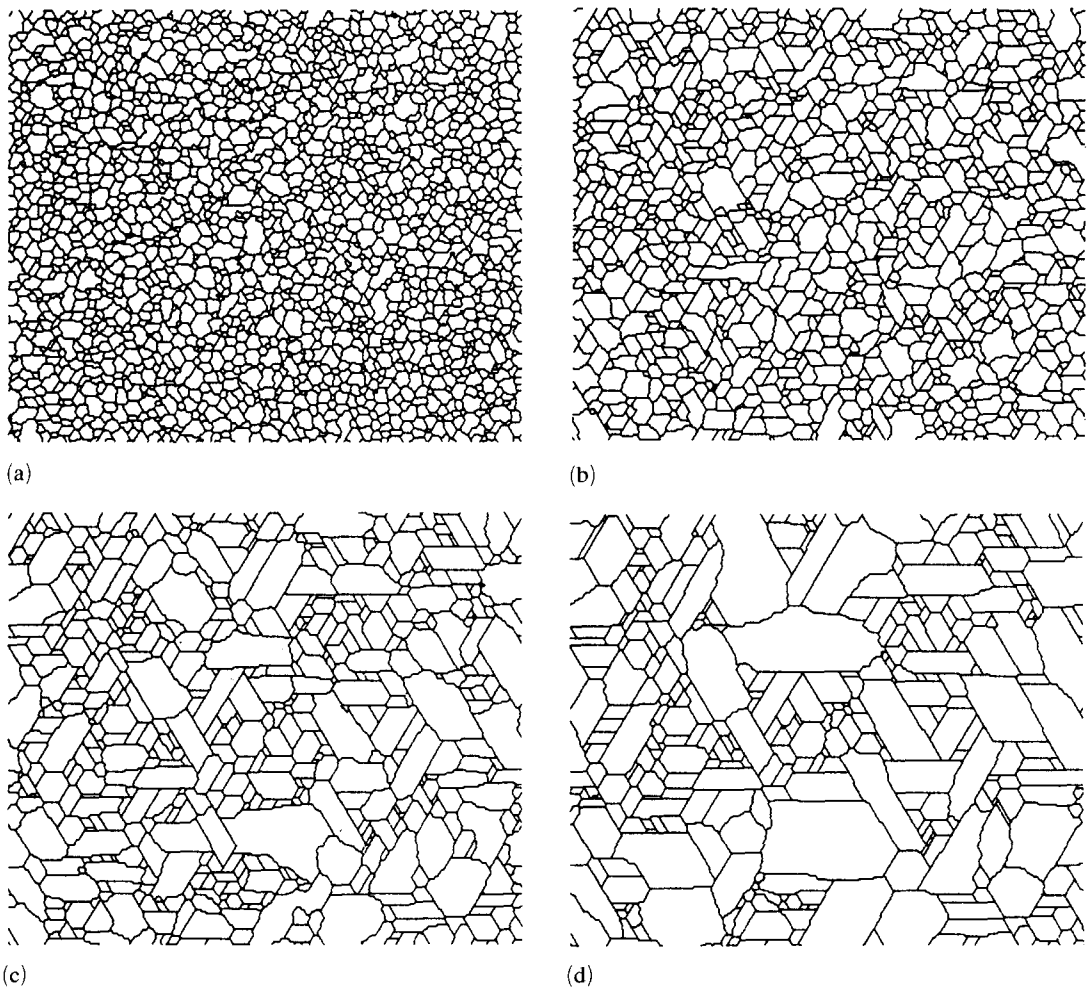

Fig. 10. The temporal microstructure evolution for the Wulff plot with two cusps (long axis = 1.6, short axis = 0.8, binding energy = 0.3): (a) initial stage; (b) 2000 MCSs; (c) 8000 MCSs; (d) 20 000 MCSs.

energy in one direction and a minimum surface energy in the perpendicular direction such that grains with a higher energy on its high energy surface compared to another grain should have a lower energy on its low energy surface. For example, one grain has a surface energy γ_1 on its high energy surface and γ_2 on its low energy surface, the other grain has a surface energy γ_1' on its high energy surface and γ_2' on its low energy surface. If γ_1 is greater than γ_1', γ_2 should be less than γ_2'. The faster growing direction of an anisotropic grain corresponds to the surface normal to the high surface energy whereas the slow growing direction corresponds to the surface normal to the low surface energy. An ellipsoidal Wulff plot and an ellipsoidal Wulff plot with one cusp, discussed above, satisfy this requirement. The planes which move fast are those with high surface energies. When some grains impinge upon each other, one grain has highest surface energy at the high energy surface and lowest surface energy at the low energy surface compared to other grains. This grain's high energy surface tends to grow fast whereas the low energy surface tends to grow slowly such that it becomes elongated and grows larger. Other grains grow slowly or are consumed by elongated grains during the growth competition. If the impinging grains have similar surface energies, they tend to remain isotropic. That is the reason that some isotropic grain regions exist in the microstructure.

After most of the boundaries become straight, the only place where the grain growth is allowed is at a trijunction. The low surface energy grains can still penetrate the grains with high surface energy along the grain boundary until they form the equilibrium trijunction.

Even though idealized two-dimensional models were employed, the observed anisotropic grain structures appear to be similar to those observed in alumina which seems to have a similar Wulff plot to the Wulff plot having one cusp [12]. In alumina, there exist large tabular grains whose basal planes have low surface energy and the plane perpendicular to the basal plane has a high surface energy [13]. The similarity of the anisotropic grain microstructure of alumina and that from our computer simulation is quite remarkable (Fig. 11).

In the case of a Wulff plot with two cusps, a grain having a low energy on its high energy surface compared to another grain should have a lower energy on its low energy surface. A few grains have the lowest surface energies on both high energy and low energy planes. These grains can grow on both sides because this will create more and more low energy surfaces. They grow very fast and consume the small grains. The microstructure evolution shows abnormal grain growth features. Depending on the relative values of the surface energies along the two cusps, anisotropic grains may appear but with low aspect ratios.

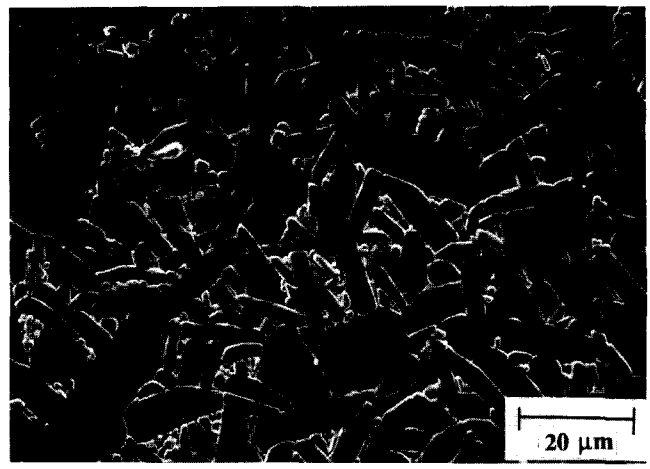

Fig. 11. A typical Al_2O_3 microstructure dominated by large anisotropic grains.

According to Eq. (4) and computer simulation results, when the surface energy is isotropic, the present model is reduced to a normal grain growth model in which all boundaries are assumed to have the same energy.

The present model for grain boundary anisotropy ensures that the bulk energies of all the grains are the same, i.e. the bulk energy of a grain is both translationally and rotationally invariant. In the model of Cai and Welch [11], the bulk energies of grains with certain orientations are lower than the others, i.e. not rotationally invariant, which results in an artifact that the chemical potential per atom (or per unit volume) in the interior of a grain varies as the grain is rotated. Therefore, part of the driving force for abnormal grain growth in Cai and Welch's model is the differences in the chemical potentials of interior atoms between grains with different orientations, which is not consistent with our conventional definition that the driving force for grain growth is the total grain boundary energy.

If minimum elements with anisotropic surface energies and those with isotropic surface energies are mixed together, the present model can be used to model the kinetics of grain growth in two-phase systems in a manner similar to that obtained by Kunaver and Kolar [10].

5. Summarizing remarks

A new model, developed for describing the anisotropy of grain boundary energy in a single-phase material, was employed in a Monte Carlo simulation

technique for studying the possibility of anisotropic grain growth. It was found that, in a single-phase polycrystalline phase, anisotropic grain boundary energy alone may lead to an anisotropic microstructure. Anisotropic grains with the lowest surface energy for the slow moving boundary and highest surface energy for the fast moving boundary, compared to the neighboring grains, can be developed. Effects of other factors such as the initial microstructure, seeding and liquid phase on the anisotropic grain growth kinetics are being examined and will be reported in future publications.

Acknowledgments

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