Determination of the Phase-Field Parameters for Computer Simulation of Heat Treatment Process of Ultra Thin Al Film

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We have simulated grain growth process of an Al ultra thin film by the phase field method. As the model of the calculation, we used the poly crystal model of Kobayashi-Warren in which the phase field and the orientation field are considered. The section of the thin film was divided into 2-dimensional meshes and the time developing equations of the phase field and the orientation field were solved numerically. Therefore the phase field method has great advantage such that it can calculate dynamic process without tracking interface, which often take complicated shape during simulation. Therefore the phase field method has been adopted to study dynamical structural formation process such as pure metal solidification and binary alloy solidification for solid solution and eutectic systems. Recently, calculation of grain growth, dislocation and electromigration become possible.

Phase field is a field parameter with arbitrary value between 0 and 1, which represents the state of phase as a function of location and time. For example in solidification problem, 0 and 1 represent solid and liquid phase respectively and the intermediate value between 0 and 1 means solid-liquid interface. Free energy of the system is expressed as functional of phase field and then the time developing equations are solved numerically under appropriate initial and boundary conditions. Thus the dynamical change of material structure can be obtained as time development of phase field.

In recent years, a wiring delay and an influence of electromigration have been more and more significant with downsizing of wire demanded for high integration of LSI. To solve this problem, crystal grain distribution should be uniform and grain coarsening by heat treatment is necessary. Since these experiments take cost and time, it is difficult to find the most appropriate heat treatment conditions. Computer experiment is a powerful tool to develop such new process with low cost and in short period.

In this research, isothermal annealing process of ultra thin Al film is simulated. As the first step, phase field parameters with an ultra thin film have been determined to perform heat treatment numerical experiments of fine crystal grains. Then isothermal annealing process at various temperatures was performed with the tuned phase-field parameters. In the following section 2 the calculation method is described and results and discussion are given in section 3.

1. Introduction

Phase field method is a newly developed simulation technique for structural formation process of various materials. It has been attracted much attention among wide range of researchers because it has clear theoretical backgrounds based on non-equilibrium thermodynamics and because it can reproduce non-equilibrium effects in material production processes. In addition, the phase field method has great advantage such that it can calculate dynamic process without tracking interface, which often take complicated shape during simulation. Therefore the phase field method has been adopted to study dynamical structural formation process such as pure metal solidification and binary alloy solidification for solid solution and eutectic systems. Recently, calculation of grain growth, dislocation and electromigration become possible.

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2. Calculation Method

2.1 Phase field method

As the model of the calculation, we used the poly crystal model of Kobayashi-Warren in which the phase field and the orientation field are considered. In this section, the time evolution equation, which should be solved numerically, is briefly described. Rotation of a crystal grain is described by the orientation field $\theta$ and by the phase field $\phi$. $\phi = 0$ and 1 represent non crystal and perfect crystal phase respectively. $\theta$ and $\phi$ are both functions of location in space and time and their time developments are described by the governing equations as follows. For orientation field $\theta$,

$$
P(\phi, \nabla \theta) \tau_0 \frac{\partial \theta}{\partial t} = - \frac{\delta F}{\delta \theta} = \nabla \cdot \left[ \kappa \nabla \theta + g \frac{\nabla \theta}{|\nabla \theta|} \right], \tag{1}
$$

and for phase field $\phi$,

$$
Q(\phi, \nabla \phi) \frac{\partial \phi}{\partial t} = - \frac{\delta F}{\delta \phi} = \alpha^2 \nabla^2 \phi - \frac{\delta f}{\delta \phi} \frac{\delta g}{\delta \phi} s|\nabla \theta| - \frac{\delta h}{\delta \phi} \frac{s^2}{2} |\nabla \theta|, \tag{2}
$$

where $\delta F$ is variation of free energy, $f$ is the homogeneous free energy density as a function of $\phi$, $g$ and $h$ are monotonically increasing functions of $\phi$ that becomes 0 at $\phi = 0$ ($g = h = \phi^2$ is assumed here). $P$ and $Q$ are kinetic coefficients concerning $\theta$ and $\phi$ respectively, $\tau_0$ and $\tau_\phi$ are relaxation times, $s$ is interface thickness, $s$ and $\alpha$ represent...
gradient correction factor of $\theta$ and $\phi$ respectively. Equation (1) and equation (2) are solved numerically. $P$ is expressed as

$$P = P(\varepsilon|\nabla \theta|)$$

$$P(u) = 1 - e^{-\beta w} + \frac{\mu}{\varepsilon} e^{-\beta w},$$

where $\beta$ and $\mu$ are correction factor to misorientation field, $\varepsilon|\nabla \theta|$. $Q = 1$ was assumed according to Ref. 1).

### 2.2 Setting of phase field parameter

The system used for calculation is a $500 \times 150$ lattice. This system is divided into a lot of areas by Voronoi division, and for each divided area initial orientation was set at random. This lattice, shown in Fig. 1, is used as the initial structure of poly crystal before heat treatment.

Figure 2 shows the averaged crystal grain radius obtained by real experiments on Al thin film heat treated at 438 K—638 K. This experimental data are from Ref. 9) and they are obtained by simulation is compared with that obtained by real experiments on ultra thin Al film at 478–483 K and 603–613 K. The data are multiplied by the magnification factors in the legend to allow an analysis of all temperatures ranges on this one graph. The points were from real experiments on ultra thin Al film. The data are shown in Fig. 2 assuming Arrhenius type. Next, the relationship between $t_\phi$ and $k$ is determined from the simulation with changing only $t_\phi$. Combining these results, $t_\phi$ is determined as a function of temperature. To verify the validity of the obtained parameter, the annealing experiment at various temperatures was conducted.

### 3. Results and Discussions

Figures 3 and 4 shows the error of the value of $k$ from that of real experiments on ultra thin Al film for 478–483 K and 603–613 K as functions of $\varepsilon$, $\alpha$ and $s$. By roughly changing the values, the error minimized parameters were found to be $\varepsilon = 0.005$, $\alpha = 0.06$ and $s = 0.06$ for 478–483 K, and $\varepsilon = 0.08$, $\alpha = 0.10$ and $s = 0.10$ for 603–613 K.

The obtained parameters are used as the tentative parameters and the values which minimize the error in the vicinity of the tentative parameters are finely searched as shown in Fig. 5. The determined parameters are $\varepsilon = 0.005$, $\alpha = 0.06$ and $s = 0.06$ at 478–483 K and $\varepsilon = 0.07$, $\alpha = 0.09$ and $s = 0.09$ at 603–613 K (see Table 2).

The temperature dependence of the grain growth of ultra thin Al film is assumed to be expressed by the relaxation time $t_\phi$ only. In this simulation, $t_\phi$ is expressed in a reduced unit because the height of the potential barrier in free energy $f(\phi)$
Fig. 3 The error of the value of $k$ at 478–483 K, $\alpha = 0.005$. The error is standard deviation of difference between calculation value and experimental value.

Fig. 4 The error of the value of $k$ at 603–613 K, $\alpha = 0.08$. The error is standard deviation of difference between calculation value and experimental value.

Fig. 5 The time development of square of averaged crystal grain radius $d_{gr}^2$ for the most appropriate parameters. (a) 478–483 K, $\varepsilon = 0.005$. (b) 603–613 K, $\varepsilon = 0.07$.

Fig. 6 The relationship of relaxation time $\tau_0$ and velocity of grain growth $k$ by simulation. (a) low temperature region. (b) high temperature region.

Fig. 7 The relationship of relaxation time $\tau_0$ and temperature $T$. (a) low temperature region. (b) high temperature region.

has not been determined. The temperature dependence of Al grain growth velocity $k$ ($\mu m^2/s$) is obtained from the 2 straight lines in Fig. 2 at 438–443 K and 478–483 K as

$$k = 1.56 \times 10^{20} \exp(-2.44 \times 10^4/T),$$

and at 603–613 K and 633–638 K as

$$k = 8.28 \times 10^6 \exp(-1.07 \times 10^4/T),$$

assuming the Arrhenius type temperature dependence. The relationship between $\tau_0$ and $k$ is determined from the various calculations with changing $\tau_0$ as shown in Fig. 6. The fitted function at 438–443 K and 478–483 K is

$$\tau_0 = 579 \exp(-719k),$$

and at 603–613 K and 633–638 K is

$$\tau_0 = 9.98 \exp(-26.7k)$$

Then, $\tau_0$ is determined as a function of $T$ at 438–443 K and 478–483 K by substituting eq. (4) to eq. (6),

$$\tau_0 = 579 \exp(-1.12 \times 10^{23} \exp(-2.44 \times 10^4/T)),$$

at 603–613 K and 633–638 K by substituting eq. (5) to eq. (7),

$$\tau_0 = 9.98 \exp(-2.21 \times 10^6 \exp(-1.07 \times 10^4/T))$$

Figure 7 shows $\tau_0$ as a function of $T$. The determined parameters are summarized in Table 2.

<table>
<thead>
<tr>
<th>Table 2 Determined phase field parameters of ultra thin Al film.</th>
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<tr>
<td><strong>Low temperature region</strong></td>
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<tr>
<td>$\varepsilon = 0.005$</td>
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<tr>
<td>$\alpha = 0.06$</td>
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<td>$s = 0.06$</td>
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<tr>
<td>$\tau_0 = 579 \exp(-1.12 \times 10^{23} \exp(-2.44 \times 10^4/T))$</td>
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Figure 8(a)–(f) are the final structure of Al grain growth between 480 K and 630 K obtained by the present simulation. Using the most appropriate parameters determined here, the temperature dependence of grain growth is clearly observed. It looks like that abnormal grain growth has occurred from figures at high temperatures, however, the figures show the final structures after a longer calculation than the usual such that the coalescence of the grains has been observed.

4. Summary

Isothermal annealing process of ultra thin Al film is simulated. As the first step, phase field parameters with an ultra thin film have been determined to perform heat treatment numerical experiments of fine crystal grains. Then isothermal annealing process at various temperatures was performed with the tuned phase-field parameters.

Phase filed parameters for the grain growth of ultra thin Al film were determined from the average crystal grain radius obtained by real experiments of ultra thin Al film at 438–638 K. The relationship of $r_0$ and $T$ was determined by combining the temperature dependence of Al grain growth velocity $k$, obtained by real experiments and the relationship of $r_0$ and $k$ obtained numerically. Using the most appropriate phase field parameters determined by the present study, the temperature dependence of Al grain growth can be reproduced.

Various annealing process will be examined in the preceding paper. The constraint of the crystal grain by the wire walls will be examined by reducing the value of $r_0$ near the wall.

REFERENCES