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Phase field simulation of heat treatment process of Cu ultra fine wire

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We have simulated grain growth process of a Cu ultra fine wire 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 fine wire was divided into 2-dimensional meshes and the time developing equations of the phase field and the orientation field were solved numerically. The isothermal annealing was simulated on the condition of the annealing temperature 523 -973 K and the annealing time 333 second, and the constant- rate annealing process was simulated on the condition of the first temperature at 373 K and the final temperature 673 -773 K with heating rate 0.45 -9.01 [K/s]. In order to clarify the effect of heating rate on the crystal structure, the post-isothermal annealing was performed in the simulation to make the thermal energy supplied to the sample equal among the various annealing processes. The change of the crystal grain radius was investigated by varying the heating rate. It was found that the crystal grain radius obtained by a certain range of the heating rate 1.29 -9.01 [K/s] is larger than that obtained by the isothermal annealing.
Key words: computer simulation, phase-field, grain growth and Cu ultra fine wire

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[1-3], dislocation[4, 5] and electromigration[6, 7] has 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 increasing more and more with downsizing of wire caused by high integration of LSI. To solve this problem, crystal grain distribution should be uniform and grain coarsening by heat treatment is necessary[8]. 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 short period.

In this research, isothermal annealing process of Cu ultra fine wire is simulated. As the first step, phase field parameters with an ultra thin wire 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 and annealing process are described and results and discussion are given in section 3.

2. CALCULATION METHOD

2.1 PHASE FIELD METHOD

As the model of the calculation, we used the poly crystal model of Kobayashi-Warren[1, 2] 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 θ and by the phase field ϕ. $\phi = 0$ and 1 represent non crystal and perfect crystal phase respectively. θ and ϕ are both functions of location in space and time and their time developments are described by the governing equations as follows. For orientation field θ,

$$P(\phi,\nabla\theta)\tau_\theta\phi^2\frac{\delta\theta}{\delta t} = -\frac{\delta F}{\delta\theta} = \nabla\bullet\left[h\varepsilon^2\nabla\theta + gs\frac{\nabla\theta}{|\nabla\theta|}\right], \quad (1)$$

and for phase field ϕ,

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

where δF is variation of free energy, f is the homogeneous free energy density as a function of ϕ, g and h are monotonically increasing functions of ϕ that becomes 0 at $\phi = 0$ ($g = h = \phi^2$ is assumed here). P and Q are kinetic coefficients concerning θ and ϕ respectively, τ_θ and τ_ϕ are relaxation times, ε is interface

thickness, s and α represent gradient correction factor of θ and ϕ respectively. Eq.1 and Eq.2 are solved numerically. P is expressed as

$$P = P(\varepsilon|\nabla\theta|)$$
$$P(w) = 1 - e^{-\beta w} + \frac{\mu}{\varepsilon} e^{-\beta w} \quad (3)$$

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

2. 2 SIMULATION OF HEAT TREATMENT PROCESS

The mesh used for calculation is a 1000 × 300 lattice. The lattice 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.

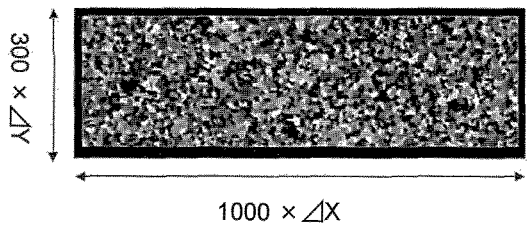


Fig. 1 The initial structure of poly crystal model in the present simulation.

The phase field parameters of Cu ultra fine wire were determined by using the method of Ref.9 as shown in Table I .

Table I Determined phase field parameters

ε = 0.0125
α = 0.0135
s = 0.0235
τ_θ = 20.5exp(-0.0907exp(-2630/T))

In order to investigate influence of heat treatment, the isothermal annealing was simulated on the condition of the annealing temperature 523 -973 K and the annealing time 333 second, and the constant-rate heating process was simulated on the condition of the first temperature at 373 K and the final temperature 673 -773 K with heating rate 0.45 -9.01 [K/s]. In order to clarify the effect of heating rate on the crystal structure, the post-isothermal annealing was performed in the simulation to make the thermal energy supplied to the sample equal among the various annealing processes.

The change of the crystal grain radius was investigated by varying the heating rate.

3. RESULTUS AND DISCUSSIONS

Fig.2 is square of crystal grain radius obtained by simulation on the condition of the first temperature at 373 K and the final temperature 673 -773 K with heating rate 0.45 -9.01 [K/s]. As the results, the rapid grain growth caused by high heating rate was confirmed.

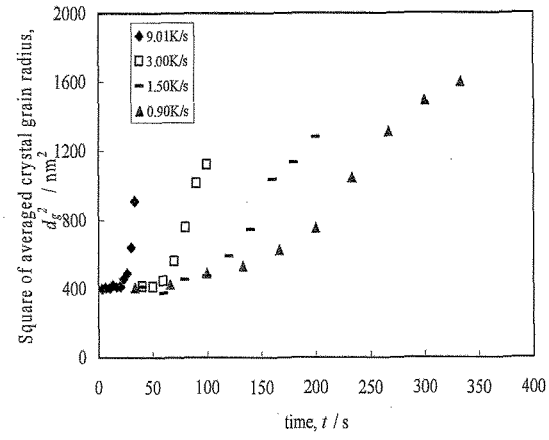


Fig. 2 Time change of the square of crystal grain radius at initial stage under various heating rate

Fig.3 is time change of square of crystal grain radius obtained by simulation of the isothermal annealing at 673 K and the final temperature 673 -773 K with heating rate 0.45 -9.01 [K/s]. The square of the crystal grain radius obtained by a certain range of the heating rate 1.29 -9.01 [K/s] is larger than that obtained by the isothermal annealing.

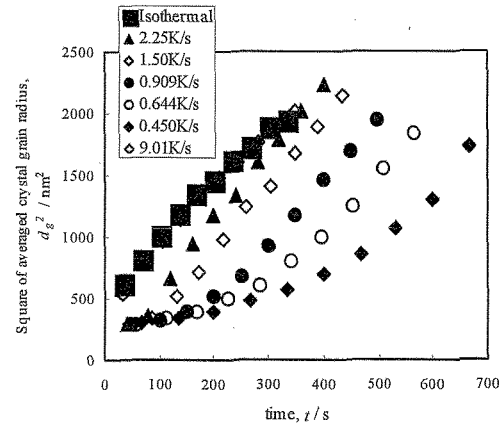


Fig. 3 Time change of the square of crystal grain radius under isothermal annealing only and constant-rate heating followed by isothermal annealing

Fig. 4 is the final grain radius obtained by simulation of the isothermal annealing at 673 K and the final temperature 673 -773 K with heating rate 0.45 -9.01 [K/s]. When the heating rate is appropriately chosen, the grain radius obtained by constant-rate heating followed by isothermal annealing is larger than that of isothermal annealing by about 7%.

Fig.5 is the distribution of the misorientation between a site and its neighbors. In the constant-rate heating case, the shoulder appeared in 1.5 - 3.0° misorientation at 73 s. As shown in Eq.1 and Eq.2, misorientation acts as a driving force of grain coarsening. Therfore the driving force for grain coarsening of this case is considered to be larger than that of isothermal annealing case. This finding can explain the result shown in Fig.4.

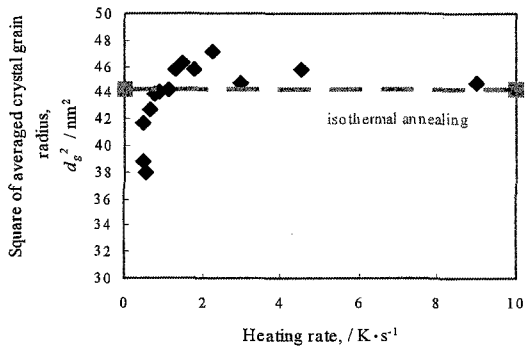
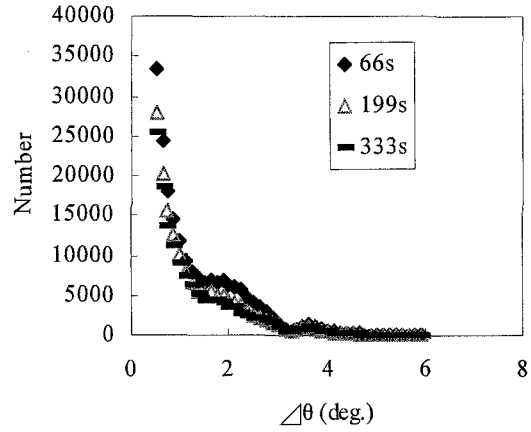
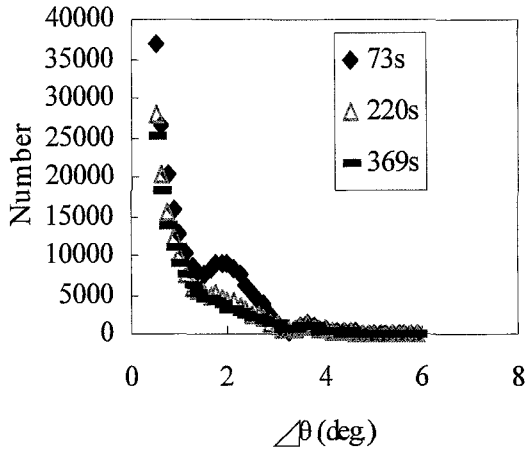


Fig. 4 Obtained final grain radius as a function of heating rate



Isothermal annealing: 333 s, 673K



Constant-rate heating followed by isothermal annealing : Heating rate 2.25 K/s

Fig. 5 Time change of the distribution of misorientation between a site and its neighbors

4. SUMMARY

We have simulated grain growth process of a Cu ultra fine wire 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 parameters for the present calculation have been determined to fit to the results of the grain growth experiment of Cu wires.

In order to investigate influence of heat treatment, the isothermal annealing was simulated on the condition of the annealing temperature 523 -973 K and the constant-rate heating process was simulated on the condition of the first temperature at 373 K and the final temperature 673 -773 K with heating rate 0.45 -9.01 [K/s]. The change of the crystal grain radius was investigated by varying the heating rate. The rate of grain growth during isothermal annealing after constant-rate heating significantly dependent on the grain size at the end of the heating treatment.

When the heating rate is appropriately chosen, the grain radius obtained by constant-rate heating followed by isothermal annealing is larger than that of isothermal annealing by about 7%. The distribution of misorientation was investigated. It was found that the effect of the misorientation is significant in grain coarsening.

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