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Effects of Impurities on the Grain Growth of Aluminium Film

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We performed simulation of the grain growth of polycrystalline Al thin film with impurities by the molecular dynamics method. The examined impurities were O and N of which affinity between Al is strong and weak, respectively. The crystallinity and orientational order of the film were estimated by 2D-Fourier transformation of the atomic structure. The present study revealed that the impurity with strong affinity to Al atom segregates at grain boundary and reduce the grain growth.

Keywords: *molecular dynamics, Aluminium thin film, grain growth, impurities.*

1. Introduction

It is very important to clarify the grain growth process of polycrystalline Al for the development of finely tuned materials. We investigated the growth process of polycrystalline Al thin films with impurities during isothermal annealing by the molecular dynamics method. We focused on the effect of impurities on the grain growth of polycrystalline Al thin film. The effect of impurities on the grain growth of Al is clarified by comparing the results with and without impurities.

After structural relaxation at low temperature (50K), the movements of individual Al atoms were calculated for isothermal annealing temperatures using the molecular dynamics method. The crystalinity and orientational order of the film were examined by 2D-Fourier transformation of the atomic structure.

2. Calculation Method

2.1 Outline of simulation procedure

The polycrystalline structure is constructed by Voronoi division and the number of grains is 20. To introduce impurities in the sample, the randomly chosen Al atoms are replaced by impurities (N or O). The concentration of impurities is set to be 2%. The movements of atoms are calculated by the molecular dynamics at 700K. The interaction between atoms is calculated by the extended Tersoff potential [1]. After relaxation at a low temperature (50K) to stabilize the structure, an annealing temperature is set. Annealing temperature is 700 K, and relaxation time and annealing time are 20 ps and 100 ps, respectively. During stabilizing the structure, pressure of 101 MPa is imposed on the system to suppress void generation in the film.

2.2 Molecular Dynamics Method

The equation of motion of Al atoms was solved numerically. The velocity Verlet method [2] is adopted as algorithm for numerical integration:

$$\begin{cases} r_i(t+\Delta t) = r_i(t) + \Delta t V_i(t) + \dfrac{(\Delta t)^2}{2m_i} F_i(t) \\ V_i(t+\Delta t) = V_i(t) + \dfrac{\Delta t}{2m_i}\{F_i(t+\Delta t) + F_i(t)\} \end{cases} \quad (1)$$

where r_i is the position of the i-th atom, V_i is the velocity, F_i is the atomic force, m_i is the atomic mass and Δt is described time interval. Δt is set to be 1 fs. The interactions between Al atoms and

between heterogeneous atoms are calculated by the extended Tersoff potential [1]. This potential is based on the Tersoff potential [3], and can reproduce the energy of a semiconductor, metal, oxide and other compounds.

2.3 Analysis method

Using two-dimensional Fourier transform (2D-FT),

$$F = \sum_j \left\{ \cos\left(k_x x_j + k_y y_j\right) + i \sin\left(k_x x_j + k_y y_j\right) \right\}, \qquad (2)$$

the obtained structure was analyzed. x_j and y_j are x and y components of the j-th atomic position, respectively, k_x and k_y are those of wave vector. $|F|^2$ is calculated and expressed as a function of k_x and k_y. From the 2D-FT, the crystalline order can be estimated for each specimen and the effects of impurities on the grain growth during annealing will be clarified.

3. Results

3.1 Grain growth of Pure Al thin film

Fig.1 shows the time change of atomic structure of the pure Al thin film during the grain growth simulation. Fig.1(a) represents the initial structure, (b) just after the structural relaxation at 50K, and (c) 120 ps after the isothermal annealing.

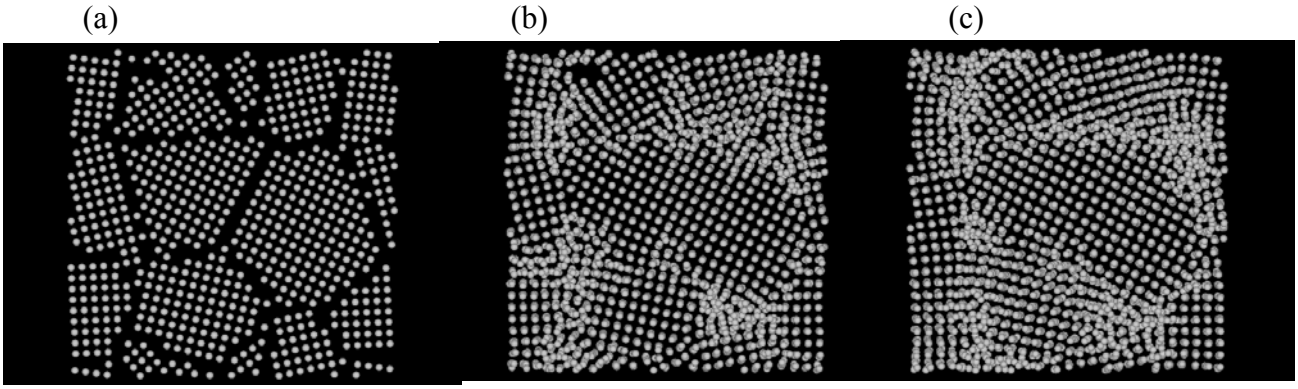

Fig.1 Plane view of the atomic structures of Al(100) oriented thin films. (a) initial structure, (b) just after the structural relaxation at 50K, and (c) 120 ps after the isothermal annealing.

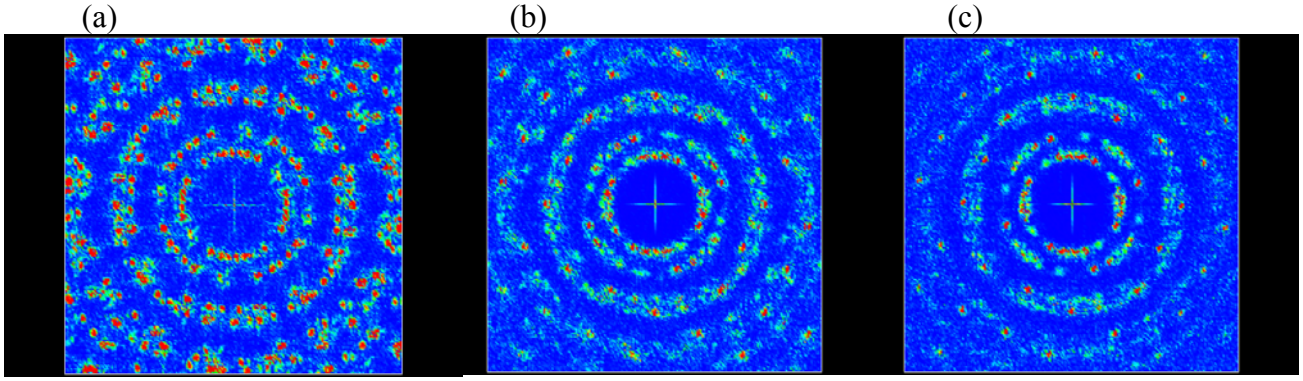

Fig.2 2D-FT patterns of the atomic structures shown in Fig.1(a)-(c). (a) initial structure, (b) just after the structural relaxation at 50K, and (c) 120 ps after the isothermal annealing.

From the comparison of Fig.1(b) and (c), it is clear that the grain growth has occurred. In addition, the voids at upper-left side in Fig.1(b) have disappeared in the final structure as shown in Fig.1(c).

Fig.2(a)-(c) are the 2D-FT patterns of the atomic structures shown in Fig.1(a)-(c), respectively. It is shown from the comparison between Fig.2(b) and (c) that the 2D-FT pattern shows ring pattern just after the structural relaxation at 50K and that the spotty peaks appears and are strengthen even in the high frequency region after the isothermal annealing. It can be concluded that the final structure has possessed high crystallinity. This fact is consistent with the grain growth of Al(100) shown in Fig.1.

3.2 Grain growth of Al thin film with impurity

The randomly selected Al atoms are replaced by impurity atoms, of which positions are the same between Al-N and Al-O systems. Fig.3(a) and (b) shows the final structure of Al-N and Al-O (100) polycrystalline films at 700K, respectively. It can be observed that the oxygen atoms segregate at grain boundaries while N atoms exists in the Al grains. Fig.4 are the 2D-Fourier transform of the Al film shown in Fig.3. The bright spots reflect the (100) oriented coarse grains of Al. The atomic structures are different as shown in Fig.3, however, both of the 2D-FT patterns are very similar.

(a) (b)

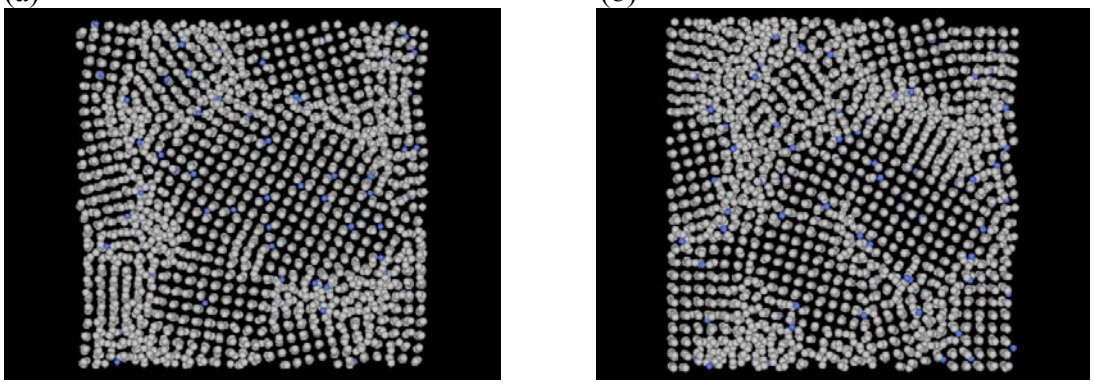

Fig.3 Final atomic structures of (a)Al–N and (b)Al–O systems after the 120ps isothermal annealing. Small circles represent impurity atoms.

(a) (b)

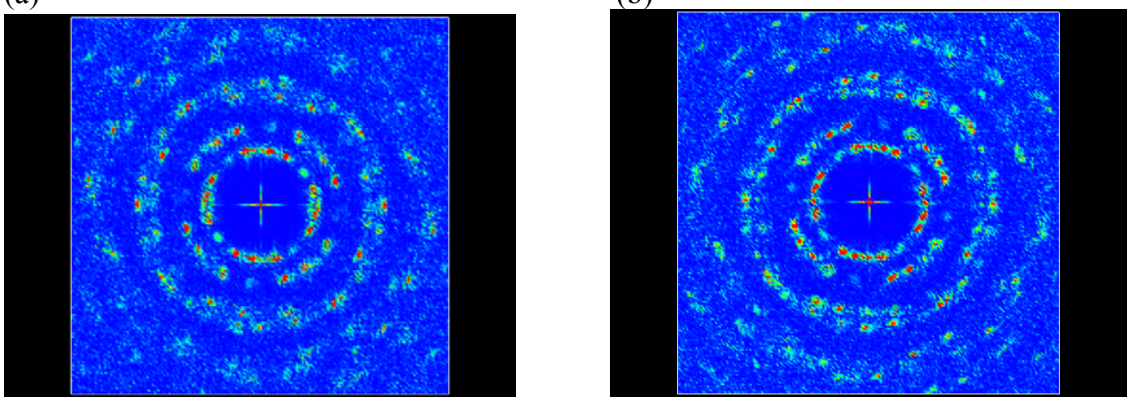

Fig.4 2D-Fourier transform of the (a)Al–N and (b)Al–O systems after the 120ps isothermal annealing. The corresponded atomic structures are shown in Fig.3.

3.3 Effect of impurity atoms on Grain growth of Al thin film

From the comparison of Fig.1(c) and Fig.3(a), it is found that the effect of N on Al grain growth is very weak because the atomic structure of Al after annealing is very similar between pure Al and Al with N. In contrast, the effect of O on Al grain growth is evident as shown in Fig.1(c) and Fig.3(b): the oxygen atoms segregate at grain boundary and prevent the Al grain growth. It can be attributed to the strong affinity of O atoms to Al. In the case of N, boundary segregation does not occur so frequently as in the case of O, because of the weaker affinity to Al than that of O.

4. Summary

We performed simulation of the grain growth of polycrystalline Al thin film with impurities by the molecular dynamics method. The interactions between Al atoms and between heterogeneous atoms are calculated by the extended Tersoff potential. The examined impurities were N and O. The crystallinity and orientational order of the film were estimated by 2D-Fourier transformation of the atomic structure. The present study revealed that the impurity with strong affinity to Al atom segregates at grain boundary and reduce the grain growth. Therefore the impurities with strong affinity to Al atom should be removed to coarsen the grains of polycrystalline Al thin film.

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