ISSN 1062-8738, Bulletin of the Russian Academy of Sciences: Physics, 2016, Vol. 80, No. 10, pp. 1287–1289. © Allerton Press, Inc., 2016.
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Effect of Temperature and Grain Size on the Stability of the Microstructure of Ag Films during Annealing

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Abstract—Grain growth in thin silver films is investigated experimentally. It is shown that the duration of the incubation period preceding linear or parabolic grain growth depends on the annealing temperature and average grain size. It is found that the lower the isothermal annealing temperature and the smaller the grain size, the longer the period of incubation, i.e., the longer the period of microstructure stability.

DOI: 10.3103/S1062873816100191

INTRODUCTION

If it was once believed that grain boundaries were responsible for parabolic grain growth $R^2 - R_0^2 = At$ and triple junctions were responsible for linear growth $R - R_0 = Bt$, the retarding influence that the formation of vacancies which accompany grain growth have on it in thin films is now a topic of discussion [1]. The authors assumed that grain growth was retarded by the formation of vacancies at the first stage, and the formation of vacancies is then responsible for grain growth. The length of the incubation period depends on the initial grain size and temperature. The authors presented the dependence of the duration of the incubation period on the initial grain size and the formula for time in physical parameters:

$$t_{\text{Incubation}} = \frac{(\delta\beta)^2 NkT}{\gamma r_0} \frac{d^2}{D_v c^{\text{eq}}}, \quad (1)$$

where D_v is diffusivity of vacancies, d is the distance between vacancy sinks; r_0 is the average initial grain size; δ is the grain boundary thickness; β is the difference between the relative bulk density and the grain boundary; c^{eq} is the equilibrium concentration of vacancies; γ is the surface tension of the boundary; N is the number of atoms per volume unit; K is the Boltzmann constant; T is the temperature; and t is time.

This formula allows us to estimate the time of microstructure stability. By varying the temperature and initial grain size, we can form the desired stable microstructure. Varying the grain size during the incubation period accounts for only a small part of the initial grain size. This means that the microstructure is stable during the incubation period.

The authors of [2] were the first to present experimental results confirming theoretical predictions [1] regarding the incubation period preceding notable grain growth during isothermal annealing.

The aim of this work was to determine quantitatively the duration of the incubation period during grain growth in silver films, depending on temperature and grain size.

EXPERIMENTAL

We selected Ag 99.99 as the material for our investigation. The samples were nanocrystalline nontextured films 0.1 μm thick, formed via thermal deposition on quartz substrates with a sugar sublayer. The substrate temperature was held at 100°C in order to form the homogeneous fine-crystalline structure.

The films were separated from the substrate by dissolving the sugar sublayer in water and then placed on special small tungsten grids. Freely suspended thin films on grids were annealed in a specially designed furnace at T = 350–450°C with the temperature maintained accurate to 1°C. Heating and cooling to the desired temperature took 100 s. The working vacuum during annealing was kept at the level of 3×10^{-3} N/m^2. The freely suspended films on grids were then placed into an electron microscope, and the structure was determined.

The fine-crystalline structure was visualized using the procedure for forming dark-field images in a transmission electron microscope. The average grain size was 0.01–0.04 μm. The ring intensity in the electron diffraction patterns from these films corresponded to randomly oriented grains.

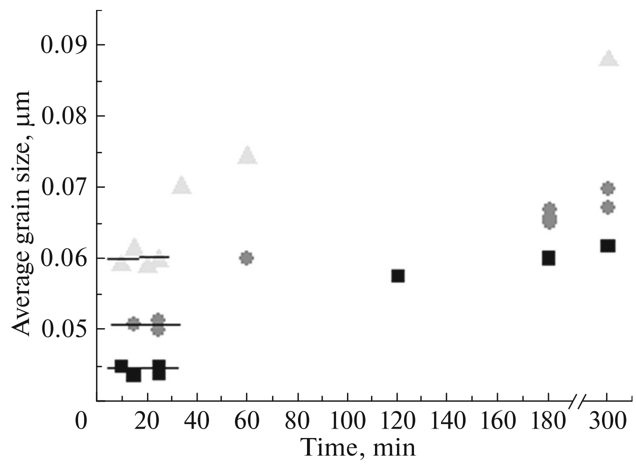

Fig. 1. Time dependence of the average grain size at 450°C.

RESULTS AND DISCUSSION

Figures 1–3 show the time dependences of the average grain size for annealing temperatures of 450, 400, and 350°C.

Grain growth with three different initial grain sizes of 0.045, 0.05, and 0.06 μm at 450°C was preceded by an incubation period 30 min long (Fig. 1). The lower time dependence of the average grain size after the incubation period corresponds to linear growth, while the upper dependences correspond to parabolic grain growth. It is known that grains grow as a result of the simultaneous motion of boundaries and triple junctions of grains, mainly under the influence of the surface tension of boundaries σ. As a rule, it is assumed that triple junctions do not retard boundary motion, and their role is reduced to maintaining the thermodynamically equilibrium angles between the boundaries. The kinetics of grain growth is determined by the mobility of boundaries m_b, and the so-called parabolic law of grain growth applies:

$$R^2 - R_0^2 = At. \tag{2}$$

If triple junctions have finite mobility m_j, they retard boundary migration. The degree of triple junction influence is determined by the relationship between boundary mobility and junction mobility $\gamma = m_b/m_j$, and grain size R. If $R \gg \gamma$, the influence of triple junctions is negligible; if $R \ll \gamma$, boundary migration is determined mainly by the mobility of triple junctions, and

$$R - R_0 = Bt \tag{3}$$

must apply instead of (2).

Such kinetics are apparent in systems with small grains, e.g., thin metallic films [3, 4]. We assume that the triple junction mobility is responsible for grain growth at a grain size of 0.045 μm, and the linear law of grain growth applies. In contrast, boundary mobility becomes responsible for grain growth at grain sizes of 0.05–0.06 μm, and parabolic grain growth is observed.

A similar pattern is seen at 400°C (Fig. 2). Grain growth is preceded by an incubation period 60 min long with different initial grains sizes of 0.045, 0.045, and 0.05 μm. Triple junction mobility is responsible for grain growth at an annealing temperature of 400°C and grain sizes of 0.035–0.05 μm, and the linear law of growth applies.

We succeeded in determining the dependence at 350°C (Fig. 3) only for an initial grain size of 0.045 μm. The incubation period was 120 min. Subsequent growth obeys a linear law.

Figure 4 shows the dependence of the incubation period on the average grain size in the period of microstructure stability. The duration of the incubation period shrinks from 120 min to 30 min with an increase in the average grain size. The smaller the

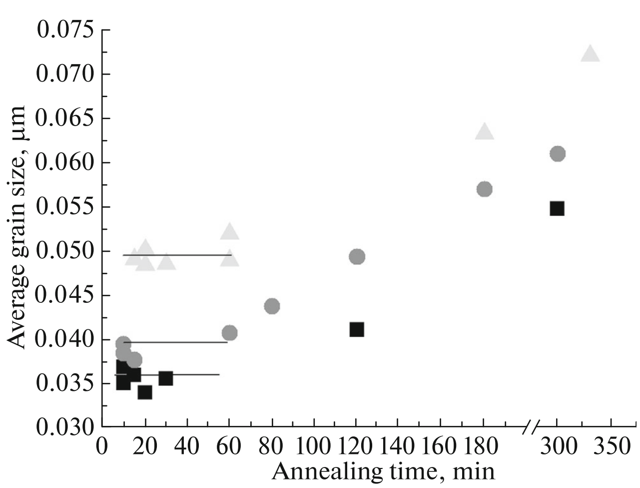

Fig. 2. Time dependence of the average grain size at 400°C.

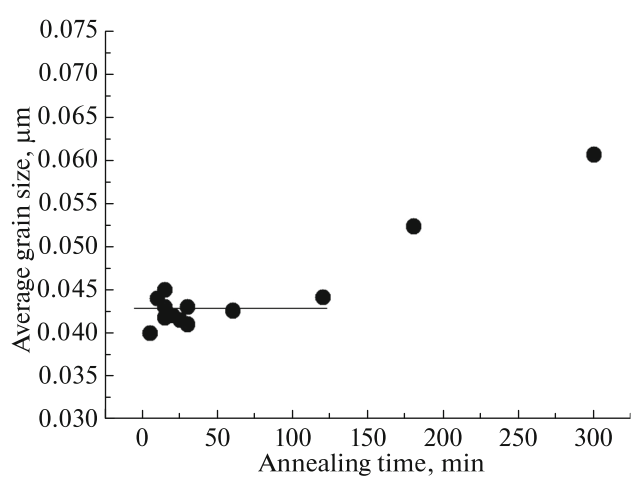

Fig. 3. Time dependence of the average grain size at 350°C.

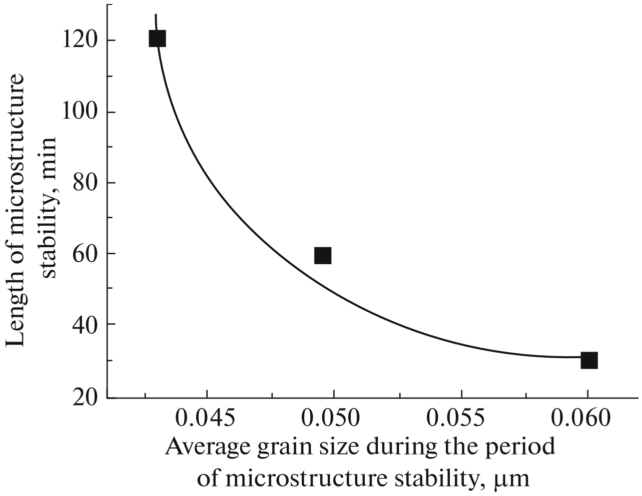

Fig. 4. Dependence of the period of microstructure stability on the average grain size.

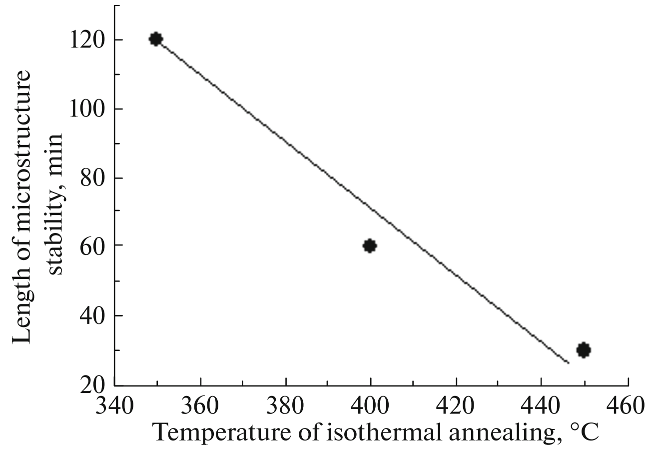

Fig. 5. Temperature dependence of the period of microstructure stability.

grain size, the greater the boundary extension and vacancy saturation of the film, retarding grain growth.

Figure 5 shows the dependence of the incubation period on the temperature of isothermal annealing. The higher the temperature, the more rapidly vacancies leave the boundaries and the weaker their retarding effect.

CONCLUSIONS

(i) Continuous grain growth was observed in the investigated range of temperatures (350–450°C).

(ii) An incubation period in which the rate of growth is low is observed upon annealing, starting at temperatures of 350–450°C.

(iii) The length of the incubation period depends on the annealing temperature. The higher the annealing temperature, the shorter the incubation period.

(iv) The length of the incubation period depends on the initial grain size. The smaller the grain size, the longer the incubation period.

ACKNOWLEDGMENTS

This work was supported by the Russian Foundation for Basic Research and the government of Moscow oblast, project no. 14 42 03556-r_tsentr_a.

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Translated by Nikolai Korovin