

Current dependence of the retardation of precipitation by direct-current stress in Al–3.3 at% Mg

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The effect of direct current (d.c.) stress on the precipitation kinetics in an Al–3.3 at% Mg alloy was investigated by the resistometric method. The reaction rate of the precipitation was found to be retarded by direct current (d.c.) stress, and the result is discussed in terms of the current-assisted-vacancy-annihilation model, which the authors have proposed to interpret the result obtained in Al–5.6 at% Zn and Al–1.8 at% Cu alloys. As a result of subtracting the apparent accelerating effect due to Joule heating, the retarding effect turns out to show a non-linear current dependence in the range of current densities from 4000 to 7000 A cm⁻² although it varies linearly up to 3000 A cm⁻².

1. Introduction

Recent developments in the miniaturization of solid-state circuit impose very heavy constraints on metal lead wires. The operating current density reaches up to about 10⁵ A cm⁻² or 10⁶ A cm⁻². Various kinds of metals or alloys have been tested as lead wires. Of these alloys, the most important is pure aluminium or some kinds of aluminium-based dilute alloys. Under these situations, the authors have investigated the effect of current stress on precipitation for various kinds of bulk aluminium alloys, such as Al–1.8 at% Cu [1], Al–5.6 at% Zn [2], Al–2.7 at% Ag [3] in order to understand the precipitation behaviour under d.c. or alternating current (a.c.) stress from a fundamental point of view.

In Al–1.8 at% Cu, the precipitation reaction was found to be retarded by d.c. stress although an opposite result has been obtained by Koppenaal and Simcoe [4]. They concluded the d.c. stress accelerates precipitation. Moreover, the retarding effect was observed by the authors more clearly in Al–5.6 at% Zn and also in Al–2.7 at% Ag, although not definitely in the latter because of the data scatter. To confirm the generality of this phenomena in aluminium-based dilute alloys, we have also investigated the effect of d.c. stress on the precipitation kinetics in an Al–3.3 at% Mg alloy, which is the same eutectic-type alloy as those already referred to.

2. Experimental methods

An as-drawn wire of Al–3.3 at% Mg, 0.4 mm in diameter, was mounted on a quartz holder as described in [1]. Every heat treatment of the samples was made with this holder. Precipitation annealing, with and without current stress, was carried out in a large water bath [5], whose temperature was controlled within an

accuracy of $\pm 0.1^\circ\text{C}$ for a given annealing temperature. The water near the sample was forcibly circulated to decrease the additional temperature increase due to Joule heating in the sample wire during power annealing. However, an additional temperature increase in the sample could not be completely removed when the applied current was increased.

The sample wire was solution heat treated at 450 °C for 60 min in air, quenched in iced water, left for 5 s and then transferred quickly into a liquid nitrogen bath. Some 10 s were spent during these treatments and the time was kept as constant as possible. Then, the resistivity of the supersaturated sample (ρ_q) was measured by the standard d.c. method. Electric current was supplied to the specimen during power annealing from a constant current source which was stabilized to within $\pm 0.03\%$ of the output value. Interrupting precipitation by quenching the sample in liquid nitrogen, the kinetics were followed by measuring the change in the electrical resistance of the sample at liquid-nitrogen temperature [1, 2]. Eight isothermal curves were thus obtained by varying the electric-current density as a parameter, $J = 0\text{--}7 \times 10^3 \text{ A cm}^{-2}$.

3. Results and discussion

Isothermal annealing curves at 70 °C with and without current stress are given in Figs 1–3. In these figures, the vertical axis expresses the fractional resistance change ($\Delta\rho = \rho - \rho_q$) normalized by the as-quenched value (ρ_q). It becomes evident [6], that in this alloy the resistivity change after quenching from the temperature region of a single phase can be attributed to the decrease of the solute concentration in the matrix due to solute segregation to dislocations, and that the resistivity decreases more rapidly with

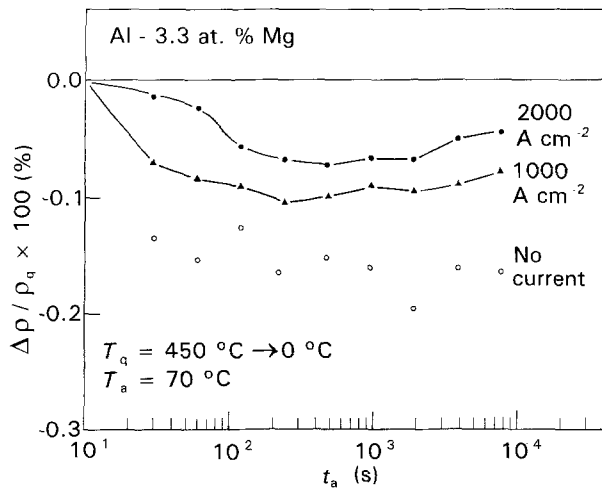


Figure 1 Isothermal annealing curves at 70°C under current stresses of 1×10^3 , 2×10^3 A cm⁻² with reference data of usual furnace annealing.

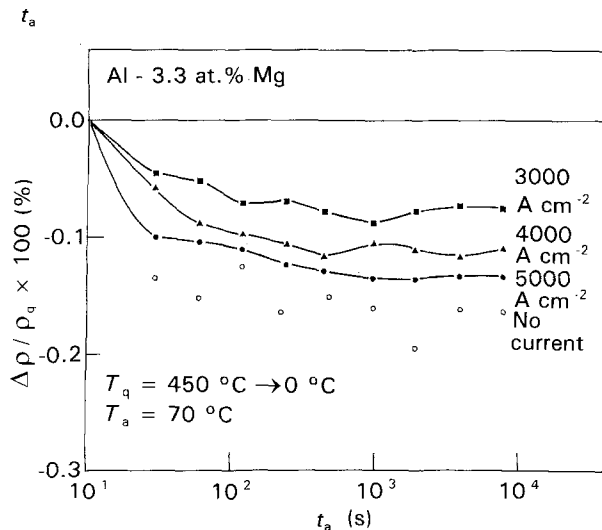


Figure 2 Isothermal annealing curves at 70°C under current stresses of 3×10^3 , 4×10^3 , 5×10^3 A cm⁻² with reference data of usual furnace annealing.

increasing annealing temperature. Then, it can be concluded that the precipitation rate is retarded by the current stress if the resistivity decreases more slowly and accelerated if it decreases more quickly than that in the usual furnace annealing. However, in the former case (retardation) another possible effect resulting from electromigration must be considered because electromigration damage such as voids and/or grain-boundary grooving also increases the resistivity of a sample irrespective of whether it is a pure metal or an alloy. Then, the resistivity increase due to the slowing down of precipitation by d.c. stress must be distinguished from that due to electromigration damage.

Some kinds of structural change induced by electromigration damage cannot completely recover the state before power annealing by high-temperature annealing without current stress. A part of the resistivity change due to the damage, therefore, becomes irreversible for the solution heat treatment. The resistivity increase due to the slowing down of precipitation by current stress is, however, completely

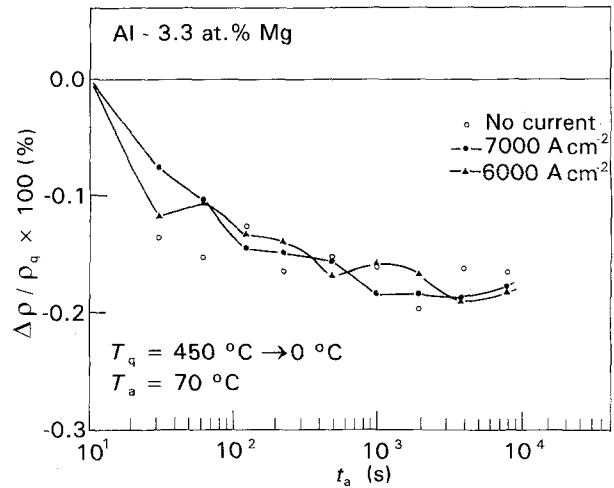


Figure 3 Isothermal annealing curves at 70°C after quenching from 450°C under current stresses of 6×10^3 , 7×10^3 A cm⁻² with reference data of usual furnace annealing.

reversible for solution heat treatment. Thus, these two contributions can be distinguished, and an increase of resistivity can be achieved by examining the behaviour of the resistivity recovery after power annealing. Moreover, there is another useful method to distinguish the resistivity increase due to the two mechanisms described above. This is to examine the time dependence of the resistivity increase caused by power annealing. In electromigration damage, the fractional increment of a sample resistivity $\Delta\rho(t)/\rho^0$ depends on the relation $\Delta\rho(t)/\rho^0 = \exp[(Bt)^n] - 1$ [7], where $\Delta\rho = \rho^J(t) - \rho^0(t)$, $\rho^J(t)$, $\rho^0(t)$ is the resistivity with and without current stress after annealing for t ; B and n are the material constants. Since the value of n is, in most cases, greater than or scarcely equal to unity, the function $\Delta\rho(t)/\rho^0$ becomes a convex function of t . For slowing precipitation, however, the fractional increment of the resistivity should follow the time dependence $\Delta\rho(t)/\rho^0 = A[1 - \exp(-\gamma t) - \gamma t \exp(-\gamma t)]$, which the authors have developed on the basis of a model of current-assisted excess vacancy annihilation. In this case, $\Delta\rho(t)/\rho^0$ becomes a concave function of t .

In the present experiment, every resistivity increment was observed after power annealing had completely disappeared by solution heat treatment, so that the effect of electromigration damage can be neglected and precipitation must be retarded.

However, an explicit comparison could not be made of the time dependence of the resistivity increment between the theoretical and experimental values because of the scattering of the experimental data. Instead, as described later, it was confirmed that the time dependence due to electromigration damage is also inappropriate for our result.

The value of $(\rho^J - \rho^0)/\rho^0 \equiv \Delta\rho(t)/\rho^0(t)$ is re-plotted as a function of current density from the isothermal annealing curves using the annealing time as a parameter and the result is given in Figs 4–6. All these curves show the same tendency: the value of $(\rho^J - \rho^0)/\rho^0$ increases with increasing current density until 2000 A cm⁻² and then decreases gradually. Finally they become even negative at the high current

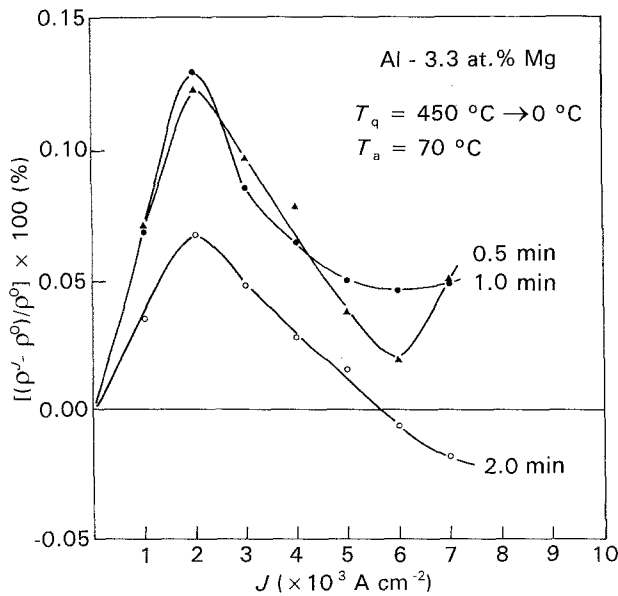


Figure 4 Current dependence of the fractional change in the sample resistivity for various power annealing times, 0.5, 1.0, and 2.0 min.

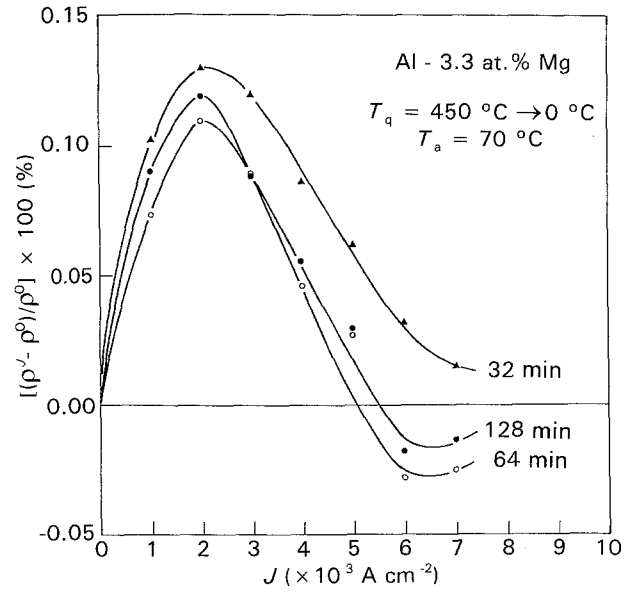


Figure 6 Current dependence of the fractional change in the sample resistivity for various power annealing times, 32, 64, and 128 min.

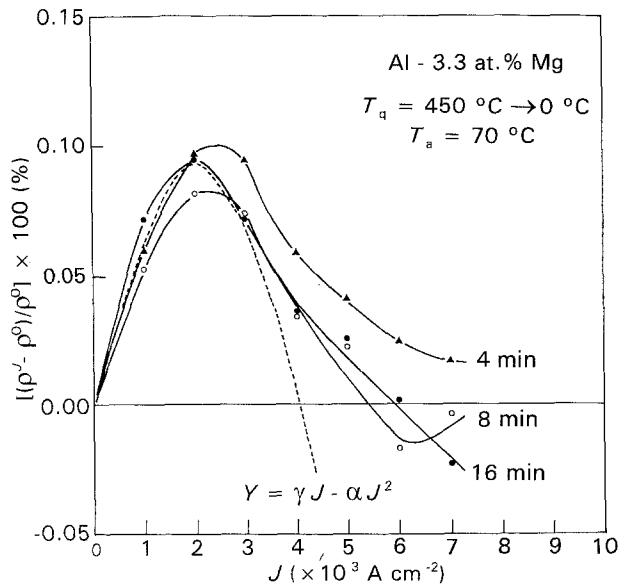


Figure 5 Current dependence of the fractional change in the sample resistivity for various power annealing times, 4, 8, and 16 min. The broken line represents a typical parabola ($Y = \lambda J - \alpha J^2$).

side for some annealing time (8, 16, 64, 128 min). Since the peak values at 2000 A cm^{-2} are nearly constant (not increasing) and tend rather to decrease, it can be concluded that the electromigration damage does not contribute to the resistivity increase observed, at least within the annealing time employed in this experiment (~ 128 min). Thus, the current dependence strongly suggests the existence of the retarding effect of current *per se* on precipitation in this alloy as in Al-1.8 at % Cu [1] and Al-5.6 at % Zn [2].

Our experimental results have been, after all, analysed on the assumption that the reaction rate is retarded if the resistivity change under power annealing slows down in comparison with that under usual furnace annealing. Hence the current dependence of the effect of current stress on precipitation,

taking Joule heating into account, can be expressed as

$$(\rho^J - \rho^0)/\rho^0 = \Gamma(J) - \alpha J^2 \quad (1)$$

where J is the current density and α is a positive numerical constant. The function $\Gamma(J)$ expresses in general the effect of current *per se* and αJ^2 expresses the effect of the temperature increase caused by Joule heating. As can be seen from Figs 4–6, the curves of the current dependence do not show the typical parabolic behaviour ($Y = \lambda J - \alpha J^2$) shown in Fig. 5 with a broken line. This suggests the existence of a non-linear current effect at the high-current side.

The experimental curves have been analysed by the following method in order to subtract the contribution of Joule heating from the measured values. By assuming a linear current dependence for $\Gamma(J)$ in Equation 1 for the weak current as in earlier papers,

$$(\rho^J - \rho^0)/\rho^0 = \lambda J - \alpha J^2 \quad (2)$$

where λ is the retarding coefficient defined in [2]. First, these values of λ and α are determined for each curve using both the maximum value of $(\rho^J - \rho^0)/\rho^0$ and the value of J_{\max} . Again, Equation 1 can be rearranged as

$$\Gamma(J) = (\rho^J - \rho^0)/\rho^0 + \alpha J^2 \quad (3)$$

Thus, the term $\Gamma(J)$ can be calculated from the experimental values of $(\rho^J - \rho^0)/\rho^0$, using the value of α obtained above, as a function of current density. The result is shown in Fig. 7, where the values of the short annealing time have been excluded because the sample temperature could not be so accurately identified because of the large ratio of the transient time to the annealing time, keeping a constant temperature compared to that of the long annealing time. As can be seen from Fig. 7, the value of $\Gamma(J)$ is positive and it depends linearly on the current density until about 3000 A cm^{-2} and the value of λ can be estimated to be $0.8 \sim 1.3 \times 10^{-6} \text{ cm}^2 \text{ A}^{-1}$. This is one-fifth as large as the value of Al-5.6 at % Zn [2] and nearly equal to

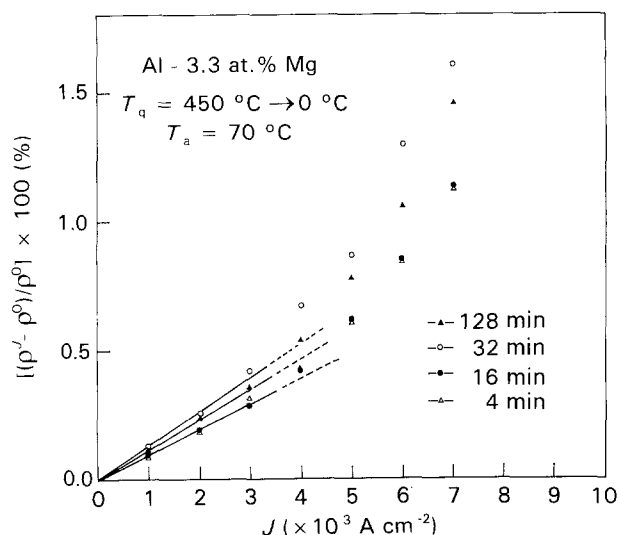


Figure 7 Current dependence of the fractional change in the sample resistivity after subtracting the contribution from Joule heating.

that of Al-1.8 at % Cu [1]. However, the deviation observed on the high-current side is beyond the scope of our weak-current theory of the current-assisted excess vacancy annihilation. Hence, at present, it cannot be decided whether this non-linear behaviour is due to the existence of another mechanism – for example, some reversible component due to electromigration damage in addition to the current-assisted vacancy annihilation – or not.

4. Conclusion

It is evident that d.c. stress retards the precipitation of solutes in an Al-3.3 at % Mg alloy and the retarding coefficient at the lower-current side was determined to be $0.3 \sim 1.3 \times 10^{-6} \text{ cm}^2 \text{ A}^{-1}$, and that the current effect shows a non-linear behaviour for current densities higher than 3000 A cm^{-2} .

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References

1. Y. ONODERA and K. HIRANO, *J. Mater. Sci.* **11** (1976) 809.
2. Y. ONODERA, J. MARUYAMA and K. HIRANO, *J. Mater. Sci.* **12** (1977) 1109.
3. Y. ONODERA, PhD thesis, Tohoku University (1991) (in Japanese).
4. T. J. KOPPENAL and C. R. SIMCOE, *Trans. Met. Soc. AIME* **227** (1963) 615.
5. Y. ONODERA and K. HIRANO, *J. Mater. Sci.* **19** (1984) 3935.
6. K. OSAMURA, T. OGURA and Y. MURAKAMI, *Kei Kinzoku* **31** (1981) 484 (in Japanese).
7. K. P. RODBELL, M. V. RODRIGUEZ, and P. J. FICALORA, *J. Appl. Phys.* **61** (1987) 2844.

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