RELAXATION EFFECT OBSERVED AT LOW FREQUENCY IN AL-CU ALLOY

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Abstract
Al-5 wt% Cu alloys have been studied by isothermal mechanical spectroscopy. The used samples were quenched and matured at room temperature. Experiments were performed in a very large frequency range (10^{-4} Hz – 50 Hz) between room and solidus temperatures. During heating, for each temperature of measurement, experiment started after a hold time long enough for a complete stabilization of the microstructure. In fact the sample has reached stability such as its hardness does not evolve and therefore the transient effects due for instance to θ’ and θ” precipitation were not observed. Nevertheless, a relaxation effect was obtained in the reversion temperature range. This effect is not thermally activated. The maximum of its peak is at about 0.1 Hz and its amplitude increases with the temperature of measurement. It completely disappears after annealing at solid solution temperature and successive slow cooling and therefore is linked to the θ precipitates. Indeed, the experimental conditions are the fact of the gradually formation of the θ precipitates at the grain boundaries of the phase α.

Keywords: Isothermal mechanical spectroscopy, Aluminium cooper alloy, precipitates, non-thermally activated effect, high temperature.

1. INTRODUCTION
The binary Al–Cu system is a well-studied precipitation strengthening system, at date; it forms the basis for a wide range of age-hardening alloys. The aging sequence in Al-Cu alloys is well stated [1] and its precipitates have been revisited and well described [2].

After solution treatment and water quench, Al–Cu alloys exhibit precipitation hardening which depends on many factors and especially on the aging temperature and time. The precipitation sequence observed on aging these alloys is often used as a model system for describing the fundamentals of precipitation hardening [3]: first Guinier–Preston zones (GPI) develop from super-saturated solid solution, and then GPII (θ”), θ’ (semi-coherent) and ends above 300 K with the transformation of the θ’ phase to the incoherent equilibrium θ phase Al2Cu.

Changes of properties during precipitation can be studied by several techniques such as density, resistivity and hardness measurements or the thermoelectric effect but also by the internal friction. The internal friction results on this alloy have been well described previously [4].

It is well known that a long aging time is necessary to obtain stable material properties. However, all these previously reported internal friction measurements have been performed in temperature sweep at a fixed frequency after quench. Consequently, the samples were in continuous evolution during heating and a large part of the described relaxation phenomena were transient effects and non classical relaxation peaks. Even for experiments at decreasing temperatures, a modification of the sample microstructure may occur if the high temperature aging was not long enough. Moreover, experiments made on samples with precipitates show an internal friction spectra exhibiting one or
several maxima due to the temperature change rate; these maxima do not appear during isothermal measurements [5]. Consequently, the aim of this work is to investigate an Al–Cu alloy quenched and annealed at various temperatures; the experiments were performed using isothermal mechanical spectroscopy (IMS) over a very large frequency range after stabilization of the sample microstructure after each temperature change.

2. EXPERIMENTAL PROCEDURE

The material used in this study was a 2024 alloy with composition Al–5 wt.% Cu–1.54 wt.% Mg–0.9 wt.% Mn cut into flat bar samples (60x5x1 mm$^3$), solution heat treated at 820 K under vacuum and then water quenched. Samples were held for 1 month at room temperature before damping experiments. Isothermal mechanical spectroscopy experiments were performed with an inverted forced torsion pendulum described in detail elsewhere [6, 7]. Measurements were conducted under vacuum of 10$^{-5}$ Torr. For this forced vibration technique, the internal friction $Q^{-1}$ is equal to $\tan \phi$, where $\phi$ is the phase lag between the applied stress and the resulting strain. The measurement frequencies were ranged between 50 Hz and 10$^{-4}$ Hz and different frequencies per decade were used. Internal friction was also measured at the pendulum free frequency ($\approx 160$ Hz) by the free decay method. Raw data were corrected according to the suspension stiffness [7]. The maximal applied strain amplitude was $\varepsilon_M = 5 \times 10^{-6}$. The specimen used for the experiments was successively heated from room temperature to 691 K step by step then cooled also by steps to 580 K and then heated again up to 823 K and finally cooled, always by steps till room temperature. For each measuring temperature, the annealing time interval was extended until reproducible damping spectra were obtained. The evolution with time of the microstructure of the sample was controlled by hardness measurements.

3. EXPERIMENTAL RESULTS

Fig. 1a shows the internal friction spectra obtained after annealing at increasing temperatures between 363 K and 553 K, the internal friction spectra can be described as an exponential background. But, above 580 K, a shouldering superimposed on this background is evidenced (Fig. 1b). This effect increases with the measurement temperature but its maximum is always at the same frequency: $\approx 0.1$ Hz for all temperatures, so it is related to a non-thermally-activated process.

For subtraction of this background, we have used a method that we have already widely detailed [8]. After the subtraction of this background, Fig. 2 confirms the increasing of the height peak with increasing the measurement temperature and shows that the maxima position of all the peaks is located exactly at about 0.2 Hz. This damping effect is linked to the presence of $\theta$ incoherent precipitates. In fact, it appeared only after annealing at the reversion temperature when the $\theta$ precipitates replaced the semi-coherent $\theta'$ phase. Fig. 3 confirms that this transformation is not thermally activated because it occurs at the same temperature for frequencies between 40 and 0.2 Hz. This decrease occurs at the same temperature for all the frequencies and corresponds to the disappearance of the $\theta$ precipitates above 800 K when the sample is in $\alpha$ phase solid solution [8].

Fig. 4, exhibits the damping curves without low frequency background corresponding to measurements made at 691K after quench and after slow cooling from this temperature to 656K. Clearly, the maximum of the peak is approximately at the same frequency: 0.2 Hz with a decreasing of its height,
this effect seems also linked to the $\theta$ incoherent precipitates; therefore the microstructure did not evolve after this annealing temperature.

This effect disappears completely after the again heat treatment at 823 K and slow cooling as shown in Fig.5a and Fig.5b. Indeed, in Fig.5a damping measurements have been performed at the same temperature (656 K) during the first heating after quench and after a first in-situ annealing at 691K and then at 823 K in the $\alpha$ solid solution domain. After the last annealing, only a low frequency background is observed and the effect shown in Fig.2 and Fig.4 at 0.2 Hz has vanished. In Fig.5b, the non-thermally effect totally disappears after the slow cooling from 823K whereas the background decreases; this decrease seems correspond to a reduction of the mobile dislocation density. In this state of the material the $\theta$ phase was only present inside $\alpha$ grain boundaries [9]. This disappearance is confirmed in Fig.6, which shows the damping measured at 1Hz versus the measuring temperature. Internal friction.

Fig.1. Internal friction spectra at various increasing temperatures after quench: (a) between 363 K and 553 K – (b) between 581K and 798 K. The annealing temperature is the same as the measuring temperature.

Fig.2. Internal friction spectra at various increasing temperatures between 581 K and 798 K after subtraction of the low frequency background.
exhibits a maximum at about 800 K and the damping measured after annealing at 820 K is lower than that measured during heating.

4. DISCUSSION
Experiments carried out on this type of alloy between ambient temperature and 553K by different authors [10, 11, 12, 13, 14, 15] working in temperature sweep at fixed frequency were therefore able to identify essentially transient effects related either to the formation of GPI zones, or the formation or evolution of precipitates θ” and θ ’ or Zener peak corresponding to a relaxation mechanism associated with point defect jumps. Furthermore, this peak can only be observed just after quench when the Cu atoms are dispersed in the supersaturated aluminum α phase. After a high temperature annealing in α solid solution (above 800 K) and low cooling, the Cu atoms are mainly in Al2Cu precipitates and the peak disappears. In all cases, these effects appear only during the first climb and are absent for measurements during cooling if the holding temperature is sufficiently high. Due to the experimental procedure used for this study, these effects cannot be observed.

In the case of semi-coherent precipitates, interface dislocation motions can explain an internal friction peak [16]. However, a non--thermally-activated effect has already been evidenced for several metallic alloys: in Al-12 wt% Mg linked to the presence of β precipitates [17], in single crystalline superalloys with γ’ precipitates in the γ-Ni3Al phases [18] or in Cu-Cu2O internal oxidized alloy [19]. For each case, the effect occurred in the reversion temperature range when a change of temperature induced a phase change at the interface of the solid solution and the precipitate.

The effect we observed can probably be explained by an interaction between dislocations and precipitates inside the α grains. We can assume that at high frequency, dislocation segments can not cross the precipitates which play the role of anchor. In this case, the segments of dislocations oscillate between the anchor points giving a peak whose frequency is related to the length the segments [20] as the non-thermally-activated effect observed at 0,2 Hz after quench and after slow cooling from 691K. But at very low frequency, all segments cross precipitates, and found continuous background exponential characteristic movements of very long dislocation segments through a grain or sub-grain. In the absence of precipitates, only the continuous background will be highlighted, as in the case of measures after annealing at 820K when the θ phase was only present inside the α grain boundaries.
Fig. 5a. Internal friction measured at 656 K after quench, after annealing at 691 K and after annealing at 823 K. (b) Internal friction spectra at various decreasing temperatures after annealing at 823 K.

Fig. 6. Internal friction measured at 10 Hz before and after annealing at 823 K.
LITERATURE