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Early Stages of Ω Phase Precipitation in Al-Cu-Mg-Ag Observed in Situ with and without Applied Stress by Small Angle X-ray Scattering

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The structural hardening obtained in high-strength aluminium alloys originates from a fine dispersion of precipitates in the matrix. The most efficient hardening shapes are non isotropic (typically needles or plates). Both interfacial energy and stress due to misfit between precipitates and matrix can explain the formation of these anisotropic precipitates.
This paper presents a preliminary study which evaluates the influence of an externally applied stress on the precipitation of plate-shaped precipitates of the Ω (Al_2Cu) phase in an Al-Cu-Mg-Ag alloy. We have used small-angle X-ray scattering (SAXS) to record in situ the formation of the Ω phase under different level of external stress. SAXS gives access to the size and the relative volume fraction of precipitates.
It is shown that different level of stress produce little difference in the observed Ω precipitates. Before the precipitation of the Ω platelets, clusters of atoms, presumably Ag- and Mg- rich, are forming. Oppositely to Ω, the shape of these clusters seems to be affected by the external stress.

Keywords: *precipitation, Al-Cu-Mg, small-angle X-ray scattering (SAXS), clusters, stress ageing.*

1. Introduction

The Al-Cu(-Mg) system, belonging to the 2000 series alloy, is an important high-strength structural hardening system [1]. As an historical model system for precipitation hardening, it has been extensively studied, especially the precipitation of the θ' (Al_2Cu) phase which forms platelets in the aluminium {001} planes. The addition of microalloying elements such as Mg and Ag is known to modify the precipitation sequence and favor the formation of another metastable phase, Ω (Al_2Cu), which precipitates as plates in the aluminium {111} planes [2-3]. These Ω precipitates have a very high aspect ratio and have a high coarsening resistance [4].

The role of the microalloying elements Mg and Ag has been studied in details by different authors [2,4]. These elements are shown to segregate at the Ω /matrix interface, presumably to lower the interfacial energy and/or accommodate elastic stresses. Reich et al. [2] also showed by atom probe tomography that before the precipitation of the Ω phase, small diffuse clusters rich in Mg and Ag appear. They are thought to act as heterogeneous nucleation site for the Ω precipitates.

In this paper, we will use small-angle X-ray scattering (SAXS) to record in situ the formation of the Ω phase under different level of external stress. SAXS enables the in situ measurement of the size and the evolution of the volume fraction of the forming phases. The formation of a precursor to the Ω in the shape of small atomic clusters, as observed by Reich et al. [2] will be also investigated.

2. Materials and Experiments details

The studied alloy is an Al-Cu-Mg-Ag alloy with composition given in Table 1. Flat tensile specimens were cut by electro-erosion from 1mm thick rolled sheets and were solution heat treated for 30 minutes at 520°C and water quenched. The ageing was performed in situ in a heating and straining apparatus designed to be operated under vacuum in the SAXS setup. A picture of the straining and heating device as well as a sketch of the tensile specimen used in our experiments can be seen on Fig. 1.

Table 1. Composition of the alloy (wt%).

Cu	**Mg**	**Ag**	**Al**
4	0.3	0.4	balance

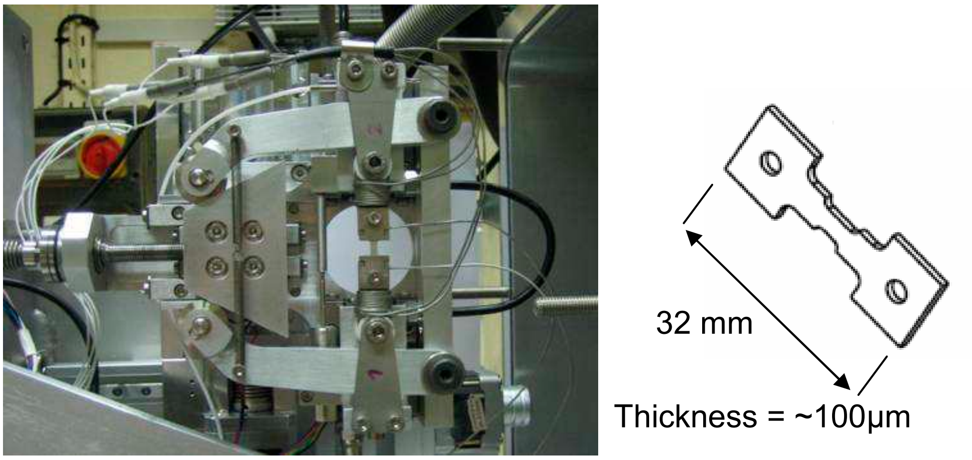

Fig. 1. *In situ* straining and heating device and geometry of the tensile specimens.

The SAXS experiments were performed on the SIMAP rotating anode setup. The X-ray energy was that of the the Cu anode K_{α}. The distance between the sample and the CCD two-dimensional detector was 0.59m (measured by silver behenate diffraction rings).

Three different in situ thermo-mechanical treatments were performed while recording the SAXS intensity:

- 0MPa sample: no stress applied + 1h temperature ramp from room temperature to 200°C followed by 1h at 200°C
- 50MPa sample: 50MPa stress applied + 1h temperature ramp from room temperature to 200°C followed by 1h at 200°C
- 100MPa sample: 100MPa stress applied + 1h temperature ramp from room temperature to 200°C followed by 1h at 200°C

The amounts of stress were chosen to remain within the elastic range of the as quenched specimen. This has been checked by observing the tensile specimen before and after the experiments to ensure that no plastic deformation occurred.

Each SAXS measurement consisted in 5 frames of 100s exposure time so that around 8-9 minutes elapsed between two measurements. The first SAXS image taken at room temperature is used as the background image, so that only clusters or precipitates originating from the thermo-mechanical treatments are responsible for the recorded signal.

3. Results and discussion

A few snapshots of the SAXS sequence are given on Fig. 2, Fig. 3 and Fig. 4 for the 0MPa sample, the 50MPA sample and the 100MPa sample, respectively. In all three samples, two distinct precipitation phases can be clearly distinguished. At early stages, one notices the appearance of a weak isotropic signal, which increases in intensity. Eventually, this signal is replaced by strong intensity streaks.

The streaking originates from the plate-like Ω precipitates (Al_2Cu). A flat precipitate scatters an intensity that is concentrated in the direction perpendicular to its habit plane (see e.g. [5]). However, in order to be able to observe this streak in intensity, the sample must be either a single crystal or strongly textured. It is very likely that the samples have a remaining rolling texture that is pronounced enough to give rise to this streaking.

The early isotropic signal is most likely to be due to the Mg and Ag rich clusters observed in atom probe tomography experiments by Reich et al. [2].

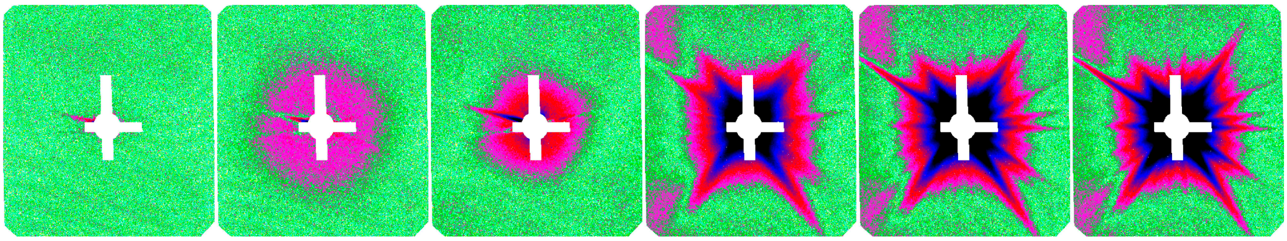

Fig. 2. Snapshots of SAXS sequence for the 0MPa sample. These snapshots are separated by approximately 20min. First, anisotropic clusters are forming followed by anisotropic precipitates.

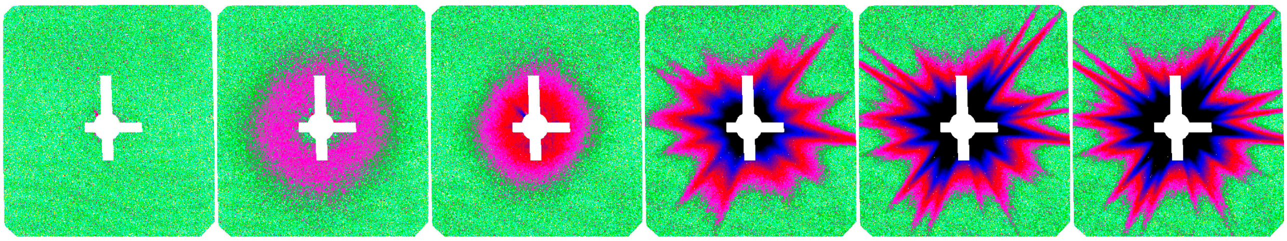

Fig. 3. Snapshots of SAXS sequence for the 50MPa sample. These snapshots are separated by approximately 20min. First, anisotropic clusters are forming followed by anisotropic precipitates.

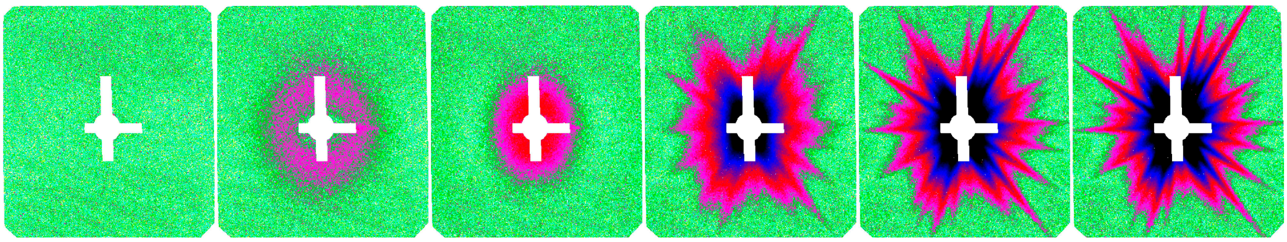

Fig. 4. Snapshots of SAXS sequence for the 100MPa sample. These snapshots are separated by approximately 20min. Notice the anisotropic shape of the diffuse spheroidal clusters (snapshots 2 and 3).

We will use azimuthally averaged signal I(q), were $q = \frac{4\pi}{\lambda}\sin\theta$ is the scattering vector and 2θ is the scattering vector. This is in principle valid only for isotropic signal, however, although the sample seems to be strongly textured, the intensity is nevertheless sufficiently spread in all direction to be able to use this approximation. This method is further validated by the behavior of the scattered intensity for images presenting streaks (Fig. 5). Indeed, the intensity (on a double log plot) is shown to follow a classical Porod law in q^{-4} in the high q region, whereas the low q region follows a q^{-2} evolution. This is a classical behavior for flat objects, which justifies the use of the azimuthally averaged intensity.

We can then assume that the intensity is in the form (see [6], pp 17-51):

$$I(q) = A\frac{2\pi}{q^2}(\Delta\rho)^2 t^2 \exp\left(-q^2\frac{t^2}{12}\right). \quad (1)$$

where A is the area of the particles, $\Delta\rho$, their electronic contrast with the aluminium matrix and t, their thickness. A Guinier-type plot Log(Iq2) vs. q2 can then be used to extract the thickness t of the precipitates. Figure 6 is an example of such a plot. It clearly shows a linear part that can be used to fit a Guinier law to extract the thickness.

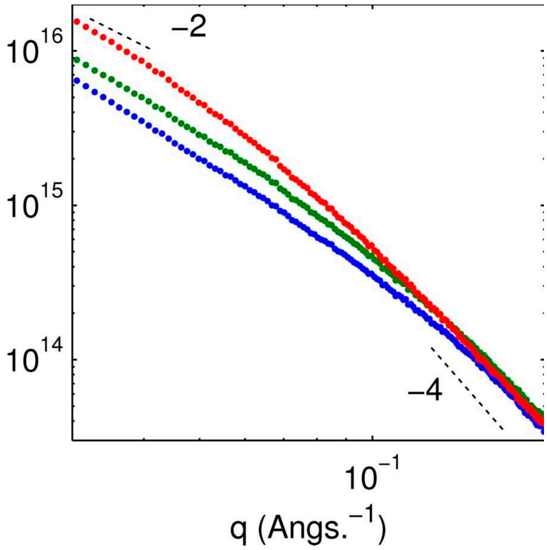

Fig. 5. Log(I) vs. Log(q) for 3 successive SAXS images of the 0MPa sample. This is a classical behaviour for flat precipitates.

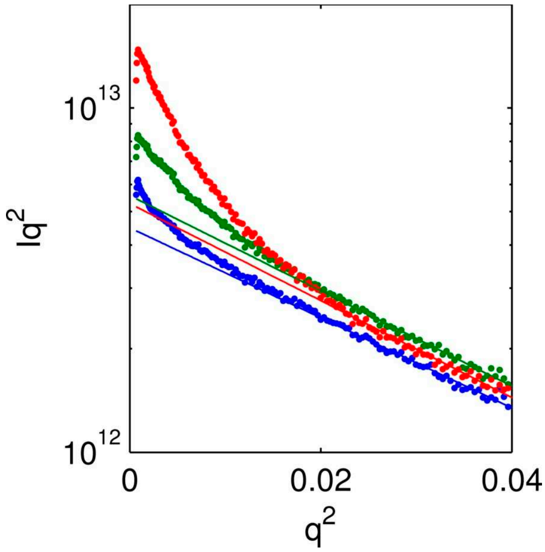

Fig. 6. Guinier type plot Log(Iq^2) vs. q^2 for the same images as Fig. 5. The slope of the linear part of the curve gives the thickness of the precipitates

For the spheroidal clusters, a simple Guinier plot, Log(I) vs. q^2, is enough to extract the radius of the clusters. The results as a function of time for the three different samples are shown of Fig. 7.

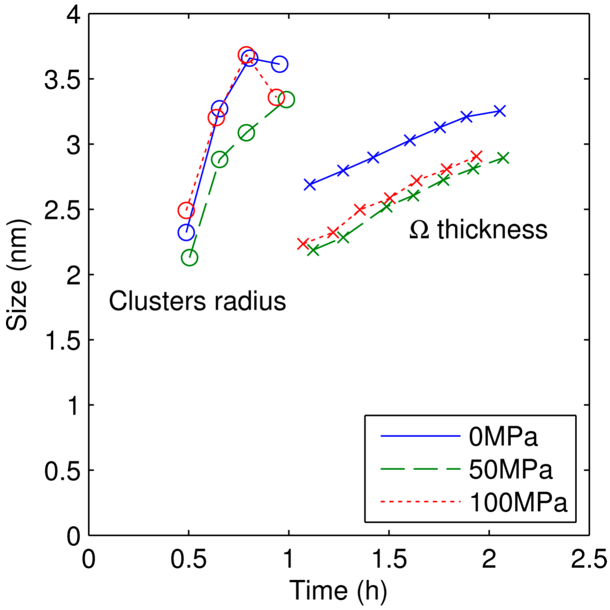

Fig. 7. Evolution of the clusters- and precipitates size as a function of ageing time for the three samples.

The clusters are first growing, before giving way to the Ω precipitates. No significant effect of the applied stress is observed on the clusters. It seems that an applied stress delays the thickening of the Ω plates, but the effect is rather subtle and may lay in the experiment uncertainty.

On the other hand, a careful observation of the scattering from the clusters in the 100MPa sample shows that there is a certain degree of anisotropy. This can be seen on the second and third snapshot of Fig. 4. To further highlight this effect, we have plotted for one SAXS image (the third snapshot of Fig. 4) the intensity as a function of the azimuthal angle for three different values of q (i.e. for three different distances from the centre).

This is shown on Fig. 8. The lines are sine fits to the data points. The lower intensity regions correspond to 90° and 270° which are the tensile directions. The higher intensity regions correspond to the direction perpendicular to the tensile direction.

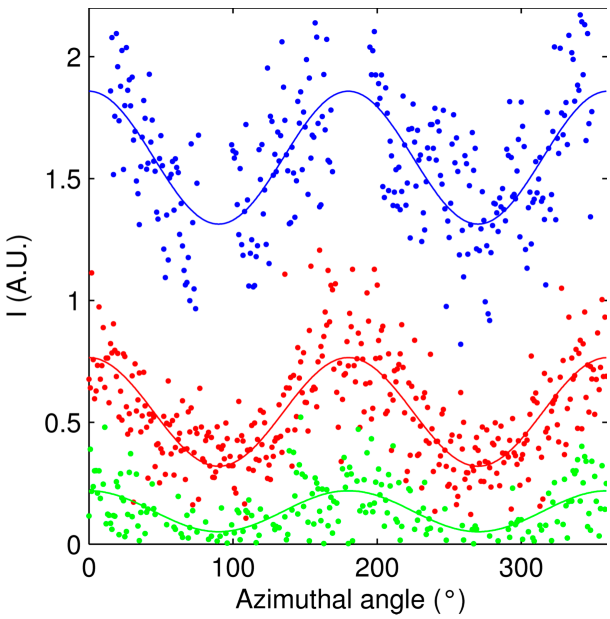

Fig. 8. Intensity as a function of the azimuthal angle for the clusters in the 100MPa sample. Three different distances from the centre are shown. The lines are sine fit to the data points.

This anisotropic signal can be explained by ellipsoidal clusters that would be either flat with a habit plane parallel to the tensile direction of elongated along the tensile direction. The aspect ratio of the SAXS signal gives an estimation of the aspect ratio of the clusters which is between 1.5 and 2.

4. Conclusion

We have assessed the possibility of SAXS to record in situ formation of the Ω phase and its precursors Mg- and Ag- rich clusters in Al-Cu-Mg-Ag. We show that these clusters grow in size before giving way to the Ω precipitates. We have attempted to show an effect of elastic stress on the precipitation of Ω Al_2Cu by comparing our results on samples subjected to 3 different levels of stress, 0MPa, 50MPA and 100MPa. The effects on Ω, if any, are shown to be very subtle. On the atomic clusters, on the other hand, there is a clear effect on the shape of the objects. When subjected to a high enough applied stress, the clusters are either flat, but more likely elongated in the tensile direction.

More extensive experiments are being performed. In particular, transmission electron microscopy is being done to record the shape of the objects and atom probe tomography experiments are planned to assess the effect of the applied stress on the segregation of Mg an Ag at the Ω/matrix interface.

Acknowledgements

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