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On chemical order and interfacial segregation in γ' ($AlAg_2$) precipitates

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Abstract

A detailed study has been carried out on γ' ($AlAg_2$) precipitates in Al–Ag and Al–Ag–Cu alloys to reconcile the conflicting reports on chemical ordering and stacking faults in this phase. High-angle annular dark-field scanning transmission electron microscopy and convergent beam electron diffraction show no indication of chemical ordering on alternate basal planes of γ' precipitates in alloys aged at 473 K for 2–23 h. Precipitates were visible as Ag-rich regions with 1–13 face-centred cubic (fcc) → hexagonal close-packed stacking faults, corresponding to γ' platelets with thicknesses ranging from 0.69 to 6.44 nm. There were no systematically absent thicknesses. Growth ledges with a riser height equal to the *c*-lattice parameter (0.46 nm) were directly observed for the first time. Genuine stacking faults within the precipitates were extremely rare and only observed in thicker precipitates. In precipitates with 1–3 stacking faults there was also substantial Ag in the surrounding fcc layers of the matrix, indicating that Ag strongly segregated to the broad, planar precipitate–matrix interfaces. This segregation is responsible for previous reports of stacking faults in γ' precipitates. The results indicate that the early stages of γ' precipitate growth are interfacially controlled.
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Keywords: Aluminium alloys; Long-range order; Interface segregation; Stacking faults; Ledgewise growth

1. Introduction

The γ' ($AlAg_2$) phase is a well-studied intermetallic phase which precipitates in Al alloys and has been used as a model system to study nucleation [1–5], ledgewise growth [6–10], and interface structure and energetics [11–13]. Despite this extensive body of work, questions remain regarding the existence or extent of chemical ordering and the nature of stacking faults widely reported in this phase [14–17].

The γ' phase can be produced by quenching an Ag-containing Al alloy from high temperatures to produce a supersaturated solid solution. Heating this solid solution to an intermediate temperature (a process termed "ageing") results in the gradual precipitation of plate-shaped γ' precipitates on the {1 1 1} planes of the Al matrix.

The ground state of the γ' phase is a hexagonal close-packed (hcp) structure with composition $AlAg_2$ and short-range order (SRO) on the basal planes [18,19]. Experimental reports indicate that, in practice, the γ' phase departs considerably from stoichiometry. An atom probe field ion microscopy study by Osamura et al. in Al–5.72 at.% Ag aged at 436 K reported an Ag concentration of 33.3 ± 1.5 at.% [20] and more recent studies using energy-dispersive X-ray analysis in Al–22 at.% Ag found an average composition of 42 at.% Ag [8].

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X-ray studies to date have also failed to find evidence of SRO [18,21] within the basal plane. Monte Carlo simulation studies have suggested an order–disorder transition temperature in the range of 45–100 K [18,21]. Such temperatures are well below the ageing temperatures required to form γ' precipitates in a finite time scale, so this might explain the absence of SRO.

There have also been conflicting reports of chemical ordering on alternate basal planes (i.e. long-range order) in γ' precipitates. Although early X-ray studies proposed a ground state with alternate layers of γ' precipitates having compositions of Al_2Ag and $AlAg_2$, these studies found compositional variations between alternate planes of at most 1.5 at.% [18,21]. A structure consisting of layers with compositions Ag and Al_2Ag has also been suggested based on high-resolution transmission electron microscopy (HRTEM) studies [6,7] that detected high and low contrast layers [11] on alternate basal planes.

Clarifying the existence and/or extent of ordering in γ' is important for models of the structure and growth of this phase. Recent density functional theory (DFT) simulations have modelled the structure as having well-defined order on alternate basal planes [2,5,22] and may require revision to represent the structure at non-zero temperatures[2] if the phase is disordered.

A further area of uncertainty regards stacking faults, which have been widely reported in γ' precipitates. The earliest reports of such defects were by Guinier [14,15], who reported stacking faults with an average spacing of only 10 Å based on small-angle X-ray scattering. Similar results were obtained via electron diffraction by Nicholson and Nutting [16], who also used the width of selected area diffraction peaks to determine the fault spacing as a function of precipitate thickness. These authors reported similar fault spacings to Guinier [14,15] and Borchers [17] for thin precipitates, and an increase in the spacing as the precipitate thickness increased to 80 Å. Despite this, HRTEM studies on thin (1–3 unit cell) precipitates did not show abundant stacking faults within γ' precipitates [3]. The discrepancies between diffraction and imaging studies have not been resolved.

The present work sets out to re-examine the structure of γ' precipitates using high-resolution scanning transmission electron microscopy. The absence of complicated phase contrast in high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM), together with the insensitivity of this technique to strain contrast, make it ideal for detecting chemical ordering. These capabilities are also well suited for examining the structure of the precipitate in order to detect and characterize any stacking faults.

2. Experimental details

Aluminium alloys with compositions Al–1.68 at.% Ag (Al–6.4 wt.% Ag) and Al–0.90 at.% Cu–0.90 at.% Ag (Al-3.4 wt.% Ag-2.0 wt.% Cu) prepared from pure elements were used in this study. Billets of each alloy were cast in air at 973 K and poured into graphite-coated steel molds. The compositions and impurity levels were measured by inductively coupled plasma atomic emission spectrometry. Impurity levels were low, with Fe present at 0.025 wt.% and Si at 0.01 wt.%. The Ni content was 0.01 wt.%, with other elements (Cu, Zr, Ti, Mn, Mg) present at levels of <0.005 wt.%.

Billets were homogenized (798 °C, 168 h), then hot-rolled (to 2 mm thickness) and cold-rolled to produce a 0.5 mm thickness sheet. Discs (3 mm diameter) punched from the sheets were solution-treated (525 °C, 0.5 h) in a nitrate/nitrite salt pot and then quenched to room temperature in water.

Solution-treated samples of Al–Ag alloy were aged in an oil bath 473 K for 2–23 h and quenched into water. Al–Ag–Cu alloys were aged for 2–4 h under the same conditions. The aged discs were mechanically thinned and then twin-jet polished to perforation using a nitric acid/methanol solution (~13 V, 253 K in 33% HNO_3/67% CH_3OH v/v). Discs were plasma-cleaned immediately prior to examination.

Foils were examined using an FEI Titan[3] 80–300 microscope operating at 300 kV. HAADF-STEM images were obtained using a convergence semi-angle of 15 mrad, providing a spatial resolution of ≈ 1.2 Å, using an inner collection semi-angle of 40 mrad.

The HAADF-STEM images were compared with images calculated by a multi-slice method using a frozen phonon approach to incorporate thermal diffuse scattering [23]. The simulations used the γ' structure proposed by Neumann, with a slice thickness of 1.4 Å and a maximum thickness of 700 Å. Atomic sites in the simulated structures were randomly occupied within each layer, and alternate layers had compositions of Ag_2Al and $AgAl_2$. These were compared with simulations in which all layers had identical compositions.

Image analysis was carried out using ImageJ software (version 3.6322U2011). Simulated images were scaled linearly to the same dynamic range as the HAADF-STEM images. The contrast in simulated images was then adjusted to match the contrast of the experimental images by modifying the gamma correction value (Γ). The adjusted intensity (I') was given by $I' = I^{1/\Gamma}$. Except where otherwise stated, all experimental and simulated images presented were adjusted for brightness and contrast in this manner, with no other manipulations.

Convergent beam electron diffraction (CBED) was performed in STEM mode using a JEOL 2100F instrument operating at 200 kV, with a 0.5 nm probe. CBED maps were obtained using the Diffraction Imaging plug-in of Gatan DigitalMicrograph software on foils aged for 23 h at 473 K.

3. Results

3.1. HAADF-STEM imaging

The γ' precipitates are readily visible in HAADF-STEM images, due to the strong atomic contrast of Ag compared

[2] DFT calculations are carried out at 0 K.

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