Quantification of the Influence of Increased Pre-Stretching on Microstructure-Strength Relationships in the Al-Cu-Li Alloy AA2195

DOI: 10.1016/j.actamat.2016.02.017

Document Version
Accepted author manuscript

Citation for published version (APA):

Published in:
Acta Materialia

Citing this paper
Please note that where the full-text provided on Manchester Research Explorer is the Author Accepted Manuscript or Proof version this may differ from the final Published version. If citing, it is advised that you check and use the publisher's definitive version.

General rights
Copyright and moral rights for the publications made accessible in the Research Explorer are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

Takedown policy
If you believe that this document breaches copyright please refer to the University of Manchester’s Takedown Procedures [http://man.ac.uk/04Y6Bo] or contact uml.scholarlycommunications@manchester.ac.uk providing relevant details, so we can investigate your claim.
Quantification of the Influence of Extreme Stretching on Microstructure-Strength Relationships in the Al-Cu-Li Alloy AA2195

B.I. Rodgers\textsuperscript{a} and P.B. Prangnell\textsuperscript{b}

The University of Manchester, Manchester, M13 9PL, UK

\textsuperscript{a}benjamin.rodgers@postgrad.manchester.ac.uk; \textsuperscript{b}philip.prangnell@manchester.ac.uk

Abstract

The effect of increasing pre-stretching to much higher levels, than are currently used in industrial practice, has been investigated on the strength, microstructure, and precipitation kinetics seen during artificial ageing an Al-Cu-Li alloy AA2195 - focusing on the behaviour of the main strengthening phase, $T_1$. Increasing the pre-strain level, to the maximum obtainable before plastic instability (15%), resulted in an increase in the T8 yield strength to $\sim 670$ MPa without a loss of useable ductility. Microstructure data have been used to deconvolute and model the effects of increasing pre-strain on the main strengthening components that contribute to this large strength increase. The precipitation strengthening model proposed by Dorin et al. [1] has been successfully employed to calculate the strengthening contribution of the $T_1$ phase and the increase in strength due to strain hardening has been modelled using X-ray line broadening measurements of dislocation density. A nearly complete absence of recovery was observed during stretching, or artificial aging, which suggests strong solute dislocation interactions occur within this material. It is shown that increasing the pre-strain prior to ageing resulted in a reduction in the strengthening provided by the $T_1$ phase, due to precipitate refinement, in favour of an increase in the strain hardening contribution. As the pre-strain was increased a transition was thus seen, in terms of the main strengthening component, from precipitation hardening to strain hardening.

Keywords: Aluminium; $T_1$ Phase; Precipitation; Work Hardening; Strengthening
1. Introduction

Third generation (Gen3) aluminium-lithium (Al-Li) alloys [2] are of great interest to the aerospace industry owing to a number of key benefits they offer over conventional aluminium (Al) alloys [3–5]. In particular, their lower density, excellent corrosion resistance, and combination of higher fatigue performance, high strength and toughness, can lead to significant weight savings. They also offer a cost advantage over CFRPs (Carbon Fibre Reinforced Polymers) which has led to Gen3 Al-Li alloys being increasingly substituted for conventional 2xxx and 7xxx series materials in new aircraft designs [6].

Previous generations of aluminium-lithium alloys contained a higher concentration of lithium (Li) and had a lower density than the new Gen3 alloys. However, these earlier alloys suffered from high anisotropy, lower toughness, and manufacturing issues, associated with the high Li level and precipitation of the metastable $\delta'$ and coarser equilibrium Li containing phases, such as $T_2$ [5,7,8]. New Gen3 alloys typically contain lower Li levels of 1 - 1.8 wt.%, which suppresses $\delta'$ formation, and have chemistries designed to promote $T_1$ as the dominant strengthening phase. These developments have proved to be extremely effective in providing high strength without the deleterious effects seen in the previous generation [1,9–11].

The conventional manufacturing route for aluminium aerospace plate involves a stretching operation after solution heat treatment, to relieve the large residual stresses developed on quenching, during which the material is typically plastically strained between 2 - 5% [12]. In Gen3 alloys this stretching operation is also critical to obtain an optimum distribution of $T_1$ precipitates, which are dislocation nucleated. It has been widely shown that a small pre-strain prior to artificial aging produces a uniform distribution of dislocations within the matrix, which act as heterogeneous nucleation sites for the $T_1$ phase [9–11]. This results in the nucleation of a fine homogeneous distribution of the $T_1$ phase throughout the material during artificial ageing, leading to exceptional mechanical performance [5,9,13,14]. Stretching also serves to accelerate matrix precipitation and thus avoid competition with grain boundary precipitation, which leads to a detrimental effect on toughness [15]. The exact nucleation mechanism is still under debate, but is known to involve prior segregation of Cu and Mg to dislocation lines [16].
The $T_1$ ($Al_2CuLi$) phase has been extensively studied and is known to form as very thin semi-coherent hexagonal plates with a $\{111\}_{Al}$ matrix habit plane [17–19]. $T_1$ precipitates have a particularly high aspect ratio and provide a greater hardening effect than the $\theta'$ phase, which forms as octagonal plates on $\{100\}_{Al}$ planes [9,20]. As it is dislocation nucleated, increasing the pre-strain increases the density of $T_1$ nucleation sites and the resultant finer precipitate distribution reduces the average diffusion field size, so the matrix is depleted of solute in a shorter timescale. This results in accelerated ageing kinetics with increasing values of pre-strain. However, the benefits of stretching have been widely reported to saturate at pre-strains of around 6 – 9% in Al-Li-Cu alloys [9,10].

The $T_1$ plates were originally thought to be shear resistant [21]. In contrast, recent research by Deschamps et al. [1,22] has demonstrated that $T_1$ precipitates are cut by dislocations during plastic deformation, across the entire range of conventional ageing treatments, as the plates are very thin (~ 1.3 nm) and their thickness is very stable with ageing time at temperatures below ~ 170 °C [22–24]. Dorin et al. have proposed a strengthening model for the $T_1$ phase, based on earlier work of Nie and Muddle [25], that considers the interfacial and stacking fault energy contributions to a $\{111\}_{Al}$ habit plane, plate-shaped, precipitate’s shear resistance. This model has been demonstrated to be able to reliably predict the yield strength contribution of the $T_1$ phase in a 2198 alloy for a wide range of plate dimensions and densities [1]. However, in this work, although it was reported that after ageing the alloy’s yield stress was relatively constant for pre-strains greater than about 2% [1], the contribution of dislocation forest strengthening to the alloy’s yield stress was not explicitly measured. It was also proposed that after recovery the residual strain hardening caused by pre-stretching compensates for a predicted reduction in precipitate strengthening, due to the reduced plate diameter that results from an increase in precipitate density with rising levels of pre-strain, and there is thus a coincidental linear correlation between strength and particle volume fraction in this alloy system.

Recent developments in rolling control technology, can enable the production of plates with variable thickness that offer potential benefits in the production of nearer-to-net-shape sections; for example in the manufacture of tapered wing skins [26]. One significant implication of applying this processing technology is the subsequent impact on the required stretching operation. The stretching of a tapered plate results in a strain gradient and for Al-
Li-Cu alloys the maximum strain that can be achieved without tensile fracture limits the taper that can be utilised without omitting the critical stretching step [26]. It is therefore important to better understand the impact of increasing plastic pre-strains, to near the plastic limit, on the peak-aged microstructure found in Gen3 plates and ultimately how this impacts on the materials peak yield strength and other mechanical properties.

While much prior work has been carried out to investigate the effects of increasing pre-strain on precipitation strengthening in Al-Li alloys within standard limits, there has been less published on the effects of more extreme stretching operations. The work presented here thus aims to examine the effect of increasing tensile pre-strains, to near the tensile plastic limit, on a typical Gen3 Al-Li alloy’s aging kinetics, microstructure and yield strength. It is also considered that, according to classical theory, more extreme stretching should perhaps lead to dynamic recovery and the formation of a dislocation cell structures [27]. This, in turn, could potentially cause microstructural heterogeneity by affecting the distribution of T1 phase. The effect of thermal treatment during artificial ageing, on reducing the strain-hardening contribution to the materials strength by dislocation recovery, is also currently largely unknown. Ultimately, this could have a profound impact on the mechanical properties of the alloy and how the different strength contributions relate to the overall peak yield strength of the material.

To this end, tensile tests, hardness testing and Differential Scanning Calorimetry (DSC) have been employed to explore the effect of extreme tensile pre-straining (i.e. above 6%) on a AA2195 alloy’s aging kinetics and yield stress, while simultaneously utilising an X-ray diffraction (XRD) peak broadening analysis to measure the effect on the residual dislocation density. High resolution electron microscopy has also been used to determine the effect of the pre-strain on the dislocation structures and to quantify the dimensions and distribution of the T1 phase. This data has then been utilised to model the different strengthening contributions that contribute to the alloy’s measured yield strength, as a function of the level of pre-strain applied prior to artificial ageing.
2. Experimental Method

A typical Gen3 Al-Cu-Li alloy, AA2195, provided by Constellium Voreppe Research Centre in France, was used in this investigation. The alloy was supplied as 22 mm thick plate in a T841 temper. The composition range of AA2195 is provided in Table 1.

<table>
<thead>
<tr>
<th></th>
<th>Cu</th>
<th>Li</th>
<th>Mg</th>
<th>Zr</th>
<th>Mn</th>
<th>Ag</th>
<th>Al</th>
</tr>
</thead>
<tbody>
<tr>
<td>Min</td>
<td>3.70</td>
<td>0.80</td>
<td>0.25</td>
<td>0.08</td>
<td>-</td>
<td>0.25</td>
<td>Bal</td>
</tr>
<tr>
<td>Max</td>
<td>4.30</td>
<td>1.20</td>
<td>0.80</td>
<td>0.16</td>
<td>0.25</td>
<td>0.60</td>
<td>Bal</td>
</tr>
</tbody>
</table>

Tensile samples were cut from the ¼ plate depth and machined in accordance with BS EN ISO 6892-1:2009 [28]. Each test sample was given a solution heat treatment (1 hour at 510°C) and water quenched. The samples were pre-stretched, by tensile stretching, to plastic strain values ranging from 3 to 15%. The upper limit of 15% was selected after analysis of the work hardening rate in the solutionised condition, which indicated that this was the maximum strain that could be reliably used while avoiding plastic instability.

Artificial aging was subsequently performed with an initial heating ramp of 25°C per hour, followed by an isothermal hold at 150°C for a range of times up to 100 hours. Pre-stretching and tensile testing was carried out using an MTS Alliance RT/100 tensile machine at a strain rate of 2mm/min, with the strain being monitored by a 25mm clip gauge extensometer. The required plastic pre-strain values were produced by elongating the tensile samples in the solution heat treated (SHT) condition. Following artificial ageing, the pre-stretched tensile samples were tested to failure to measure the effect of pre-stretch on the material’s tensile properties.

The effect of pre-strain on the aging kinetics was first determined by measuring hardness curves for samples with varying degrees of pre-strain, ranging from 3 to 15%, using an Instron RT2100 machine with a V044 indenter and a 0.5Kg load. An average of five measurements was taken for each condition. Differential Scanning Calorimetry (DSC) analysis was also performed, using a Netzsch – STA 449C Jupiter instrument, on samples subjected to varying degrees of pre-strain and at different stages through their artificial ageing treatment. This enabled the effect of pre-strain on the relative volume fraction of the T₁ phase to be determined from the integral of the T₁ precipitation exothermic peak in the
DSC curves, following the method proposed by Dorin et al. [29]. The DSC samples were prepared by cutting slices from the gauge length of stretched tensile specimens using a Struers Minitom with a diamond cutting wheel. A baseline was measured using an annealed sample of pure aluminium and subsequently subtracted from the results obtained from each test sample. The DSC scans were carried out at a heating rate of 20°C/min from 20 - 510°C.

TEM samples were prepared by twin-jet electropolishing using a solution of 80% Methanol and 20% Nitric Acid at -30°C and 14V. Transmission Electron Microscopy (TEM) was used to observe the effect of extreme stretching on the dislocation structure using a Tecnai G2 T20 microscope operating at 200kV. Scanning TEM (STEM), using a High Angle Annular Dark Field (HAADF) detector, was used to image the T1 precipitates’ size and spatial distribution. STEM imaging was conducted using an FEI Titan 80-200 FEG-TEM fitted with dual CEOS aberration correctors.

X-Ray Diffraction (XRD) was used to determine the effect of pre-strain on the dislocation density by the line broadening method [30]. Measurements were taken using a Bruker D8 Discover diffractometer equipped with a VANTEC-500 detector. Data was collected using a monochromatic Co Kα radiation (λ = 0.17902nm) with a 0.6mm divergent slit, 35kV and 40 mA over a 2θ range of 40-100°. The diffraction peaks were fitted using a Rietveld refinement.

3. Results
3.1 Effect of Pre-strain on the Aging Kinetics
3.1.1 Age Hardening Curves
In Figure 1 age hardening curves are compared, as a function of increasing pre-stretch up to 15%, on heat treating the 2195 alloy at 150°C for up to 100 hours. These results demonstrate that increasing the pre-strain increases the hardness of the material both in the T351 and T8 tempers, but the effect appears less pronounced after artificial ageing. Figure 1 (a) reveals a similar level of softening occurring for each pre-stretch condition, during the initial heating ramp, owing to the reversion of solute clusters produced by natural ageing [20] and an expected increase in ageing kinetics with increasing level of pre-strain [9,10]. However, it can be seen from Figure 1 that in contrast to some previous
reports (e.g. [10]) the aging kinetics and peak hardness achieved continues to increase into the more extreme stretching regime, albeit at a reducing rate. It can further be seen from Figure 1 (b) that the peak hardness of the pre-strained samples all exhibited a level maximum plateau that remained stable when they were artificially aged for up to 100 hours.

![Aging Hardness Curves](image)

**Figure 1. Age hardening curves; (a) early stage aging (b) prolonged artificial ageing.**

### 3.1.2 Volume Fraction of the T₁ Phase during Ageing

Integration of DSC curves has been used to track the relative volume fraction of the T₁ phase during artificial ageing. As outlined by Dorin et al. [29], this is a reliable approach in this alloy because there is a clearly defined exothermic peak in the DSC curves and when stretched the T₁ phase dominates the precipitate sequence. An example data set is provided in Figure 2 (a) for samples subjected to a 3% pre-strain and aged at 150°C for increasing times, between 0h (end of the heating ramp) and 22h (peak aged). Just below 300°C an exothermic peak can be seen caused by precipitation of the T₁ phase, followed by a broad dissolution peak. As the samples undergo artificial ageing the peak area can be seen to diminish, due to the increasing volume fraction of T₁ precipitates present prior to performing the DSC scan.
Normalisation of the integrated area of the T₁ exothermic peak in each sample, relative to the zero hour case for the same pre-stretch, can therefore be used to determine the relative volume fraction of the T₁ phase at a given ageing time. Figure 2 (b) shows the corresponding evolution of the relative volume fraction of the T₁ phase, determined by this approach, in the 3% stretched example. The same analysis was subsequently repeated for all the samples with pre-strain values ranging from 3 to 15% (see Figure 3). In each case the experimental points were fitted using the Avrami (JMAK) law [31] given by:

\[ f_V = 1 - \exp(-Kt^n) \]

Equation 1

Where; \( f_V \) is the transformed volume fraction, \( t \) is the aging time and \( K \) and \( n \) are constants.

![Figure 2. (a) Example DSC curves for the 3% pre-strain condition, with increasing aging times at 150°C, showing the T₁ precipitation exotherm in the temperature range 200 – 300 °C and (b) the evolution of the relative volume fraction of the T₁ phase as a function of aging time, determined by integration of the precipitation exotherm peak area.](image-url)
Figure 3. Fitted JMAK curves showing the evolution of the relative volume fractions of the \( T_1 \) phase, as a function of aging time at 150\(^{\circ}\)C, with pre-strain values ranging from 3\% to 15\%.

In Figure 3, JMAK curves, fitted to DSC data following the procedure described above, have been plotted on the same graph for increasing levels of pre-strain. These curves again demonstrate an increase in the aging kinetics with rising levels of pre-strain. In agreement with the hardness curves in Figure 1, it can be seen that the rate of acceleration diminishes as the pre-strain value increases into the extreme stretching regime and for pre-strain values above 10\% there is little difference in the curves.

3.2 Effect of Pre-strain on Microstructure

3.2.1 Dislocation Structures

Figure 4. Bright field TEM Images showing the uniform dislocation density seen in the AA2195 alloy following pre-strains of (a) 3\%, (b) 9\% and (c) 15\%.
The images in Figure 4 show the high dislocation densities seen in the AA2195 alloy in the T351 temper, in regions of similar thickness, after applying pre-strains of 3, 9 and 15%. At this magnification the images reveal a uniform, but increasing, density of dislocations in dense forests throughout the aluminium matrix with rising levels of pre-strain. Both high angular resolution EBSD and systematic tilting were used on the samples with the highest pre-strain, to investigate the possibility of the formation of cell structures. In Figure 5 a bright field tilt series is provided at a lower magnification from the sample with the highest pre-strain of 15%. This again shows a uniform dislocation density and that there is little evidence of cell formation. Although some recovery would be expected in pure aluminium at this strain level [32], it can be noted that in the alloy studied this appears to have been retarded. The contrast change seen in the tilt series is, however, indicative of small systematic variations in crystallographic orientation owing to the formation of very diffuse micro shear bands. Overall, the dislocation structures are similar to those found in high Mg content Al alloys at this strain, where solute has a strong effect on inhibiting recovery [33].
3.2.2 Dislocation Density Measurements

Because it is extremely difficult to reliably measure the high dislocation densities shown in Figure 5 by TEM after artificial ageing, when dislocations are masked by precipitate coherency contrast, x-ray diffraction peak-broadening analysis was utilised to complement the qualitative TEM observations. According to the work of Ungar [34,35], diffraction peak broadening can be reliably correlated to the dislocation density within a crystalline material when the dislocations are uniformly distributed in forests, as was the case in the alloy.

Figure 5. Bright field TEM two beam condition tilt series of the dislocation structure in AA2195 after a 15% Pre-Strain; (a) 0° (b) 2° (c) 4° (d) 6° tilt around [1\bar{1}0].
investigated here (Figure 4). A high resolution 2-theta scan of the \{111\} diffraction peak was therefore performed (using 0.015° 2θ steps) to measure the effect of increasing pre-strain on peak broadening, in both the pre-stretched and artificially aged conditions. A Gaussian function was subsequently fitted to each \{111\}_\text{Al} diffraction peak in order to calculate the dislocation density using the method described below. The \{111\}_\text{Al} reflection was chosen for the calculations as it provides a high intensity and good angular resolution for the peak width. Figure 6 shows examples of fitted diffraction peaks with increasing levels of pre-strain from samples in the T351 and T8 tempers.

![Figure 6. Fitted Diffraction Peaks (a) 3% Pre-Strain T351 (b) 3% Pre-Strain T8 (c) 15% Pre-Strain T351 (d) 15% Pre-Strain T8.](image)
The dislocation densities (ρ) were calculated from the integral breadth of the \{111\}_{Al} peak profile in reciprocal space, $B_s^*$ using the method proposed by Vermeulen et al. [30];

$$B_s^* = k^* g^* b^* \sqrt{\rho} \quad \text{Equation 2}$$

Where; $b^*$ represents the Burgers vector (0.286nm for Aluminium), $g^*$ is the length of the reciprocal lattice vector given by $\sqrt{h^2+k^2+l^2} / a$, $k^*$ is a dimensionless coefficient related to the distribution and the orientation of the dislocations, with respect to the diffraction vector, and can be taken as 1 for a random distribution, $B_s^*$ is calculated from the real space breadth ($B_s$) using;

$$B_s^* = \frac{B_s \cos \theta}{\lambda} \quad \text{Equation 3}$$

Where; $\lambda$ is the wavelength of the X-Ray beam and $B_s$ is the real space integral breadth given by Equation 4.

$$B_s^{3/2} = B_m^{3/2} - B_i^{3/2} \quad \text{Equation 4}$$

Where; $B_m$ is the measured breadth of the aluminium peak and $B_i$ is the contribution of instrumental broadening to the total peak width, measured here using a Corundum standard.

The dislocation densities calculated by this method are plotted against the level of pre-strain for both the T351 and T8 aged samples in Figure 7. The results reveal a linear increase in dislocation density with increasing pre-strain, which demonstrates an absence of recovery during pre-straining at room temperature to remarkably high levels (15%). Furthermore, perhaps more surprisingly, Figure 7 also shows hardly any apparent change in the dislocation density, as interpreted from the line broadening, after artificial ageing. Overall, these results can therefore be interpreted as demonstrating an almost entire absence of static recovery during the artificial aging heat treatment of $150^\circ C$ for 22 hours.
Figure 7. Line broadening dislocation density measurements, showing the estimated dislocation density as a function of pre-strain for samples in both the T351 and T8 (22 hour at 150°C) Temper.

3.2.3 Distribution and Size of the T₁ Phase

STEM-HAADF images of the T₁ phase seen in the peak aged samples, with increasing levels of pre-strain, are shown in Figure 8 taken close to a <110>₂₃ zone axis. Qualitatively, it is apparent from these images that there is a reduction in the T₁ plate diameter with the level of prior plastic strain. Further investigations were conducted using STEM-HAADF imaging in order to remove unwanted diffraction contrast and enable more accurate measurements of the T₁ plate dimensions. A low magnification image is provided using this technique in Figure 9, to illustrate the exceptionally uniform distribution of the T₁ phase that was still seen in the sample subjected to the highest pre-strain level of 15%. Higher magnification images in Figure 10, confirm the thin dimensions of the T₁ phase and show the presence of only a minor fraction of θ' precipitates within the aged material.

Measurements of the T₁ plates’ size distributions were made manually from STEM-HAADF images using ImageJ image processing software with increasing pre-strain (Figure 11 (a-c)). These results demonstrate a continued decrease in the average plate diameter in the T8 condition with increasing pre-strain up to the extreme value of 15%. They also clearly show a reduction in the width of the precipitate diameter frequency distributions with increasing pre-strain, suggesting an increase in the homogeneity of the distribution of the precipitates. In line with previous studies [1,9], High resolution TEM imaging further confirmed that the
thickness of the T₁ plates remained effectively constant at approximately 1.3nm, in all the samples examined (e.g. Figure 10 (a)).

(a)

![STEM HAADF images showing T1 precipitates seen in AA2195 in the T8 condition (close to the <110>Al zone axis) for (a) 3% and (b) 15% Pre-Strain.](image)

(b)

**Figure 8.** STEM HAADF images showing T1 precipitates seen in AA2195 in the T8 condition (close to the <110>Al zone axis) for (a) 3% and (b) 15% Pre-Strain.

**Figure 9.** STEM HAADF image showing the uniform distribution of the T₁ phase seen in the peak aged AA2195 alloy following a 15% pre-strain (<110>Al Zone Axis).
Figure 10. STEM HAADF images of (a) a single $T_1$ plate and (b) an example of a $\theta'$ precipitate within the peak aged AA2195 subjected to a pre-strain of 15% ($<110>$$_{Al}$ zone axis).
Figure 11. Evolution of the mean $T_1$ plate diameter and size distribution for three pre-strain values; (a) 3%, (b) 9%, and (c) 15%, measured from HAADF-TEM images. (d) Evolution of the average plate diameter and calculated number density with increasing pre-strain.

By assuming a constant peak volume fraction in the peak aged condition (Figure 3) [1,10] and a constant plate thickness the number density of the $T_1$ phase could be evaluated from [12];

$$N = \frac{4f_v}{\pi D^2}$$  

Equation 5

Where; $t$ is the average plate thickness, $D$ is the average plate diameter and $f_v$ is the volume fraction.
The resultant calculated precipitate number densities are plotted in Figure 11 (d) and demonstrate a corresponding increase in number density as the average precipitate diameter reduces with level of pre-strain.

In addition to the STEM observations, further information can be garnered from the fitted Avrami curves shown in Figure 3. It has been shown theoretically that the value of the Avrami exponent (Equation 1) can elucidate changes in the nucleation mechanism, or homogeneity and morphology of precipitate phases [36,37]. In Figure 12 a systematic reduction in the value of the exponent (n) was seen with increasing pre-strain. In keeping with the STEM observations, this behaviour can be attributed to an increase in the homogeneity of the size distribution of the $T_1$ phase with pre-strain.

![Figure 12. Change in the JMAK exponent (n) with increasing pre-strain.](image)

3.4 The Effect of Extreme Stretching on the T8 Yield Stress

The average mechanical properties (from 3 tests) obtained from tensile testing the peak aged, pre-stretched, samples are summarised in Figure 13. This reveals a significant increase in the peak yield stress of the material with increasing pre-strain, which continued into the higher pre-strain regime and with the greatest pre-stretch applied of 15% ultimately reached a maximum value of 668 MPa. However, this increase in strength also led to a modest decrease in strain to failure from $\varepsilon_f = 11\%$, for the conventional 3% level of pre-stretch, to $\varepsilon_f = 7.5\%$, with a 15% pre-stretch.
Figure 13. Yield Stress (0.2% proof) and ductility (plastic strain to failure) in the T8 Temper (22 hrs at 150°C), plotted as a function of pre-Strain.

4. Strength Modelling
Following the work of Shercliffe and Ashby [38] and other authors [39,40] the main strengthening terms of interest can be summed by the following simplified relationship;

\[ \sigma_y = M (\tau_B + \Delta\tau_p + \Delta\tau_P) \]  \hspace{1cm} \text{Equation 6}

Where; \( \sigma_y \) is the yield strength of the material, \( \tau_B \) is the constant base shear strength, \( \Delta\tau_p \), and, \( \Delta\tau_P \), are the increase in strength due to strain hardening and precipitation, respectively, and M is the Taylor factor (usually assumed to be about 3.1 for a material with a relatively weak texture [41,42]).

The base strength of the solution treated alloy prior to pre-straining and ageing will result from a combination of the intrinsic shear resistance of the lattice (\( \tau_i \)) and solid solution strengthening (\( \tau_{ss} \)). Recent research has demonstrated that a significant amount of Cu often remains in solution after aging [43,44]. Therefore, for the sake of simplicity, and following the approach of other authors [1,45,46] it is assumed that to a first approximation this term will remain constant during age hardening, as any change from a loss of solid solution strengthening will be relatively small compared to the high strength increase from precipitation [47].
As the XRD peak broadening results revealed an absence of significant recovery during artificial ageing, it is reasonable to assume that any increase in the yield strength resulting from strain hardening will be largely retained in the peak-aged material. The strain hardening contribution can thus be easily determined using the increase in yield stress seen in true stress-strain curve produced during tensile stretching of the T3 material. Any further increase in the yield stress in the T8 temper can then be attributed to precipitation of the main strengthening T₁ phase, which can be obtained by subtraction of the strain hardening contribution and base strength from the total measured yield strength. In the first instance, the two main strengthening contributions can hence be separated simply using the tensile-stress strain data, as shown in Figure 14. From this analysis it is apparent that the strengthening contribution from precipitation of the T₁ phase diminishes with increasing pre-strain and the contribution from strain hardening is higher than might be expected, owing to the observed lack of recovery. Indeed, with pre-strains greater than around 10% the strength increase from strain hardening becomes greater than that from precipitation. This preliminary interpretation will be further justified below by separately modelling the strain and precipitation hardening contributions.

![Figure 14](image.png)

*Figure 14. Effect of Pre-Strain on the main components contributing to the yield strength in the T8 temper, as a function of pre-strain.*
4.1 Calculation of the Strain and Precipitation Hardening Contributions

4.1.1 Strain hardening

In addition to the above approach, the measured dislocation density values obtained by XRD can be used to estimate the contribution from forest hardening to the overall yield strength of the material from standard expressions relating dislocation density to shear strength [12] given by:

\[ \Delta \sigma_p = M \alpha \mu b \rho^{1/2} \]  

Equation 7

Where; \( \alpha \) is a dimensionless constant between 0.2 – 0.5 [48], \( \mu \) is the shear modulus, \( b \) is the burgers vector (\( \approx 0.286 \text{nm for Al} \)), and \( \rho \) is the dislocation density.

When compared to the change in strength of the T351 samples with pre-strain, using the dislocation density data shown in Figure 7, Equation 7 gives very reasonable agreement with the experimentally determined yield stress, as can be seen from Figure 15 (a). It can further be seen in Figure 15 (c) that there is also very little difference between the predicted increase in yield stress from strain hardening, following artificial ageing, determined from the dislocation density measured by the XRD peak broadening.

4.1.2 Precipitation hardening

Recent literature has demonstrated the efficacy of a model originally developed by Nie and Muddle, [25] and subsequently validated by Dorin et al. [1], for predicting the strengthening contribution of the thin shearable T1 plates. The model is based on the assumption of (Friedel) statistical interactions between a dislocation and weak obstacles, where the obstacle strength is controlled by the increase in interfacial energy on shearing a precipitate. The thin disc geometry of the T1 precipitates, with respect to their \{111\}Al habit plane and the \langle011\rangleAl shear direction, is explicitly considered by Dorin et al. [1], who also included a stacking fault energy term, giving:

\[ \Delta \tau_p = \frac{1.211D_{eff}^{3/2}}{t^2} \sqrt{\frac{b f_e}{\Gamma}} \]  

Equation 8
Where; \( \Delta \tau_p \), is the increase in the critical resolved shear stress due to precipitation, \( D \), is the average precipitate diameter, \( t \) is the thickness, and \( \gamma_{eff} \) is an effective interfacial energy term that considers both the interfacial and stacking fault energy contributions associated with shearing a precipitate. The other terms have their usual meaning; \( b \) is the Burgers vector (\( \approx 0.286 \)nm for Al), \( f_v \) is the volume fraction of the T1 phase, and \( \Gamma \) is the dislocation line tension. It should be noted that this model is only applicable for T1 strengthened alloys in the under-aged and peak aged conditions and if substantial over-ageing occurs Orowan by-passing would need to be considered. However, for the conditions studied here, where the plates did not thicken during ageing, this would not be of concern [1].

In Figure 3 it is apparent that the volume fraction of the T1 phase is constant for ageing times in the peak aged plateau hardness region and it has also been shown to remain unchanged with increased levels of pre-strain, which does not affect the level of solute retained in solution at equilibrium [1,10]. Therefore, a representative value of volume fraction has been taken from the literature for an alloy with a similar composition of \( f_v = 3.2\% \) [1]. Using these parameters with equation 8 a value of \( \gamma_{eff} = 0.089 \)Jm\(^{-2}\) was found to give an excellent fit to the experimentally determined yield stress contribution, as can be seen in Fig. 15(b).

### 4.1.3 Total predicted strength increase

When the contributions calculated for strain and precipitation hardening are summed, using Equation 6, it can be seen from Figure 15 (c) that the predicted yield strength agrees extremely well with the effect of increasing levels of pre-strain on the yield stress measured in the tensile tests for the peak aged samples.
Figure 15. Comparison of the calculated contributions to yield strength with measured values for; (a) Strain hardening obtained from Equation 7, using dislocation densities measured by XRD and (b) Precipitation of the T₁ phase using Equation 8, with data measured by TEM. In (c) the predicted contributions are combined and compared to the effect of pre-strain on the measured tensile yield stress in the T8 temper.
5. Discussion

When viewed all together, the results reveal that increasing the level of pre-strain prior to ageing, to higher strain levels than are normally applied in industrial practice, has a more significant effect than formerly thought on the microstructure and strength of Gen3 Al-Li alloys. For example, previous work has commonly suggested that the microstructure refinement benefits of pre-stretching tend to saturate at strain levels of about 6% [9,10]. The DSC results used here, to track the volume fraction of precipitation during ageing (Figure 3), showed that acceleration of the precipitation kinetics continued for pre-stretching above 6 %, but at a rapidly diminishing rate, and then the kinetics did not change significantly for pre-strains above about 10%. However, there was still a continual increase in yield stress for greater pre-strains (Figure 13). For the alloy studied, in the T8 condition, this led to a remarkably high yield strength that approached 700MPa.

An important factor in the effect of pre-straining on the T1 phase, and its role in precipitation hardening, is clearly the influence this has on the dislocation nucleation site density in the material prior to artificial ageing. However, as will be discussed further below, strain hardening was found to have a larger direct effect than anticipated on the yield strength after artificial ageing, where significant recovery is usually expected during thermal treatment and strengthening is normally considered to be dominated by precipitation hardening.

The TEM images in Figures 4 and 5 show that in the AA2195 alloy a very uniform distribution of dislocations, in the form of dense tangled forests, was still seen at high pre-strain levels. In addition, perhaps more unexpected, was the fact that the XRD results indicated an almost complete absence of recovery, both during stretching and subsequent artificial ageing, across the full range of pre-strain values investigated. In the first case this can be inferred from a linear dependence of plastic strain on the calculated dislocation density (Figure 7) and, in the second, from the fact there was virtually no change in the integrated breadth of the Al matrix {111} diffraction peak profiles in the pre-stretched samples after the ageing heat treatment (Figure 6). Furthermore, when the level of strain hardening was calculated from the dislocation densities measured by XRD line broadening, which reached around $4 \times 10^{14}$ m$^{-2}$ with a pre strain of 15%, this was found to accurately predict the increase in yield stress seen after stretching (Figure 15 (a)). In addition, when combined with the
precipitation hardening model proposed by Dorin et al. [1], the same approach also gave good predictions for the strength of the material in the T8 temper (Figure 15 (c)). The TEM images, XRD data and modelling results are thus self-consistent with the supposition that very little recovery took place, either dynamically during stretching, or statically, during the thermal treatment involved in artificial ageing.

This lack of recovery is important in that it has a very significant influence on the material’s strength in the T8 temper at high pre-stretch levels and suggests strong solute interactions occur with dislocations in the alloy studied. Mg has long been known to strongly inhibit recovery in Al-alloys [27,49] and the images of the dislocation forests shown in Figures 4 and 5 are extremely similar to those seen in non-heat treatable Al-Mg alloys deformed to equivalent strains that have Mg levels of over 5 wt.% [33]. Mg has often been associated with a reduction in the stacking fault energy (SFE) in Al, which in turn restricts cross slip. However, its main effect, on inhibiting recovery and dislocation cell formation, is from the drag of solute atmospheres and the thermal activation of solute atoms away from climbing jogs with screw dislocation segments [33,50]. In the current material the Mg level was less than 1 wt%, so such a strong effect would not be expected from this element, but the Cu content in AA2195 is around 4 wt%. Relative to Mg, the influence of Cu on recovery in Al solid solutions has historically received less attention. While Cu also increases the SFE [51], when compared at the same atomic concentration, it has been found to have a greater influence on both solid solution strengthening and reducing the rate of recovery [33,47]. Furthermore, recent atom probe studies have shown clear evidence of Mg and Cu segregation to dislocations in the early stages of ageing [16]. In Al-Cu-Li-Mg alloys the nucleation of the T1 phase on dislocations has now been well documented [9–11]. Thus, it is highly likely that the ageing process has a powerful effect on inhibiting recovery of dislocation structures in an alloy where there is such a strong association between age hardening and dislocation assisted nucleation.

The increase in dislocation density with strain was also found to affect the precipitate size and size distribution of the T1 phase, with respect to the diameter of the plates. Measurements from STEM images (Figure 11) demonstrated a continued decrease in the T1 plate’s average diameter, as well as a reduction in the spread in their size distribution, and a corresponding increase in number density with increasing pre-strain levels above 9%. The
spatial distribution of the $T_1$ phase was also found to reflect the high and uniform dislocation density seen in the samples subjected to large pre-strains and increased in density and homogeneity throughout the matrix even up to pre-strain levels of 15% (Figure 9). The greater and more uniform nucleation site density provided by stretching to higher strain levels, thus led to a reduction in the length individual precipitates could grow to before solute was depleted from the surrounding matrix by overlap of neighbouring diffusion fields. The reduced variability in distance between nucleation sites, caused by the high dislocation density, can also be correlated to the reduction observed in the spread in the $T_1$ plates' size distribution with higher levels of pre-stretching, which was also shown to lead to a reduction in the JMAK exponent, $n$, with increased pre-strain (Figure 12).

In contrast, the $T_1$ plate thickness remained constant (at approximately 1.3nm) with increasing pre-strain values into the more extreme stretching range. This result is in line with previous investigations which have shown that at standard artificial aging temperatures (i.e. 155°C), once nucleated, the thickness of the $T_1$ phase remains constant at a single minimum structural unit high for extremely long ageing times, owing to its high coherency strain [1,20]. $T_1$ plate thickening has only been observed to occur at higher ageing temperatures (e.g. 190 °C) through the nucleation of new growth ledges which add four new atomic layers as they progress across the broad faces of the plate [1,52,53].

The predictions plotted in Figure 15 showed that, when the large contribution from strain hardening is taken into account, the thin plate precipitation hardening model proposed by Nie and Muddle [25], gave accurate predictions for the effect of pre-strain on the T8 yield stress. In applying this model all the precipitate parameters were measured directly from HAADF-TEM images, apart from volume fraction, which was taken from the literature. The only adjustable parameter used was the effective interfacial energy, which with a fitted value of $\gamma_{eff} = 0.089$ Jm$^{-2}$ gave a good agreement to the experimentally determined yield stress. This value of $\gamma_{eff}$ is extremely close to the interfacial energy for the $T_1$ precipitate obtained by Dorin et al. [1] of $\gamma_i = 0.085$ J m$^{-2}$, in their original model, and for thin $T_1$ plates formed by ageing at lower temperatures Dorin et al. have further shown that the $\gamma_{eff}$ is dominated by $\gamma_i$ as the stacking fault energy, $\gamma_{SF}$, is only about 6% of $\gamma_i$. 

26
As pointed out by Dorin et al. [1], for most metallurgists it is counter intuitive that refinement of a precipitate size distribution by stretching an age-hardening alloy can lead to a reduction in the strength contribution from precipitation. However, for the T₁ phase this case has been well argued in their series of papers [1,19,20,22,54,55] and is a direct consequence of the thin nature of the T₁ precipitates, which leads them to be sheared by dislocations. The results presented here fully corroborate this finding, in that when the larger than expected effect of strain hardening is subtracted from the yield stress, it is very apparent that the strengthening contribution from precipitation reduces with increasing pre-strain in the high strain range investigated (Figure 15). As discussed by Dorin et al. [1], this behaviour can be correlated to the reduction in diameter of the T₁ plates that occurs on increasing the level of stretching, which is caused by the increase in nucleation site density. With a constant plate thickness and volume fraction, this results in a higher number density of precipitates (Figure 11 (d)), but with thin disc shaped particles the increase in statistical interactions with a dislocation line from a greater obstacle density in the slip plane is outweighed by the larger effect that reducing the plate diameter has on the obstacle’s strength.

Thus, while previous research has attributed the increase in yield strength seen in alloys like AA2195 with increased pre-strain to an increase in the number density of fine T₁ precipitates [9,14], our results reveal the opposite, in that we see a reduction in the T₁ strengthening contribution with increasing levels of pre-strain and that the progressive increase in strength that results from high levels of pre-strain is a consequence of the higher level of strain hardening retained in the material owing to exceptionally low rates of recovery. This conclusion is exemplified by Figure 15, where, for the 15% pre-stretched condition, the increase in strength estimated from strain hardening is about 50% greater than that from the T₁ precipitates. In fact, at this pre-strain level, the increase in strength predicted from precipitation hardening has reduced by 40% relative to that seen for a conventional industrial stretching treatment involving a 3% pre-strain.
6. Conclusions

It has been demonstrated that the increase in yield strength seen in Gen3 Al-Li-Cu alloys, on stretching following solution treatment, continues to much higher pre-strain levels than are currently used in industrial practice. The practical limit that can be applied, owing to plastic instability in tensile stretching the solution treated alloy, was determined to be at a plastic strain of about 15%. Nevertheless, at this pre-stretch level the yield strength of the AA2195 alloy investigated was found to increase to ~ 670 MPa, compared to ~ 580 MPa with the standard stretch and T8 temper.

A linear increase in dislocation density with pre-strain was determined from XRD measurements and in the TEM a very uniform dislocation distribution was still seen at high pre-strain levels. Surprisingly, little reduction in dislocation density was detected after artificial ageing, suggesting that recovery was greatly inhibited. This has been attributed to a strong solute interaction between dislocations and the Cu and Mg in solution, as well the strong segregation of Cu and Mg to, and the nucleation of T₁ plates on, dislocations during the early stages of artificial ageing.

Measurements from STEM images revealed a continued decrease in the T₁ plate’s average diameter, as well as a narrowing of their size distribution and an increase in number density, with increasing pre-strain levels to 15%. The spatial distribution of the T₁ phase was also found to reflect the uniform dislocation density seen in the samples subjected to large pre-strains and this led to a reduction in the JMAK exponent (n) with increased pre-strain. However, the thickness of the T₁ plates remained constant (at approximately 1.3nm) into the more extreme stretching range.

Good agreement was found between the experimental results and predictions of the effect of pre-stretch on the strain and precipitation hardening contributions to the alloy’s yield stress. In agreement with the work of Dorin et al. [1], it was found that the strength contribution from precipitation hardening decreased with increasing pre-stretch, owing to the strong dependence of the obstacle strength on the plate diameter and refinement of the T₁ precipitates with increasing dislocation density. In contrast, due to the low level of recovery, the contribution from strain hardening was shown to increase parabolically with
pre-stretch. Thus, while previous research has attributed the increase in yield strength seen in alloys like AA2195 with increased pre-strain to an increase in the number density of $T_1$ precipitates, the results here suggest the opposite. In that, there is a reduction in the $T_1$ strengthening contribution with increasing levels of pre-strain and the increase in strength that results from high levels of pre-strain is a consequence of the high level of strain hardening retained in the material. In fact, it was found that for pre-stretch levels greater than 10% the contribution from strain hardening exceeded that from precipitation.

**Acknowledgements**

The author would like to thank the EPSRC funded LATEST2 Programme (EP/G022402/1) and Primetals Technologies Ltd for financially supporting this project, and Constellium for the supply of materials. Particular thanks are owed to Dr. Richard Cindery for facilitating the project and providing continued encouragement and support. Thanks are also given to the technical staff at the University of Manchester who assisted in the manufacture of test pieces and in the use of laboratory equipment. In particular, thanks go to Matthew Smith for his assistance in operating the TITAN microscope.

**References**


