Influence of multi-step heat treatments in creep age forming of 7075 aluminum alloy: Optimization for springback, strength and exfoliation corrosion

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ABSTRACT

Multi-step heat treatments comprise of high temperature forming (150 °C/24 h plus 190 °C for several minutes) and subsequent low temperature forming (120 °C for 24 h) is developed in creep age forming of 7075 aluminum alloy to decrease springback and exfoliation corrosion susceptibility without reduction in tensile properties. The results show that the multi-step heat treatment gives the low springback and the best combination of exfoliation corrosion resistance and tensile strength. The lower springback is attributed to the dislocation recovery and more stress relaxation at higher temperature. Transmission electron microscopy observations show that corrosion resistance is improved due to the enlargement in the size and the inter-particle distance of the grain boundaries precipitates. Furthermore, the achievement of the high strength is related to the uniform distribution of ultrafine $\eta'$ precipitates within grains.

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1. Introduction

The 7xxx series aluminum alloys have been widely used as structural materials in aeronautical industries due to their attractive comprehensive properties, such as low density, high strength, ductility, toughness and resistance to fatigue [1–3]. However, the 7xxx series aluminum alloys have some application limits in forming integral wing panels by traditional manufacturing process due to their poor assembling ability and the increase of weight. Therefore, developing a new superior forming technique becomes a major issue for possible applications of high strength aluminum in aircraft components. Considered the accuracy to assemble, creep age forming (CAF) with easy operating is preferred and developed [4,5].

CAF is a combined age hardening process with the stress induced deformation/forming of metal parts. The fundamental mechanism is the stress relaxation phenomenon that can arise when a heat treatable alloy is artificially aged. In this process, a solution treated material sheet is loaded onto the tool and then heated to a temperature for artificial ageing. During the thermal exposure, the constituents of the metal precipitate and alter the microstructure of the material. At the same time, stress relaxation occurs due to creep and permanent deformation of the material takes place [5–7]. One of the greatest challenges in this process is the springback which occurs when the load is released. Therefore, final shape of the component is somewhere between its original shape and the tool shape. Springback occurs due to the limited ageing period which is required to achieve proper mechanical properties [8]. High levels of springback often result in the formed component being out of tolerance, extra after-work problems during final assembly and also deficient aerodynamic behavior [9].

On the other hand, the 7xxx series aluminum alloys in high level of tensile strength are sensitive to localized...
corrosion, such as intergranular corrosion, exfoliation corrosion and stress corrosion cracking (SCC) [10]. Among the various localized corrosion, exfoliation corrosion is particularly relevant to plates presenting grains of large aspect ratio and subjected to environmental exposure where potential trapping and concentration of water can occur, such as riveted areas in aircraft structure [11]. The resistance of such alloys to exfoliation corrosion is generally increased through overaging treatments, which unfortunately, significantly decrease the mechanical strength. In order to achieve a good combination between strength and corrosion resistance, lots of effort has been carried out to acquire multi-step heat treatments.

In terms of experimentation, assessment of the susceptibility of aluminum alloys to exfoliation corrosion has been proposed by various procedures. Alloy and aircraft manufacturers commonly use tests such as those set forth in ASTM G66, ASTM G85, and ASTM G34 (the so-called EXCO, i.e. exfoliation corrosion, test). However, most of these tests, although widely accepted, yield only qualitative results. The EXCO test is based on the visual examination of a corroded surface. Therefore, a method that would provide a quantitative evaluation of exfoliation corrosion sensitivity would be useful to optimize heat treatment [12]. Recently, some works have been published using impedance technique to characterize the exfoliation corrosion of aluminum alloys, establishing a better understanding of corrosion attack and improving the reliability of the technique as a tool for characterizing this corrosion process [13].

To date, a limited number of studies have been published which focus on the principles of CAF technique and on application to selected standard aerospace aluminum alloys [5,8]. But, there is no report for optimizing the process with respect to properties in the 7xxx series aluminum alloys. In this paper, the effect of multi-step heat treatments on the tensile properties, exfoliation corrosion and springback of the 7075 aluminum alloy was studied. These results can give indispensable information for optimizing the CAF process.

2. Experimental Procedure

The material used in this study was a commercial 7075 aluminum alloy sheet with 3 mm thickness. The chemical composition of the alloy is given in Table 1.

Sheet specimens with the dimensions of 400 mm × 100 mm × 3 mm were used for CAF tests. The tool surface was in a cylindrical form with a curvature radius of 310 mm which leads to a maximal displacement \(d_{\text{max}}=65\,\text{mm}\) in the middle section of the tool. Fig. 1 shows the tool surface.

All specimens were solution treated at 475 °C for 50 min and water quenched to room temperature. Then CAF was performed as following sequence:

1) The specimen was mounted on the tool (lower die).
2) The load (upper die), weighing 60 kg, was placed on the specimen to deform it into complete contact with the tool.
3) The whole assembly was placed in the furnace and isothermally heated to the temperature of forming.
4) After forming for certain times, the tool, load and specimen were cooled out of the furnace.

The amount of springback was measured at the center of the length as shown in Fig. 2 according to Eq. (1):

\[
\text{Springback(\%)} = 100\left(\frac{\delta}{d_{\text{max}}} - 1\right)
\]

More details about CAF procedure and test facilities can be found in our previous paper [14].

The room temperature tensile property tests were performed using an Instron 5500 universal testing machine at a tensile velocity of 2 mm/min. The stress-strain curves were used to determine yield (0.2% offset) points.

The accelerated exfoliation corrosion (EXCO) test was performed according to the standard EXCO test as described in ASTM G34-01 [15].

The EXCO test solution was 4.0 M NaCl + 0.5 M KNO\(_3\) + 0.1 M HNO\(_3\). The initial apparent pH of the solution was 0.4. The solution temperature was maintained at 25 °C using a thermocouple.

The polarization curve and electrochemical impedance spectroscopy (EIS) experiments were carried out using EG&G PRINCETON APPLIED RESEARCH 273A electrochemical system. A saturated calomel electrode (SCE) was used as the reference electrode, the platinum sheet was served as the counter-electrode, and the studied alloy was used as the working electrode. The equivalent circuits simulating the electrochemical response of the system were constructed using Zview software.

Microstructural observations were carried out with a Transmission Electron Microscope (TEM). The TEM specimens were prepared by the standard twin-jet electropolishing method using...
70% methanol and 30% nitric acid solution below −20 °C, and then characterized by JEOL electron microscopy operated at 200 kV.

3. Results and Discussion

3.1. Tensile Properties and Springback

Fig. 3 demonstrates the variations in yield strength of the alloy at 150 °C and 190 °C for different forming times. At forming temperatures of 150 °C and 190 °C, the yield strength gradually increased with the increase of forming time and reached maximum yield strength of 450 MPa and 300 MPa after 24 h and 6 h forming time, respectively. This behavior mainly derives from precipitation hardening and strain hardening during CAF of alloy [9]. It appears that maximum yield strength occurred in the 150 °C/24 h and 190 °C/6 h forming condition and therefore this condition selected as the base for multi-step heat treatment (see Table 2).

Fig. 4 shows the springback and tensile properties of the samples after CAF at different heat treatments. It should be noted that the tensile properties and springback of the sample formed at 150 °C/24 h (sample A) are also presented for comparison with samples formed by multi-step heat treatments. The ultimate tensile strength (σ_{UTS}), yield strength (σ_{0.2}) and springback (R %) of sample A are 535 MPa, 450 MPa and 37, respectively. The σ_{UTS}, σ_{0.2} and R% for the sample B are 400 MPa, 250 MPa and 30, respectively, which are lower than the sample A. It is clear that for sample B, the tensile properties decline significantly. So two-step heat treatment is not favorable. For the three-step heat treated samples (C, D and E samples), the strength first increase and then decrease with increasing forming time at 190 °C. The maximum strength is obtained at the sample D. The σ_{UTS}, σ_{0.2}, and R% of sample D are 530 MPa, 440 MPa and 34 respectively. Considered the springback and strength, A, B and D samples were selected for evaluation of exfoliation corrosion susceptibility and microstructural characterization.

3.2. Exfoliation Corrosion Behavior

Fig. 5 indicates the surface morphologies of the samples after immersing in the EXCO solution for 48 h. Regarding the corroded surface, it is obvious that the lifted layers peeled off from the surfaces of A and B samples are more than the Sample D, which indicates that these samples are more susceptible to exfoliation corrosion than that of Sample D. Meanwhile, it is also found that Sample B is a little more susceptible to exfoliation corrosion than the Sample A, as seen in Fig. 5a and b.

In order to compare the exfoliation corrosion susceptibility of 7075 Al alloy under different heat treatments, EIS diagrams were carried out in EXCO solution. Fig. 6 shows the experimental EIS plots for all samples. The Nyquist plots (Fig. 6a) revealed only a depressed capacitive arc in the high-medium frequency range which indicates that the corrosion process is under activation control. Moreover, an inductive loop in the low frequency range related to the initiation of pitting corrosion [16,17]. Scientists demonstrated that inductive behavior is more likely promoted by the weakening of the aluminum oxide layer due to the anodic dissolution of aluminum alloy. Besides,
inductive resistance can be produced in inducing period of pitting of the oxide film. As soon as the pits are initiated, inductive resistance disappears [18,19]. Considering the Bode-phase plots (Fig. 6b), it can be noted that all samples present only one time constant. Conde and de Damborenea [13,16] believed that the onset of delamination marks the appearance of the two time constants. When the two time constants were detected and the low frequency inductive loop vanished, delamination was initiated. Conversely, when the attack is minimal and only some localized pits appear on the working electrode, the corresponding impedance diagram was constituted by a single time constant. Thus, the exfoliation corrosion can be detected by means of the appearance of two time constants. According to the results which can be seen in Fig. 6b, it is obvious that two time constants cannot be differentiated and the inductive loop can be seen in all samples, so it indicates that the high exfoliation corrosion susceptibility of samples cannot be expected after 48 h immersion in EXCO solution.

In order to give quantitative support to experimental results and better explanation about the susceptibility to corrosion, impedance parameters were obtained by the Zview software using an equivalent circuit model [20,21], drawn in the inset of Fig. 6b. The physical meaning of the equivalent circuit elements is shown as following: R_s is the ohmic resistance of the electrolyte; C_1 is the capacitance of the film; R_p is the pore resistance of the film; C_2 is the capacitance of the double layer of the aluminum alloy; and R_c is the polarization resistance of the electrode. Based on the circuit, the typical EIS parameters which are listed in Table 3 can be calculated. It is clear from the values of the elements are listed in Table 3 that sample D has higher R_p or the porous layer polarization resistance than that of A and B samples. Moreover, The C_1 (The capacitance of porous layer) for sample D exhibits the lowest measured values, which can be associated with both an increase in the passive layer thickness and a decrease in the oxide film dielectric constant [22]. Generally, a system with lower C_1 and higher R_p is more resistant to corrosion [20]. These were further confirmed that the sample D showed higher

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**Fig. 5** – Corrosion morphologies of 7075 aluminum alloy after 48 h of immersion in EXCO solution (a) Sample A; (b) Sample B; (c) Sample D.

**Fig. 6** – (a) Nyquist diagram; (b) Bode-phase diagram with equivalent circuit model for all immersion samples in EXCO solution after 48 h immersion.
corrosion resistance while the sample B showed the lower corrosion resistance.

The polarization curves after immersion for 48 h in EXCO solution are shown in Fig. 7. Electrochemical characteristics derived from the polarization curves are also listed in Table 3. It is seen that the corrosion current density ($I_{corr}$) of sample D is much lower than that of A and B samples, while the polarization resistance ($R_p$) of sample D is the greatest. According to the results, it can be concluded that the corrosion behavior of sample D is higher than other samples. In general, it can be seen that the polarization results are in agreement with the EIS measurements and the corrosion resistance of sample D is the highest after 48 h immersion in EXCO solution.

### 3.3. Microstructural Characterization

In order to investigate the effect of microstructure on the properties, microstructural characterization was performed by Transmission electron microscopy. The TEM bright field images of the 7075 aluminum alloy heat treated at different conditions are shown in Fig. 8. As could be seen in Fig. 8a for sample A, a high density of ultrafine precipitates is distributed homogeneously in the matrix. The analysis of selected area electron diffraction (SAED) pattern and High Resolution Electron Microscopy (HREM) image showed that the GP zones and $\eta'$ precipitate are the main phases in the matrix. Also the precipitates at grain boundaries with continuous distribution belong to $\eta$-MgZn$_2$ phase.

Forming at 150 °C/24 h plus 6 h at 190 °C shows a significant coarsening of the precipitates (Fig. 8b). The density of the precipitates for sample B is much lower than sample A, which is attributed to one of the reason why its tensile strength is much lower than the other samples. The increase in the forming temperature enhances the diffusion rate of the solute atoms. Furthermore, dislocations act as short circuit diffusion path and consequently accelerate the precipitation and over-aging [23]. Analysis of SAED indicates that the $\eta'$ and $\eta$ precipitates are dominant phases in this condition. As could be seen in this figure, precipitates have two types of morphology: spherical and rod-like. Liu et al. [24] confirmed that the rod-like precipitates are the equilibrium $\eta$ phase.

<table>
<thead>
<tr>
<th>Sample</th>
<th>$R_p$ (Ω.Cm$^{-2}$)</th>
<th>$C_1$ (μF.Cm$^{-2}$)</th>
<th>$n_1$</th>
<th>$R_{po}$ (Ω.Cm$^{-2}$)</th>
<th>$C_2$ (FCm$^{-2}$)</th>
<th>$n_2$</th>
<th>$R_{ct}$ (Ω.Cm$^{-2}$)</th>
<th>$E_{corr}$ (V vs. SCE)</th>
<th>$I_{corr}$ A/Cm$^2$</th>
<th>$R_p$ (Ω.Cm$^{-2}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample A</td>
<td>6.707</td>
<td>1916.1</td>
<td>0.71435</td>
<td>22.79</td>
<td>-4.99×10$^{-2}$</td>
<td>0.83176</td>
<td>-11.06</td>
<td>-0.773</td>
<td>5.66×10$^{-4}$</td>
<td>51.5</td>
</tr>
<tr>
<td>Sample B</td>
<td>7.992</td>
<td>2113.9</td>
<td>0.73444</td>
<td>16.09</td>
<td>-1.162</td>
<td>0.8374</td>
<td>-2.362</td>
<td>-0.798</td>
<td>12.32×10$^{-4}$</td>
<td>29.33</td>
</tr>
<tr>
<td>Sample D</td>
<td>7.28</td>
<td>1267.1</td>
<td>0.72531</td>
<td>82.06</td>
<td>-2.36×10$^{-2}$</td>
<td>0.81679</td>
<td>-21.19</td>
<td>-0.834</td>
<td>1.89×10$^{-4}$</td>
<td>212.3</td>
</tr>
</tbody>
</table>

**Table 3** - Results of the electrochemical impedance measurements and potentiodynamic polarization in EXCO solution after 48 h immersion.

**Fig. 7** – Potentiodynamic polarization curves for all immersion samples in EXCO solution after 48 h immersion.

**Fig. 8** – TEM bright field images and corresponding selected area diffraction pattern of the alloy at different CAF condition (a) Sample A; (b) Sample B; (c) Sample D.
The microstructure of sample D is shown in Fig. 8c. It appears that the precipitates in the matrix are similar to that of sample A, however, the precipitates are slightly denser and coarser. The precipitates are discontinuous at the grain boundaries. As in the above discussion, the main precipitates inside the grains at 150 °C/24 h forming condition are fine and homogenously distributed GP zones and $\eta'$ precipitates. It is obvious that the dissolution of GP zones and nucleation of $\eta$ precipitates can be occurred at 190 °C. The dissolution of GP zone offer great amounts of solution atoms and is beneficial for the precipitation in the subsequent forming treatment. After re-forming at 120 °C/24 h the $\eta$ phase nucleated at 190 °C growth [22,25]. Furthermore, some coarse $\eta'$ precipitates are observable within the grains. Wang et al. [26] proposed that formation of some coarse $\eta'$ precipitates in the matrix can be attributed to the dissolution of the GP zones which decrease the nucleation sites for the $\eta'$ precipitates.

The precipitate free zones along grain boundaries could be seen in Fig. 9. These zones negatively influence mechanical properties because they provide locations for easy dislocation motion. The width of precipitate free zones for A, B and D samples are 70 nm, 110 nm and 70 nm, respectively. Therefore, another reason of the loss of strength in the samples is due to formation of precipitate free zones.

In general, solute depletion and vacancy depletion theory describe the formation of the precipitate free zones as a result of heat treatment of materials. At high temperatures during the CAF the $\eta$ phase has optimal conditions for the growth and subsequent coarsening. During these processes a great amount of solute elements is consumed which results in the depletion of solutes in the matrix near the growing $\eta$ phase boundary as the new solute atoms may come only by the re-dissolution of metastable $\eta'$ phase near the grain boundary [27]. This is in good agreement with experimental results.

A consensus has not yet been reached on the mechanism of exfoliation corrosion, and several views have been proposed. It has been suggested that exfoliation corrosion belongs in the same category as inter-granular corrosion and stress corrosion cracking. Severe inter-granular corrosion will promote exfoliation corrosion, and the wedging stress caused by corrosion products plays a significant role in development of exfoliation corrosion [28].

The inter-granular corrosion was mainly attributed to the difference in potential between the matrix and grain boundary precipitates and influenced by a range of the microstructure, such as the size and inter-spaces of precipitates, precipitate free zone and solute concentration gradients. It is well known that $\eta$ precipitates are noted as being anodic to the Al alloy matrix. Therefore, galvanic reactions between the $\eta$ precipitates at the grain boundaries and the alloy base at their adjacent periphery are inevitable. Grain boundaries with continuous $\eta$ precipitates become more susceptible anode channel and offer a path for inter-granular corrosion [29].

As in the above discussion, the nearly similar exfoliation corrosion susceptibility of A and B samples is attributed to the similar distribution of grain boundaries precipitates. This feature of grain boundaries precipitates shows the greatest exfoliation corrosion sensitivity. It should be noted that the higher exfoliation corrosion susceptibility of sample D is due to the higher volume fraction of the $\eta$ phase at the grain boundaries. The exfoliation corrosion resistance of sample D is greatly increased which is in good accordance with the result of microstructural characterization.

It is worth mentioning that during inter-granular corrosion process, the corrosion products accumulate at the grain boundaries. Moreover, the corrosion products have a higher specific volume than the aluminum alloy. Obviously, more materials will be converted during exfoliation corrosion. The expansion of the corrosion products results in high tensile stresses (wedging stress) on the grain boundary. These wedging stresses cause the lift off of surface grains or even of entire surface layers [30].

Furthermore, the exfoliation corrosion can initiate from pitting corrosion, which originates from the active sites where point defects accumulated [17]. During the CAF process the samples were stressed, the number of active sites such as dislocation can increase, which improved the corrosion process.

In CAF process two phenomena (age hardening and creep/stress relaxation) occur. Ageing is a process that can increase the strength of a metal, while creep during ageing is the
mechanism to promote forming of the formed shape of the part. For forming temperatures higher than 0.4 T_M (T_M is absolute melting point), as they are done within this work, the dominant range of creep is primary and steady state creep, which mainly shows dislocation creep [14,31]. Therefore, the deformation mechanism is related to the dislocation motion and interaction with precipitates. The interaction of dislocation/precipitate occurs by two different mechanisms: particle bypassing mechanism and particle cutting mechanism. It was suggested that as the precipitate is less than a certain critical size, the strengthening effect through Orowan bypassing mechanism is greater than that through dislocation shearing mechanism [10,32].

It is often reported that the usual precipitation sequence of 7xxx series aluminum alloys can be summarized as [33–35]:

7xxx → GP zones → η’ → η(MgZn2)

where GP zones are Guinier–Preston zones, η’ is a metastable hexagonal phase and η is an equilibrium phase, MgZn2.

At the beginning of the CAF the alloy is in solution treated condition. Thus, the dislocations can move without hindrance (primary creep). When precipitation proceeds the moving dislocations will be pinned and reduces creep rate. Creep continues only if dislocations are able to disappear again (dislocation recovery) [9,26]. During CAF at 190 °C the dislocation recovery and relaxation of internal stresses can take place. As a consequence, more creep strain is accumulated. This is a favorable aspect of the forming process as it tends to reduce springback. It is worth to mention that based on the results obtained in this research CAF at (150 °C/24 h + 190 °C/40 min + 120 °C/24 h) was selected as the optimal condition.

4. Conclusions

In summary, a multi-step heat treatment in CAF of 7075 aluminum alloy has been presented that simultaneously improves the properties and springback. This significant improvement is related to the microstructural changes in CAF. The main points of the present study are summarized in the following statements:

• Homogenously distribution of fine η’ precipitates within grains, leading to the maintaining high strength.
• Heterogeneous nucleation of the η phase by dislocations and discontinuous distribution at the grain boundaries. This grain boundary η morphology results in the low exfoliation sensitivity.
• Dislocations annihilation at higher forming temperature and consequently more creep strain (lower springback).

REFERENCES


