Age Hardening in 6061Al Alloy and Al₂O₃/6061Al Composite

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6061 aluminium alloy and the alloy containing 15% volume fraction of Al₂O₃ particles were aged at 398 and 443 K. It was found that the hardness in reinforced alloy reached its peak value at much shorter ageing time compared to the monolithic alloy when aged at 443 K. However, the reinforced material did not show any acceleration in precipitation hardening when aged at 398 K. Transmission electron microscopic investigation showed that hardness increased as fine needle-shaped zones (GP-II or called β⁺) appeared, and the material attained peak hardness when the structure contained a high density of needle-shaped zones. As the ageing continued, the zones became coarser and fewer in number which led to decrease in the hardness. The presence of large Al₂O₃ particles accelerated coarsening of the β⁺ zones resulting in peak hardness which occurred at shorter ageing time in reinforced material compared to that in the monolithic alloy at 443 K. However, at a lower ageing temperature of 398 K, the coarsening of the zones was very sluggish in both reinforced and monolithic materials.

It was found that deformation prior to ageing of the material could modify the formation and coarsening of the precipitates. The microscopic observations revealed that formation of fine needle-shaped β⁺ zones was depressed by formation of β' phase in deformed material. Since the β⁺ zone is the only hardening agent in 6061Al alloy, reduction or absence of β⁺ can considerably or totally diminish the hardening ability in the deformed alloy.

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I. Introduction

Recently, several investigations have been carried out to study age hardening behaviour in aluminium alloy containing discontinuous reinforcements such as SiC and Al₂O₃ whiskers or particles. It has been found that existence of these reinforcements could accelerate precipitation or ageing in the matrix, and peak hardness could appear earlier than that in the monolithic alloy. One of the reasons for accelerated ageing is considered to be due to the high dislocation density generated from thermal mismatch between the reinforcement and the matrix, or residual stress field near reinforcements. However, accelerated ageing has not been observed in the reinforced materials when they are aged at some what low temperature. It is envisaged that this may be due to difference in ageing sequence of the phases at different temperatures but no systematic microstructural study has been pursued to support this suggestion.

The effect of deformation on ageing sequence in 6061Al alloy and its composites has been studied by several researchers. It was found that straining prior to ageing could modify ageing sequence in these alloys. It has been suggested that formation of the GP-II (β⁺) zones might diminish and the formation of the β' phase could be accelerated in deformed material. However, no detailed microstructural investigation has been carried out to study the effect of straining on the formation of such phases. The principal aim of this work is to study the precipitation behaviour and hardness response in 6061Al alloy containing large Al₂O₃ particles and the matrix 6061Al alloy during ageing. In order to compare ageing behaviour of the composite at different temperatures, the samples were aged at two temperatures, 443 K which is close to a recommended ageing temperature for 6061Al alloy and a lower temperature 398 K. The experimental investigation was also carried out in deformed samples given 55% reduction prior to ageing.

II. Experimental

6061 aluminium alloy and 6061 aluminium containing 15% volume fraction of Al₂O₃ particles used in this work were supplied by Alcan (Canada) Ltd. in the form of extruded bars. The Al₂O₃ particles are about 25 µm in diameter and are distributed uniformly in the matrix as shown in Fig. 1. Both reinforced and unreinforced alloys were first cast and then hot extruded. Geometry and dimensions of the ageing samples machined from extruded bars are shown in Fig. 2. Prior to ageing, the samples were solution treated at 813 K for 2 h and water quenched. Solution treated samples were aged at 398 and 443 K for 680 and 50 h, respectively, and then water quenched. In order to study the effect of straining prior to ageing, the composite samples were strained 55% and aged at 398 K up to 780 h. Ageing was monitored by hardness measurements using Vickers hardness test with 5 kg of load.

Transmission electron microscopy was undertaken to examine the changes in precipitation and structure in the samples immediately after ageing. Differential scanning calorimetry (DSC) was used to analyse precipitation
kinetics. DSC specimens were heated from 298 to 773 K at a heating rate of 5 K/min. The specimens were quenched from temperature at which exothermic peaks appeared, and the precipitate structures corresponding to the exothermic peaks were examined by transmission electron microscopy.

III. Results

Figures 3 and 4 are plots of hardness versus ageing time for both reinforced and unreinforced materials aged at 398 and 443 K, respectively. It can be seen that the hardness of the composite is much higher than that of the monolithic alloy after solution treatment. High hardness resulted from the high density of dislocations and elastic strain field generated by thermal mismatch between the reinforcement and the matrix during quenching. As the ageing time increases, the hardness increases gradually in both alloys till the peak hardness values are attained. The curves of both materials are almost parallel to each other which suggests that the rate of increase in hardness for both reinforced and unreinforced alloys are similar indicating that the composite has a comparable age hardening capability as the monolithic alloy.

Both reinforced and unreinforced alloys attained their peak hardness values after 480 h of ageing at 398 K (Fig. 3). However, this was not the case when these materials were aged at 443 K (Fig. 4). The time to attain the peak hardness at 443 K was between 10 to 16 h for the reinforced alloy whilst 28 h for the monolithic alloy indicating that ageing was accelerated in the composite at this temperature.

Figure 5(a), (b) and (c) are micrographs taken from the composite samples aged at 398 K for 190, 480, and 680 h, respectively. The sample aged for 190 h shows the dot-like precipitates and the short needle-shaped phase. It should be noted that hardness after 190 h is much lower than the peak value. When the sample exhibits peak hardness after 480 h, the sample displays mainly dense and fine needle-shaped structure. As the ageing continues, the needle-shaped zones become coarser and the density of
the zones decreases (Fig. 5(c)). Figure 6(a), (b) and (c) are the TEM micrographs of reinforced alloy aged at 443 K for 6, 10 and 44 h, respectively. Similar ageing sequence is observed in these samples but precipitates are much coarser compared to that aged at 398 K. It can be seen that the needle-shaped zones in the sample aged for 10 h at 443 K (Fig. 6(b)) are coarser and the density of the zones is lower compared to that in the sample aged at 398 K for 480 h (Fig. 5(b)). Furthermore, over aged sample at 443 K for 44 h shows zones which are much coarser compared to those at 398 K for 680 h indicating rapid coarsening of the zones at high temperature.

The DSC thermograms of the two materials are shown in Fig. 7. Two exothermic peaks for the composite appear at lower temperature than that for the monolithic alloy. The results of DSC analysis are given in Table 1. First and second peaks for the composite occurred at lower temperatures compared to that for the monolithic alloy.

The precipitate structures at the two peaks for 6061Al alloy are shown in Fig. 8(a) and (b). These structures corresponding to exothermic peaks have been reported to be
Table 1  The initial reaction temperature ($T_i$), peak temperatures ($T_{p,1,2}$) and heat capacities ($\Delta H_{p,1,2}$) of the two materials.

<table>
<thead>
<tr>
<th></th>
<th>6061Al</th>
<th>Al$_2$O$_3$/6061Al</th>
</tr>
</thead>
<tbody>
<tr>
<td>$T_i$ (K)</td>
<td>467</td>
<td>465</td>
</tr>
<tr>
<td>$T_{p1}$ (K)</td>
<td>505</td>
<td>501</td>
</tr>
<tr>
<td>$\Delta H_{p1}$ (J/g)</td>
<td>3.93(453–533 K)*</td>
<td>4.50(453–523 K)</td>
</tr>
<tr>
<td>$T_{p2}$ (K)</td>
<td>553</td>
<td>547</td>
</tr>
<tr>
<td>$\Delta H_{p2}$ (J/g)</td>
<td>1.28(423–573 K)</td>
<td>1.28(423–573 K)</td>
</tr>
</tbody>
</table>

*Peak integration temperature range.

$T_{p1}$ and $T_{p2}$: First and second exothermic peak temperatures.

$\Delta H_{p1}$ and $\Delta H_{p2}$: Heat reaction capacities in the first and second peak regions.

$\beta^+$ (or GP-II) and $\beta'$ phases by Dutta. However, the structures obtained in this work (Fig. 8(a) and (b)) are coarser compared to those found by Dutta et al. A possible reason for such a difference in structure might be the lower heating rate used in the present work compared to that employed by Dutta et al.

Hardness curves for deformed and undeformed composite aged isothermally at 398 K are shown in Fig. 9. Deformed material has higher hardness compared to undeformed material at the beginning of ageing, and it increases gradually till the maximum value is attained after 190 h of ageing. On the other hand, hardness of undeformed alloy increases gradually in the first 100 h of ageing and then increases rapidly to its peak value after 480 h. The DSC thermograms for both deformed and undeformed composites are shown in Fig. 10. The deformed sample exhibits one small exothermic peak at 443 K and a large peak at 490 K (third peak at 640 K is a recrystallization peak).

![Fig. 8](image_url) Precipitate structures in 6061Al samples quenched from peak exothermic temperatures shown in Fig. 5. (a) first peak and (b) second peak.

![Fig. 9](image_url) Hardness of Al$_2$O$_3$/6061Al alloy in both deformed and undeformed conditions versus ageing time at 398 K.

![Fig. 10](image_url) Differential scanning calorimetric analysis of deformed and undeformed Al$_2$O$_3$/6061Al composite. Temperature range: 302 to 773 K and heating rate: 5 K/min.
tion reaction), while the first peak in undeformed sample occurred at 501 K and the second peak at 547 K. These results indicate that the deformation accelerates the reaction significantly in this material.

The structures of deformed samples after ageing were examined by TEM microscopy. Figure 11(a) and (b) are the micrographs of deformed samples aged at 398 K for 190 h and 780 h respectively. Heavily dislocated structure is present in peak aged sample (190 h) but the zone structure cannot be seen in this condition. However, coarse rod-shaped precipitates can be observed in the over-aged sample after 780 h of ageing.

IV. Discussion

1. Ageing sequence in Al₂O₃ particle reinforced 6061Al composite

The shape of the ageing curves for both reinforced and unreinforced materials is quite similar indicating that the composites has comparable age hardening capacity as the monolithic alloy. By correlating aged structure (Figs. 5 and 6) with the hardness measurements, it can be seen that increase in the hardness is related to formation of dot or needle-shaped zones (Figs. 5(b) and 6(b)) and these zones are identified as GP or GP-II ($\beta''$) zones\(^\text{10,16,17}\). As ageing time increases, the density of the zones becomes higher, and the dot shaped zones changes to fine needle-shaped zones in the sample at the peak hardness. Coarsening of the $\beta''$ zones occurs when the alloys are over-aged and in addition to coarse $\beta''$ zones, some long needle-shaped precipitates probably $\beta'$ can be seen in the sample which has been over-aged for 44 h at 443 K (Fig. 6(c)). These observations suggest that $\beta''$ zone is the principal hardening agent in this alloy and the age hardening and over-ageing are due to the formation and coarsening of the $\beta''$ zones.

2. Effect of ageing temperature on ageing behaviour of 6061Al alloy and its composites

Both reinforced and unreinforced materials reached peak hardness after 480 h at 398 K, but it only took about 16 h for the composite and 28 h for the monolithic alloy to attain the peak hardness at 443 K. It is obvious that increase in temperature accelerates ageing of the alloys due to increase in diffusion rate which in turn enhances both nucleation and growth rates of the $\beta''$ zones. It should be noted that the increase in growth rate of $\beta''$ zones seems to be more pronounced as the temperature increases and this can be seen from structures in the samples with peak hardness aged at 398 and 443 K. The zone structure in the sample aged at 398 K is much finer and denser compared to that in the sample aged at 443 K indicating that growth of the zones is faster at high temperature. The occurrence of the fast growth of the zones reduces the ageing time required to reach the peak hardness significantly but this occurs at the cost of hardening capacity (Table 2). The data show that the values of $\Delta H$ (difference between solution treated hardness and peak hardness) for both composites and 6061Al alloy are lower in samples aged at 443 K than that in samples aged at 398 K indicating acceleration in zone growth at higher temperatures.

![Fig. 11 Microstructures of deformed Al₂O₃/6061Al alloy aged at 398 K for (a) 190 h and (b) 780 h.](image)

Table 2 Age hardening of two materials at different ageing temperatures.

<table>
<thead>
<tr>
<th>Temperatures</th>
<th>Materials</th>
<th>$H_s$</th>
<th>$H_p$</th>
<th>$\Delta H$</th>
</tr>
</thead>
<tbody>
<tr>
<td>398 K</td>
<td>Al₂O₃/6061Al</td>
<td>94</td>
<td>138</td>
<td>44</td>
</tr>
<tr>
<td></td>
<td>6061Al</td>
<td>75</td>
<td>117</td>
<td>42</td>
</tr>
<tr>
<td>443 K</td>
<td>Al₂O₃/6061Al</td>
<td>94</td>
<td>129</td>
<td>35</td>
</tr>
<tr>
<td></td>
<td>6061Al</td>
<td>75</td>
<td>115</td>
<td>40</td>
</tr>
</tbody>
</table>

$H_s$—Hardness of solution treated samples; $H_p$—Peak hardness of aged samples; $\Delta H$—Difference between $H_s$ and $H_p$. 
It has been reported that increase in ageing temperature could change ageing sequence in Al alloys\(^{11(12)7(18)}\). The DSC analysis in the present study shows that the zone structures are different at different temperature, that is, the \(\beta''\) zones form at about 493 K whilst the \(\beta'\) zones appear at 543 K. However, in certain temperature range below 493 K, the zone structure has not changed. For example, at both ageing temperatures of 398 and 443 K, the structures in aged samples consist of \(\beta''\) zones but the coarser structure is present at 443 K compared to that at 398 K.

3. **Effect of reinforcement on ageing behaviour of the alloy**

The results showed that presence of large volume fraction of ceramic reinforcements did not change ageing sequence in the composite in the ageing conditions used in this work. However, the composite did exhibit accelerated ageing at 443 K. DSC analysis showed that two exothermic peaks for formation of \(\beta''\) and \(\beta'\) in the composite moved to lower temperature compared to the monolithic alloy indicating that both formation of \(\beta''\) and \(\beta'\) phases are accelerated in the composite. However, in the present work accelerated ageing is not observed when the composite is aged at 398 K. It seems that the effect of reinforcements on the formation and growth of the zones may be diminished at low temperature of ageing. One probable explanation for such behaviour is as follows.

Residual strains and dislocations introduced by reinforcement could increase diffusivity and thus enhance nucleation and growth rates of the zones and intermetallic precipitates. However, it seems that the presence of high density of dislocations and residual strains have mainly increased growth rate of the zones during ageing at 443 K. This can be observed from DSC thermograms which show that the composite exhibits a quite similar initial reaction temperature but a lower peak temperature (first peak) compared to the monolithic alloy. It can therefore be said that as the reaction (precipitation) starts, it finishes at a shorter time in the composite, which could be as a result of high growth rate of the zones. High growth rate of the zones in the reinforced material can also be related to difference in \(\Delta H\) values (Table 2). It can be seen that \(\Delta H\) is lower for the composite compared to the matrix alloy when the both are aged at 443 K. Lower \(\Delta H\) for the reinforced material could be due to higher coarsening rate of the zones, which in turn decreases its ability to acquire maximum hardness during ageing at 443 K. However, at low temperature, 398 K, both nucleation and growth rates are low because of low diffusivity of solute elements. In addition to this, the dislocation activity is sluggish at low temperature, which in turn leads to a limited effect on ageing. The formation and growth of the zones in the reinforced materials may therefore not be very different from that in the monolithic alloy at low temperature. It can be seen that both the composite and the monolithic alloys attain peak hardness values at same time and \(\Delta H\) values are also similar (Table 2). As mentioned in the previous section that the ageing sequences in 6061Al alloy and Al\(_2\)O\(_3\)/6061Al composite at 398 and 443 K are similar and the hardening agent is \(\beta''\) phase at these temperatures. The accelerated ageing of the reinforced alloy observed at 443 K seems thus due to increased growth rate of \(\beta''\) zones at higher temperature and not as a result of change in the ageing sequence.

4. **Ageing behaviour of deformed Al\(_2\)O\(_3\) composite**

The overall effect of deformation on ageing behaviour in reinforced alloy generally involves the contribution from two processes—age hardening as well as recovery of deformed structure. Firstly, formation of the zones and the precipitates during ageing can lead to age hardening. On the other hand, recovery occurring in deformed material will result in decrease in hardness. The ageing curve of deformed material could therefore be considered as a result of combination of these two processes which might result in a lower hardening rate compared to the material in undeformed condition. However, the microstructure of the deformed sample in the present work did not exhibit significant recovery even after ageing for 780 h at 398 K (Fig. 11). This indicates that a lower peak hardness for the deformed material compared to undeformed material is due to degradation of age hardening capability instead of recovery.

A DSC study of the deformed composite has shown that first exothermic peak occurred at 443 K is very small and the second peak which appears at 490 K overlaps with the first peak. As discussed earlier that the first peak relates to the \(\beta''\) zone whilst the second peak corresponds to the \(\beta'\) phase, which means that the formation of the \(\beta''\) zone is depressed by formation of the \(\beta'\) phase. Since the \(\beta''\) zone is the only hardening agent in this alloy, the reduction or absence of \(\beta''\) considerably or totally diminish the hardening ability in the deformed material. This could be the reason why the deformed sample displays less hardening compared to undeformed material.

Suppression of \(\beta''\) zones in deformed material has been considered as a result of the zone forming elements, Si and Mg being trapped by the tangled dislocations after heavy cold deformation\(^{90}\). However, from DSC analysis in the present study, it was found that formation of both \(\beta''\) and \(\beta'\) phases was accelerated but the \(\beta'\) phase formed so rapidly that it started forming well before the complete formation of \(\beta''\) zones occurred. Generally, \(\beta''\) zones are uniformly nucleated in the matrix prior to \(\beta'\) phase. However, when diffusivity increases at high temperature or due to the presence of heavy strain in the material, the formation of \(\beta'\) phase can be considerably accelerated and the formation of \(\beta''\) zones will be reduced. The effect of deformation on the ageing behaviour is very different from that of reinforcements. The reason is that the deformed sample has much higher density of dislocations and defects such as vacancies compared to undeformed reinforced material. In addition of these defects, substructure boundaries introduced by defor-
mation could also act as diffusion channels leading to a considerably high formation rate of $\beta'$ phase in the deformed material.

V. Conclusions

(1) Age hardening in 6061 aluminium alloy was achieved by the formation of GP zone, but the peak hardness was contributed by a fine needle-shaped $\beta''$ phase (or called GP-II zone) at temperatures above 398 K. The hardness decreased as the materials were over-aged as a result of coarsening of $\beta''$ phase.

(2) The existence of large particles could accelerate the formation and coarsening of the $\beta''$ phase, and subsequently, ageing was accelerated. However, this could only be observed while the alloy was aged at high temperature. When the temperature was low, the formation and coarsening of the $\beta''$ phase became sluggish, and accelerated ageing in the reinforced alloy was hardly observed.

(3) Deformation could diminish formation of $\beta''$ zones and accelerate the formation of $\beta'$ phase in 6061Al composite during artificial ageing. This could lead to degradation of hardening ability of the alloy.

REFERENCES


