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Aging Characteristics of Short Mullite Fiber Reinforced Al-4.0Cu-1.85Mg Metal Matrix Composite

W. Li, J.P. Long, S. Jing, B.L. Shen, S.J. Gao, and M.J. Tu

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Mullite short fiber reinforced Al-4.0Cu-1.85Mg composite and its base alloy were fabricated by squeeze casting. The age-hardening behavior, precipitation procedure, microstructure of dislocation and precipitates, and the interfacial structure have been studied by means of hardness measurement (HB), differential scanning calorimetry (DSC), and analytical transmission electron microscope (ATEM), respectively. The short mullite fiber in the composite induces high dislocation density in the near vicinity of the interface after it is solutionized and quenched in ice water, and suppresses or delays the formation of GPB zones. The aged hardness of the composite is always higher than that of its base alloy, but there appears little difference between the time needed in the composite and in the base alloy to reach the peak hardness, which means that the acceleration effect of mullite fiber in the precipitation of Al-Cu-Mg alloy is not great enough. Mg also reacts with Al and SiO_2, resulting in the formation of spinel ($MgAl_2O_4$), which depletes Mg in the matrix and finally hinders the aging acceleration in the testing composite.

Keywords aging, Al-Cu-Mg alloy, composite, hardness, short mullite fibers

1. Introduction

Many investigations have verified that the presence of ceramic reinforcements (whisker, fiber, and particulate) can considerably influence the age-hardening behavior of aluminum alloys. The main results are [1-7] (1) GP or GPB zone formation is suppressed in the composites; (2) the precipitation of metastable phases such as θ′, S′, or β′ is accelerated in the composites; and (3) the time needed to reach peak aging is shortened in the composites compared with the unreinforced base alloys.

Some researchers have indicated that the suppression and acceleration effects of GP (B) zone and precipitation are much weaker, and in some cases both the antisuppression and the deceleration are also observed.[8]

Mullite fiber is a less expensive reinforcement for making metal matrix composites and so is a potential candidate for commercial applications in the near future.[9-12] Although the mechanical properties or wear resistant behavior of mullite fiber-reinforced aluminum-based metal matrix composite is discussed briefly in the literature,[9-12] the aging behavior of short mullite fiber-reinforced Al-Cu-Mg metal matrix composite has rarely been investigated. This article evaluates the effect of mullite short fiber on the precipitation kinetics and age-hardening efficiency in a mullite/Al-4.0Cu-1.85Mg composite. The interfacial microstructure and its relation to the aging behavior of mullite/Al-4.0Cu-1.85Mg composite have also been studied.

W. Li, J.P. Long, S. Jing, B.L. Shen, S.J. Gao, and **M.J. Tu,** School of Manufacturing Science and Engineering, Shichuan University, Chengdu 610065, China. Contact e-mail: wli@sc.homeway.com.cn.

2. Experimental Procedure

The matrix used in the current study is an Al-4.0Cu-1.85Mg aluminum alloy. The reinforcement is 18 vol.% short mullite fibers of 3-10 μm diameter and 20-100 μm length. The composite was fabricated by squeeze casting and the unreinforced Al-4.0Cu-1.85Mg alloy was cast simultaneously in the same solidification process: melt temperature 800 °C, preform temperature 450 °C, die temperature 300 °C, and infiltration pressure 60 MPa with 2 min of holding during infiltration. Small specimens of $40 \times 20 \times 10$ mm^3 and $\phi 4.5 \times 2$ mm^3 for hardness measurement and differential scanning calorimetry (DSC) analysis were cut from the main body, respectively. After solution treatment at 495 °C for 8 h, the specimens were quenched in ice water and immediately subjected to isothermal aging at 190 °C (hardness measurement), or stored in a refrigerator (below 0 °C) while awaiting the DSC test.

Brinell hardness measurements were conducted at room temperature after air-cooling from the aging temperature using a steel ball (ϕ5 mm) indenter and 7.35 kN for 30 s. Each reported hardness value is the average result of more than three measurements. DSC runs were carried out at a heating rate of 10 °C min^{-1} from 40-500 °C and under a dynamic dry argon atmosphere using a TA2910 DSC apparatus (Thermal Analysis Co., USA). The DSC thermograms were normalized to the mass of the matrix alloy.

Specimens for transmission electron microscopy (TEM) observation were prepared by standard methods involving mechanical grinding, polishing, and dimpling followed by ion milling of foils to perforation on a liquid nitrogen-cooled specimen stage to eliminate further aging during the thinning period. Morphological and microstructural studies were performed in a JEM-200CX TEM (Japanese Electric Microscopy, Japan) operating at an accelerating voltage of 160 kV.

For comparison, unreinfored Al-4.0Cu-1.85Mg alloy samples were prepared with the same thermal and mechanical treatments and tested under the same experimental conditions as the composite samples.

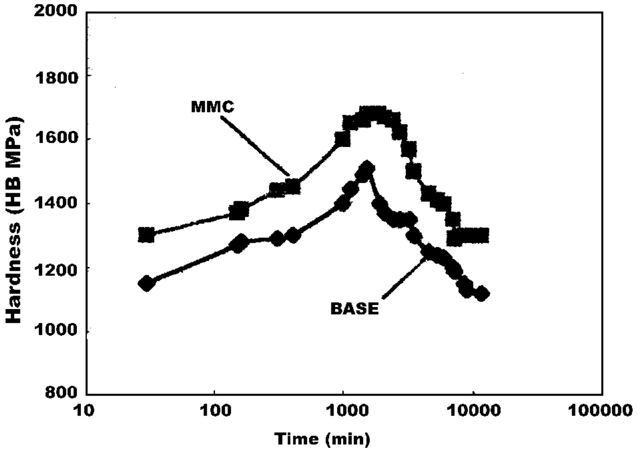

Fig. 1 Age-hardening curves of mullite/Al-4.0Cu-1.85Mg composite and Al-4.0Cu-1.85Mg alloy (aged at 190 °C)

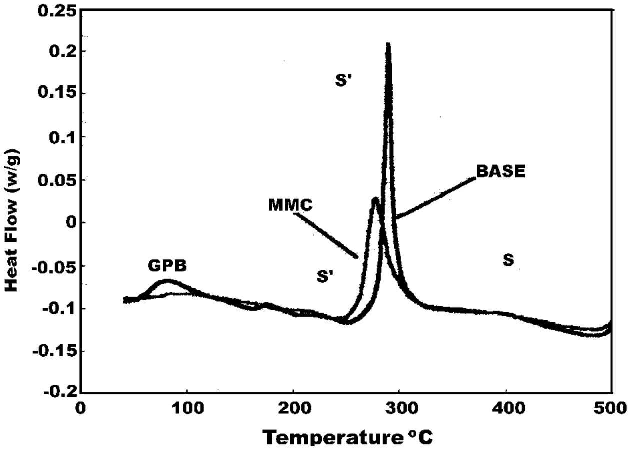

Fig. 2 DSC thermograms of mullite/Al-4.0Cu-85Mg composite and Al-4.0Cu-1.85Mg alloy (10 °C/min)

3. Results and Discussion

3.1 Age-Hardening Behavior

Figure 1 shows the age-hardening behavior of both mullite fiber-reinforced Al-4.0Cu-1.85Mg composite and the monolithic base alloy. Both curves represent the same typical age-hardening procedure; namely, hardness increases at early time and peak hardness appears at a specific time, then decreases the measured hardness as the aging time increases. However, the hardness of the composite is always higher than that of the monolithic base alloy, indicating that the composite is reinforced by the short mullite fiber, and that there is only a small difference between the times needed to reach the peak aging in the composite and in the base alloy. This later result is different from what many researchers have reported in literature;[1-7] i.e., reinforcement may accelerate the aging progress and largely shorten the time needed to reach the peak aging.

3.2 DSC Analyses

Figure 2 shows DSC thermograms of the reinforced and the unreinforced Al-4.0Cu-1.85Mg alloys, and Table 1 lists the peak temperatures, enthalpy, and activation energy values for precipitation reactions in the monolith and the composite.

Although the whole trend of the two curves is similar, which means that the age-hardening sequence in the two materials is identical, the two curves are different because of the addition of short mullite fibers.

From previous investigations in the literature,[1-7] it is believed that the exothermic peaks represent the formation of GPB zones, S'' and S' precipitates, and S (Al_2CuMg) stable phases, respectively, from low to high temperatures, whereas the endothermic peaks correspond to the dissolution of above zones or precipitates respectively. In the case of monolithic alloy, the onset and the dissolution of the GPB zone, S'' coherent precipitates, S' semicoherent precipitates, and noncoherent stable phase S (Al_2CuMg) are all observed. This finding follows well the ordinary sequence of ternary Al-Cu-Mg alloys, i.e., supersaturated solid solution (SSS) $\rightarrow$ GPB $\rightarrow$ S'' $\rightarrow$ S' $\rightarrow$ S (Al_2CuMg). For short mullite fiber reinforced Al-4.0Cu-1.85Mg metal matrix composite, however, the GPB zone formation is heavily suppressed, but the precipitation of S'' and S' phases is accelerated (Fig. 2 and Table 1).

Table 1 Peak Temperatures, Enthalpy, and Activation Energy Values for S' Precipitation Reactions in the Monolith and in the Composite

Materials	Tp, °C (a)	*H*, J/g	*Q*, kJ/mol
Al-4.0Cu-1.85Mg	288.63	−21.53	129.49
Mullite/Al-4.0Cu-1.85Mg	277.14	−22.82	113.72

(a) Tp, Peak performance of the DSC curves

3.3 Dislocation and Precipitates Morphology

Figure 3 shows the dislocation array in the composite sample after solution treatment and quenching in ice water. The highly densified dislocations in the near vicinity of mullite fiber matrix interface arise as a result of the mismatch of thermal coefficients between the reinforcement and the matrix during quenching from the solutionizing temperature. When aged at lower temperatures, the GPB zone formation of Al-Cu-Mg alloys is dominated by vacancy diffusion. The existence of denser dislocations acts as a dislocation sink, which delays the enrichment of Cu or Mg clusters, and hence suppresses the formation of the GPB zones. However, when aged at relatively higher temperatures, the precipitation of both coherent S'' and semicoherent S' is more energetic and the acceleration could be explained by the following possible mechanism: (1) a greater interfacial area, caused by additional reinforcement fibers and finer grain-subgrain structure of composites, provides more preferential nucleation sites for precipitation and enhances solute diffusion in composites; and (2) high dislocation density reduces the activation energy (as in Table 1) for the solute diffusion. Dislocation could become a rapid diffusion path of the solute. This mechanism is suggested by most researchers,[1-7,10-12] and the current study corresponds well to it.

Figure 4 shows the morphology of S' precipitates in the composite sample. This result is also observed in the monolithic base alloy, but both the amount and the magnitude of S'

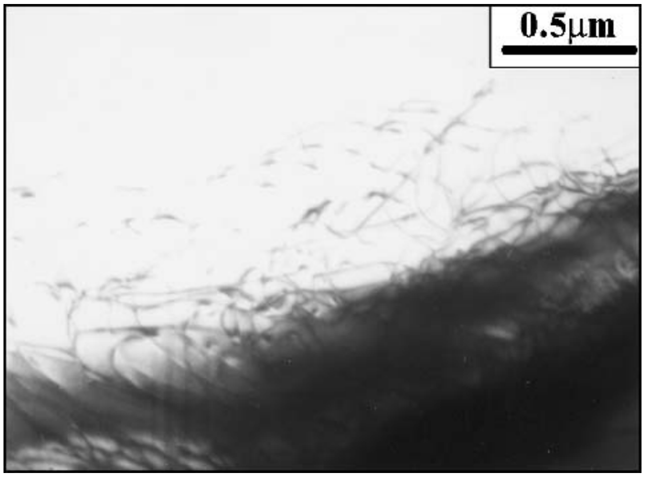

Fig. 3 Bright field TEM image of dislocations in the composite

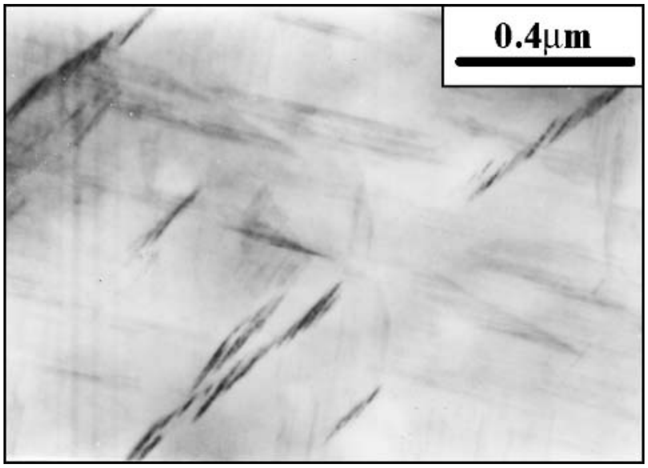

Fig. 4 Bright field TEM image of S′ precipitates in the composite

phases are relatively smaller at the same aging stage. Therefore, there exists an acceleration of aging, to some extent, in the mullite short fiber reinforced Al-4.0Cu-1.85Mg composite.

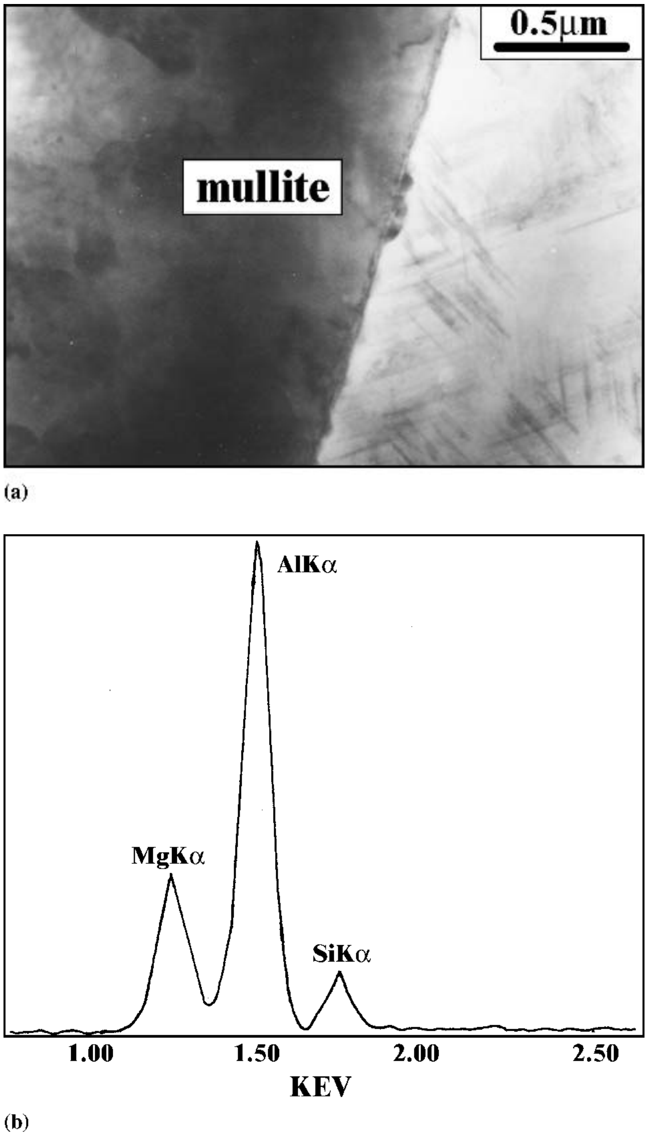

Fig. 5 Interfacial microstructure **(a)** and the spinel composition **(b)** of mullite/Al-4.0Cu-1.85Mg composite (ATEM image and EDAX)

3.4 Interfacial Reaction and Its Effect on the Aging Behavior of Mullite/Al-4.0Cu-1.85mg Composite

Many investigators have reported interfacial reactions in many aluminum-based metal matrix composites (MMCs), especially in MMCs that contain magnesium.[13-15] Although mullite fiber ($3Al_2O_3 \cdot 2SiO_2$) is stable when used as reinforcement in a matrix of Al-Cu-Mg alloy,[1] some interfacial reactions, as shown in Eq 1, may occur in the composite:

$$2SiO_2(s) + 2Al(l) + Mg(l) = MgAl_2O_4(s) + 2Si(l) \qquad \text{(Eq 1)}$$

Here, SiO_2 comes from the fiber itself[1-3,8] or the binder. $MgAl_2O_4$ is formed from Eq 1 during the infiltration fabrication of the composite[3-15] and may grow further in the process of heat treatment. Figure 5 shows the interfacial microstructure (Fig. 5a) and the related energy disperse analysis of x-ray (EDAX) analytical results of the interfacial reaction product (Fig. 5b). When reaction occurs at the mullite fiber/Al-4.0Cu-1.85Mg matrix interface, interfacial reaction products formed as a result of the reaction will alter the chemical composition of the matrix alloy. The more interfacial reaction products that are present, the less magnesium in the matrix. Therefore, there might be fewer precipitates in the composite than in the unreinforced base alloy when aged at a given temperature and at a given aging stage, resulting in the minor difference between the peak aging times.

4. Conclusions

1) Short mullite fiber can reinforce Al-4.0Cu-1.85Mg ternary alloy. The aged hardness of the reinforced composite is always higher than that of the unreinforced base alloy during the whole aging procedure. There is little acceleration of

aging in the composite compared with its unreinforced monolith on the basis of hardness measurement.

2) GPB zone formation is heavily suppressed in the short mullite fiber-reinforced Al-4.0Cu-1.85Mg composite because of the high density of dislocation in the near vicinity of interface. The precipitation of S″ and S′ phases is accelerated to some extent in the testing composite on the basis of DSC analysis. Both the reinforced and the unreinforced materials represent the same precipitation sequence.
3) Mullite fiber reacts with magnesium and aluminum during the fabrication procedure of mullite/Al-4.0Cu-1.85Mg composite. The product of the interfacial reaction is mainly spinel ($MgAl_2O_4$). It is the interfacial reaction that results in the depleting of magnesium in the Al-Cu-Mg matrix, and hence alters the age-hardening of short mullite fiber-reinforced Al-4.0Cu-1.85Mg composite.

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