J Mater Sci (2006) 41:5402–5406
DOI 10.1007/s10853-006-0243-0

LETTER

Ageing behavior of Al–4.5 wt% Cu matrix alloy reinforced with Al_2O_3 and $ZrSiO_4$ particulate varying particle size

Sanjeev Das · S. Das · K. Das

Received: 1 April 2005 / Accepted: 11 July 2005 / Published online: 5 August 2006

Introduction

Age hardening behavior of Aluminum metal matrix composite has been of great interest of present research. The nature of change in kinetics and magnitude of hardening during ageing of composites depends on matrix material [1, 2], type of reinforcement including its size, shape and volume fraction [2, 3], method of synthesizing the composite [2], post fabrication treatment [4, 5] and temperature of ageing [1, 2]. It has been conclusively shown that the presence of ceramic reinforcement such as SiC/Al_2O_3 (whiskers, particle or short fibers) lead to acceleration in the ageing kinetics when compared with the unreinforced alloy [3–13]. This behavior generally has been attributed to enhanced nucleation and growth due to the presence of high matrix dislocation densities, which is generated due to the coefficient of thermal expansion (CTE) mismatch between the matrix and the reinforcements [14]. Matrix dislocations have been found to reduce the incubation time for heterogeneous nucleation of strengthening precipitates, as well as to increase solute diffusivity in the composite matrix. However, it has also been reported that the precipitation sequence was not affected by reinforcement of the aluminum alloy but the volume fraction of GP zones decreased with increasing reinforcement [15, 16]. Selection of ceramic reinforcement in the current research has been constricted to few reinforcement types like Al_2O_3, SiC and B_4C. Limited work has been reported on fabrication and characterization of $ZrSiO_4$ reinforced with aluminum metal matrix composites. $ZrSiO_4$ possess high hardness, high modulus of elasticity, and excellent thermal stability. Banerji et al. found improvement in hardness, abrasive wear resistance, elastic modulus, 0.2% proof stress, and UTS of cast Al–3 wt% Mg alloy due to dispersions of $ZrSiO_4$ particles [17, 18], which makes $ZrSiO_4$ reinforced composite an important material for industries like aerospace, military, automobiles etc. Also, the coefficient of thermal expansion of $ZrSiO_4$ is comparable to that of SiC. Hence the influence of $ZrSiO_4$ reinforcement in ageing behavior of Al–4.5 wt% Cu matrix composite was found to be interesting.

In the present investigation, a comparative study on ageing behavior of Al_2O_3 and $ZrSiO_4$ reinforced in Al–4.5 wt% Cu composite synthesized by stir casting route was carried out. Microstructural characterization has been carried out to investigate the effect Al_2O_3 and $ZrSiO_4$ particulates on the microstructure of Al–4.5 wt% Cu matrix composite. Effect of particle size and type on ageing hardening behavior of Al–4.5 wt% Cu were studied and correlated.

Experimental details

Nominal composition of the alloy matrix used in this study was 4.5 wt% Cu–Al (bal.) and it was reinforced with 15 vol.% of Al_2O_3 and $ZrSiO_4$ particulates. The ceramic particles of size ranges 44–74 μm and 74–105 μm were selected for the experiment. A stir casting setup was used consisting of a resistance furnace and a stirrer assembly as shown in Fig. 1. Al–4.5 wt% Cu alloy was prepared in an induction furnace. Approximately 700 g of Al–4.5 wt% Cu alloy was then remelted at 820 °C in the resistance furnace in a stir casting setup. Preheating of ceramic powders at 450 °C was done in a resistance furnace, placed near stir

S. Das (✉) · S. Das · K. Das
Department of Metallurgical and Materials Engineering, Indian Institute of Technology, Kharagpur, West Bengal 721302, India
e-mail: sanjeev_das2002@yahoo.com

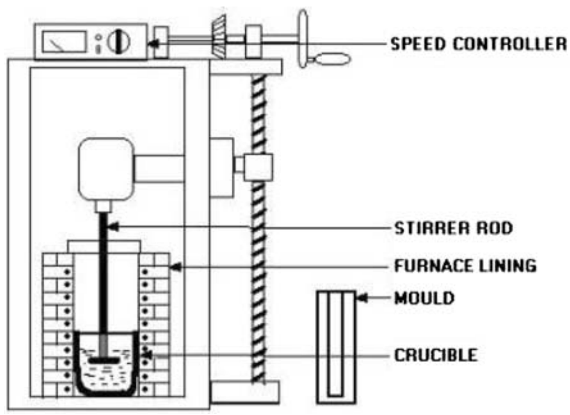


Fig. 1 Schematic diagram of the setup for preparation of particulate composite

casting setup, to remove moisture and gases from the surface of the particulates [19]. The stirrer (graphite) was then lowered vertically up to 20 mm from the bottom of the crucible (total height of the melt was 60 mm). The speed of the stirrer was gradually raised to 600 rpm and the preheated ceramic powders were added with a spoon at the rate of 20–30 g/min into the melt. After addition of ceramic powders, stirring was continued for 10 min for better distribution. The melt was then poured in metal mould of similar dimensions.

Ageing studies were carried out to obtain the peak hardness value with respect to time for the composites reinforced with Al_2O_3 and $ZrSiO_4$ particulates. Identical specimens (10 mm × 5 mm × 2.5 mm) acquired from the ingot were solutionized at 540 °C in a muffle furnace for 2 h, followed by water quenching. Then the samples were immediately kept in refrigerator to avoid diffusion of solute atoms at room temperature. For ageing treatment a low temperature oven has been used. The temperature selected for ageing treatment was 180 °C. The specimens were taken out at different intervals of time and microhardness of the matrix was measured to study the effect of particle size and type on age hardening behavior of the alloy.

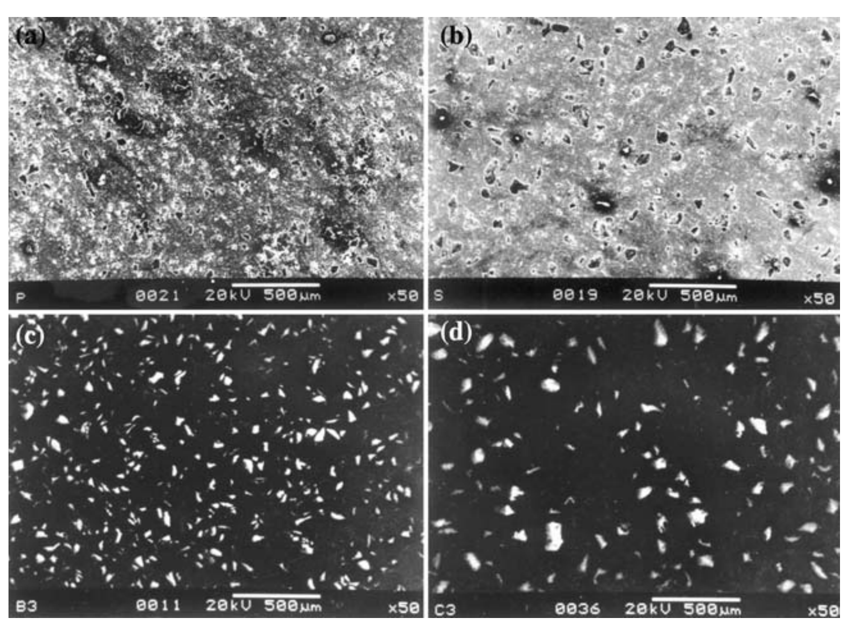
Microstructural characterization studies were primarily accomplished using an optical microscope and scanning electron microscope (SEM) equipped with an energy dispersive spectroscope (EDS). The monolithic alloy and composite samples were metallographically polished and etched (Keller's reagent) before examination.

Results and discussion

Microstructural characterization

Scanning electron micrographs at lower magnification show uniform distribution of Al_2O_3 and $ZrSiO_4$ particulate throughout the alloy matrix (Fig. 2). Scanning electron micrographs at high magnification shows the particle–matrix interfaces (Fig. 3). It is observed that the surface of Al_2O_3 particulates is not as smooth as compared to $ZrSiO_4$ particulates in the alloy matrix. Optical micrographs of the as-cast alloy and composites are shown in (Fig. 4). The grain sizes of composites were found at a reduced proportion compared to monolithic alloy. This can be attributed to the particle restricted growth phenomenon as ceramic particulates slows down the velocity of the solidification front, locally the solidification time increases and more nuclei can form leading to a refinement of grain size [20]. Finer particles can be

Fig. 2 Scanning electron micrographs showing particle distribution of composite reinforced with (**a**) Al_2O_3 particles of size range 44–74 μm, (**b**) Al_2O_3 particles of size range 74–105 μm, (**c**) $ZrSiO_4$ particles of size range 44–74 μm, (**d**) $ZrSiO_4$ particles of size range 74–105 μm



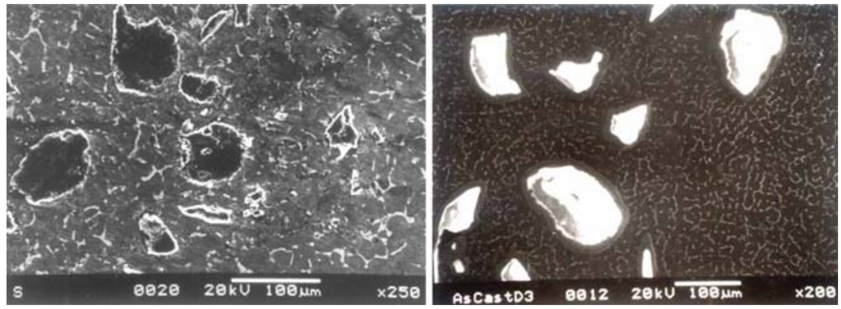


Fig. 3 Scanning electron micrographs showing particle–matrix bonding of (**a**) Al_2O_3 and (**b**) $ZrSiO_4$ reinforced composites

observed near the coarse particle in the case of Al_2O_3 reinforced composite, where as $ZrSiO_4$ reinforced composite does not so such accumulation of finer particles. Although the proper reason for these finer particles are not clear. This can be due to the breakage of Al_2O_3 particles, as CTE of Al_2O_3 is higher than $ZrSiO_4$, effect of thermal fluctuation on Al_2O_3 particle is likely to be more severe during melting and stirring process.

Line profile analysis (LPA) across the particle matrix interfacial region revealed a high concentration of Cu at the interface for both Al_2O_3 and $ZrSiO_4$ reinforced composites (Fig. 5). It can be attributed to the presence of enhanced dislocation density at the interfacial region due to the difference in the thermal coefficient of expansion between the ceramic particle (Al_2O_3 and $ZrSiO_4$) and the alloy matrix, promoting the dislocation-assisted diffusion of alloying elements from the adjacent dislocation-lean areas of the matrix. A similar enrichment of Cu in SiC particulate–matrix (Al–2 wt% Cu) interfacial region has been reported by Gupta et al. [12].

Ageing studies

Figure 6 shows the variation in microhardness as a function of ageing time with varying particle size for Al_2O_3 and $ZrSiO_4$ particulate reinforced composite. It is found that

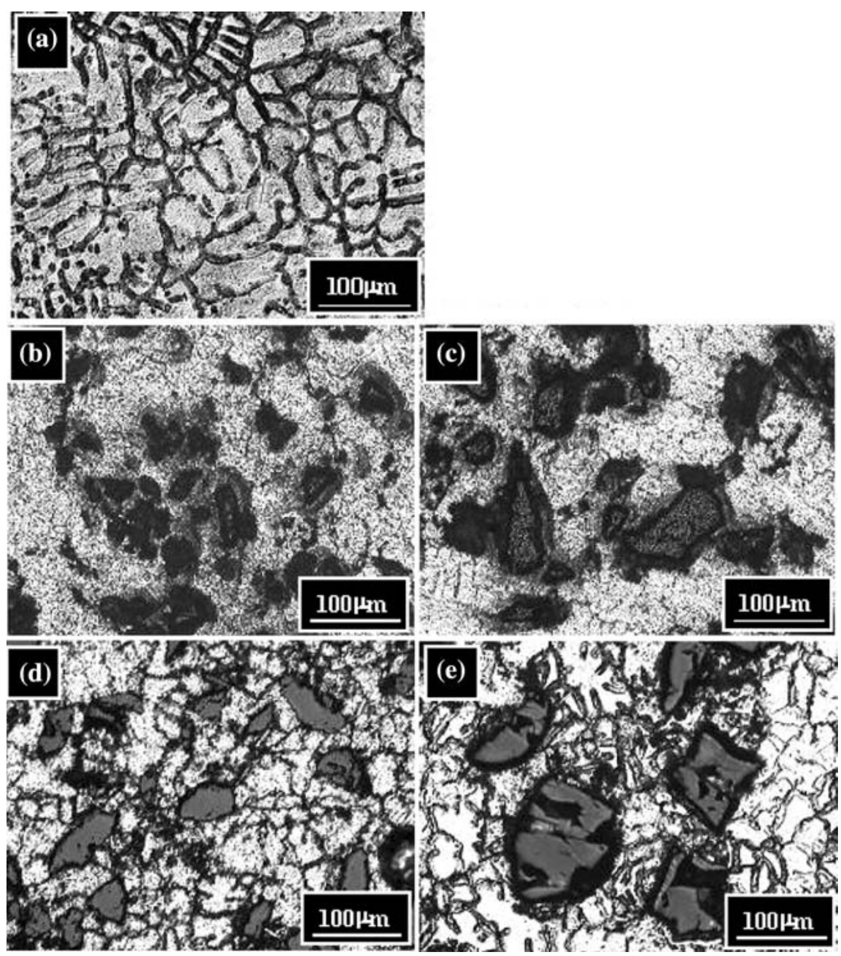


Fig. 4 Optical microstructures of (**a**) monolithic alloy without reinforcement, and composites with (**b**) Al_2O_3 particles of size range 44–74 μm, (**c**) Al_2O_3 particles of size range 74–105 μm, (**d**) $ZrSiO_4$ particles of size range 44–74 μm and (**e**) $ZrSiO_4$ particles of size range 74–105 μm

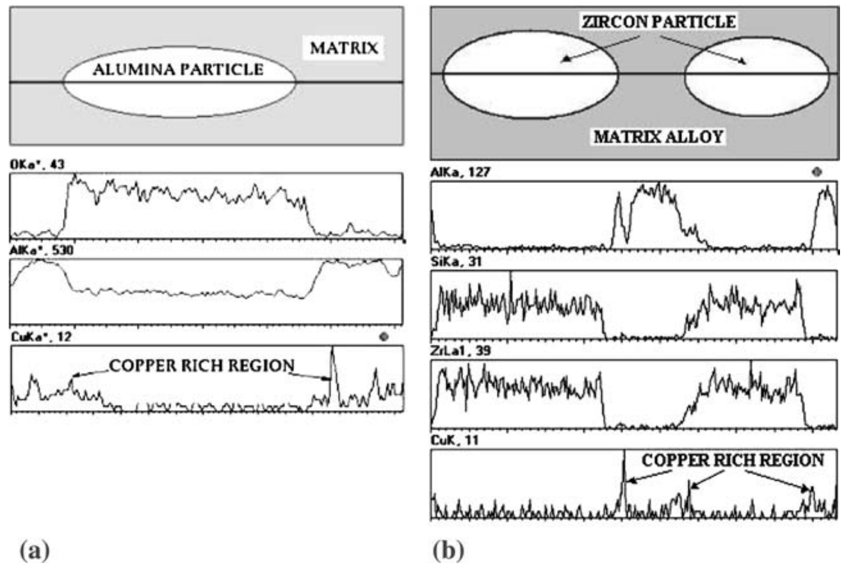


Fig. 5 Line profile analysis (LPA) across particle–matrix interfacial region for (**a**) Al_2O_3 and (**b**) $ZrSiO_4$ reinforced composites

time to attain peak hardness of composites reinforced with Al_2O_3 and $ZrSiO_4$ particles are less compared to monolithic alloy. This can be attributed to matrix dislocations that reduce the incubation time for heterogeneous nucleation of precipitates, as well as increase solute diffusivity in the composite matrix. It is also observed that the decrease in particle size decreases the time to attain peak hardness for both the composites reinforced with different particle type. This is due to high amount of dislocations generated by smaller particles in the matrix as there interparticle distance is lesser compared to that of larger particles that acts as nucleating sites for precipitation. Comparing the nature of the ageing curves it is observed that composite reinforced with smaller particle size have a firm rise in hardness up to peak hardness and then the curve shows a rather constant hardness with respect to ageing time without significant softening. Where as, in the case of composites reinforced with larger particles size, a swift raise and drop in hardness is observed with respect to ageing time. It can be attributed to the increased number of particle–matrix interfaces in the case composite reinforced with fine particles, which generates more dislocation (nucleating sites for precipitation) that accelerates the age hardening and obstructs the softening by restricting precipitates coarsening.

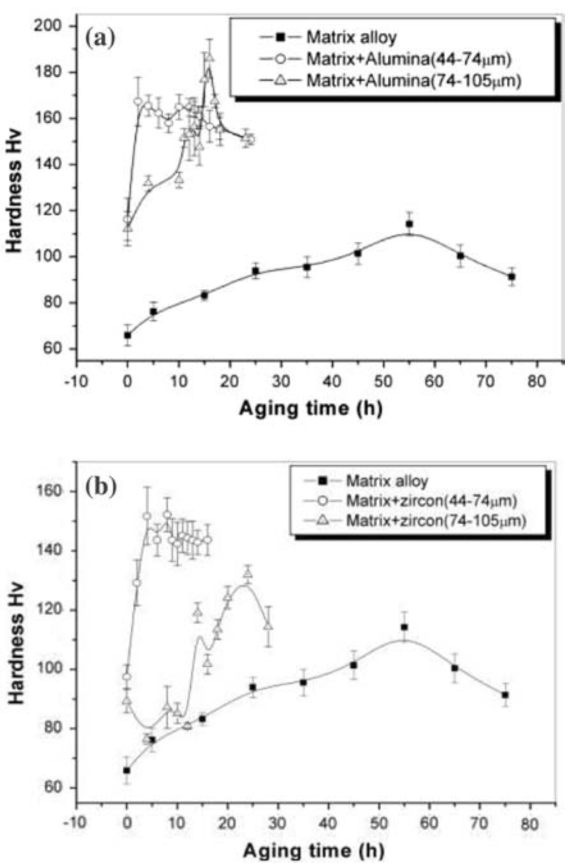


Fig. 6 The variation in microhardness as a function ageing time varying particle size for (**a**) Al_2O_3 and (**b**) $ZrSiO_4$ particulate reinforced composite aged at 180 °C

Figure 7 shows the variation in microhardness as a function ageing time for composites varying particle type of size ranges 44–74 µm and 74–105 µm. It is observed that time to attain peak hardness for composite reinforced with Al_2O_3 particulate was less than composite reinforced with $ZrSiO_4$ particulate. According to earlier studies CTE mismatch between the matrix and the reinforcements is responsible for high matrix a dislocation density that reduces the incubation period for θ'' precipitation [14]. Which implies that if the difference in CTE of particulate and the matrix is higher, more would be the dislocation density and it is supposed to take less time to attain peak

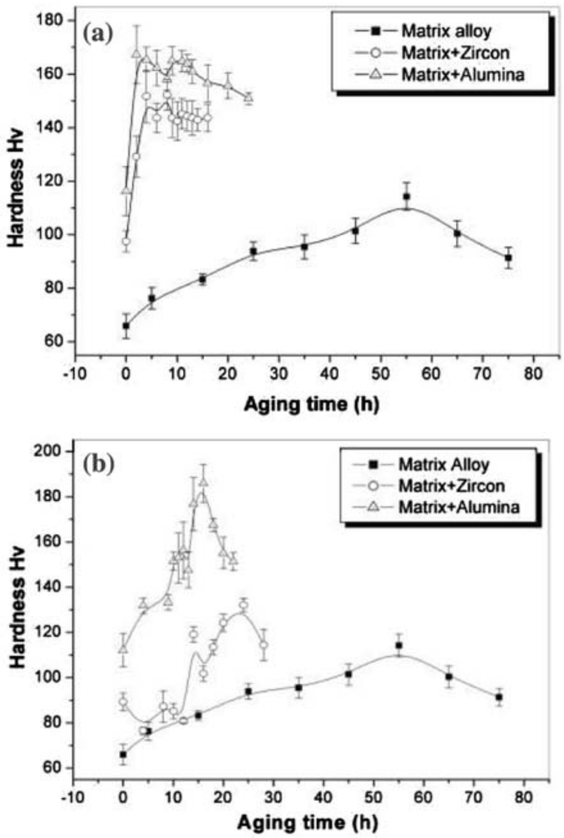


Fig. 7 The variation in microhardness as a function ageing time of composites varying particle type of sizes (**a**) 44–74 μm and (**b**) 74–105 μm aged at 180 °C

hardness. However, the comparison of age-hardening behavior for both the composites shows contradictory results (difference in CTE between Al_2O_3 and Al–4.5 wt% Cu is less than $ZrSiO_4$ and Al–4.5 wt% Cu) as time to attain peak hardness for Al_2O_3 particulate reinforced composite was slightly less than $ZrSiO_4$ particulate reinforced composite. This can be attributed to the accumulation of fine particles formed during synthesis for Al_2O_3 particulate reinforced composite. These fine particles enhance the ageing kinetics of composites reinforced with Al_2O_3 particles compared to $ZrSiO_4$ reinforced composite. Higher hardness of the composite reinforced Al_2O_3 particles compared to $ZrSiO_4$ reinforced composite can be attributed to the affect of particle hardness on the matrix alloy.

Summary

Al_2O_3 and $ZrSiO_4$ can be dispersed uniformly in Al–4.5 wt% Cu alloy by stir casting route. Acceleration in age hardening can be observed in the case of composite reinforced with ceramic particles compared to matrix alloy. Composite reinforced with smaller particle size accelerates age hardening compared to larger particle reinforced composite. It was found that Al_2O_3 reinforced composite show high acceleration in age hardening compared to $ZrSiO_4$ particulates, which is due to the breakage of Al_2O_3 particles during melting and stirring process.

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