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Effect of microstructural features on the ageing behaviour of Al–Cu/SiC metal matrix composites processed using casting and rheocasting routes

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In the present study, microstructural variation in Al–2 wt% Cu/SiC composites was accomplished by synthesizing them using conventional casting and partial liquid phase casting (rheocasting) routes. Microstructural characterization studies conducted on the rheocast composite samples revealed a finer grain size, minimal porosity, uniform distribution of SiC particulates, and a superior matrix − particulate interfacial integrity when compared to the conventionally cast composite samples. Furthermore, the results of interfacial characterization studies revealed that the presence of porosity associated with either individual SiC particulate or SiC clusters significantly influence the constitutional characteristics of the interfacial region. Results of ageing studies revealed an accelerated ageing kinetics in case of rheocast samples when compared to the conventionally cast composite samples. The results of ageing studies were finally rationalized in terms of the difference in microstructural characteristics of the rheocast and conventionally cast composite samples.

1. Introduction

The flexibility associated with tailoring the physical and mechanical properties of metal matrix composites (MMCs) as required by the end application have made them suitable for a spectrum of applications including energy, automobile and aeronautical sectors [1].

The variation in the properties of MMCs can be achieved by a judicious selection of matrix material, type of reinforcement and the heat treatment procedure [2]. The full potential of the metal and ceramic combination, however, is strongly influenced by the processing associated microstructural evolution [3]. The microstructural features of paramount importance in MMCs are: (a) grain size, (b) porosity, (c) distribution of ceramic particulates, (d) interfacial integrity between ceramic particulates and metallic matrix and (e) constitutional characteristics of the interfacial region [1–4]. The impact of these microstructural features in improving the mechanical properties has been extensively reported [1–4]; however, not much information is available regarding their influence on the precipitation behaviour of the strengthening phases.

Accordingly, in the present study Al–2 wt% Cu/SiC composite samples were synthesized using liquid phase (conventional casting) and partial-liquid phase (rheocasting) techniques in order to achieve contrasting microstructural features. The composite samples thus obtained were microstructurally characterized using scanning electron microscopy and heat treated according to conventional T6 heat treatment procedure. Particular emphasis was placed to correlate the effect of microstructural features of the conventionally cast and rheocast composite samples with their respective ageing behaviour.

2. Experimental procedure

2.1. Materials

The nominal composition of the matrix alloy used in the present study was (in wt%): 2.0 Cu–Al (bal). Silicon carbide (α-SiC) particulates with an average size of 23 μm were selected as the reinforcement phase.

2.2. Processing

The synthesis of the metal matrix composites used in the present study was carried out using conventional casting and rheocasting techniques. The synthesis of MMCs using conventional casting was carried out according to the following procedure. The metal ingots, prior to melting, were cleaned using emery papers followed by subsequent washing with water and acetone in order to minimize the thickness of the oxide film and to eliminate other surface impurities. The cleaned metal ingots were melted to the desired superheating temperature (940 °C). SiC particulates, preheated to 900 °C were then added into the molten metal stirred using an impeller. The composite melt thus obtained was poured into cylindrical steel moulds (25 mm diameter, 178 mm height). In all the

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cases, a stirring time of SiC particulates in the melt was maintained between 10 and 15 min. Regarding the rheocasting of MMCs, the synthesis process involved: superheating the properly cleaned metal ingots in a graphite crucible, addition of preheated SiC particulates, in the liquid metallic melt and stirring of the composite mixture in the liquid phase regime and the two phase regime in order to achieve the uniform distribution of SiC particulates in the metallic matrix. The composite material thus obtained was allowed to solidify in the crucible and was subsequently remelted (in the same crucible) followed by casting into cylindrical steel moulds.

2.3. Quantitative assessment of SiC particulates

Quantitative assessment of SiC particulates in the composite samples was carried out using a chemical dissolution method. This method involved: (i) measuring the mass of composite samples, (ii) dissolving the samples in hydrochloric acid, followed by (iii) filtering to separate the ceramic particulates. The particulates were then dried and the weight fraction determined [4].

2.4. Density measurement

Density measurements were carried out in order to ascertain the volume fraction of porosity in the Al–Cu matrix. Density measurements were carried out using Archimedes' principle following the procedure discussed in reference [3].

2.5. Ageing studies

Ageing studies were carried out in order to obtain the peak hardness temperature and time conditions for the conventionally cast and rheocast metal matrix composites. Specimens (25 mm diameter, 7 mm height) taken from as-processed rods were solutionized for one hour at 490 °C, quenched using cold water and aged isothermally at 160 °C for various intervals of time. Rockwell B hardness measurements were made using a 1.58 mm diameter steel ball indenter with a 100 kg load.

2.6. Microstructural characterization

Microstructural characterization studies were conducted on the conventionally cast and rheocast samples in order to investigate the grain size, distribution of ceramic particulates and the presence of porosity. Grain size was measured using the linear intercept method, as described in ASTM E112-88.* Particular emphasis was placed to examine the precipitation behaviour and segregation of copper in the metallic matrix in the immediate vicinity of ceramic particulates.

Microstructural characterization studies were primarily accomplished using an optical microscope and a Jeol scanning electron microscope equipped with EDS (energy dispersive spectroscopy). The composite samples were metallographically polished prior to examination. Microstructural characterization of the samples was conducted in both etched and unetched conditions. Etching was accomplished using Keller's reagent (0.5 HF–1.5 HCl–2.5 HNO_3–95.5 H_2O).

3. Results

3.1. Macrostructure

Macrostructural characterization conducted on the as-processed, machined and polished conventionally cast specimens revealed the presence of macropores and the macrosegregation of SiC particulates. These features, however, could not be detected on the machined and polished surfaces of rheocast composite specimens in the as-processed condition.

3.2. Quantitative assessment of SiC particulates

The results of acid dissolution experiments are summarized in Table I. The weight percentages of SiC particulates was estimated to be approximately 13.9% for conventionally cast composite specimens and 10.9% for the rheocast composite specimens.

3.3. Density measurement

The results of density measurements conducted on the as-processed conventionally cast and rheocast composite specimens revealed density values of 2.57 and 2.75 $g\,cm^{-3}$, respectively. The volume percent of the porosity computed using the experimentally determined density values and the results of acid dissolution tests are shown in Table I.

3.4. Ageing studies

The results of ageing studies conducted on the conventionally cast and rheocast composite samples are shown in Fig. 1. The results exhibit the presence of a well-defined hardness peak at 9 h for the conventionally cast composite samples and at 6 h for the rheocast composite samples. The results also reveal that the maximum peak hardness is achieved in the rheocast composite samples followed by the conventionally cast composite samples.

3.5. Microstructural characterization

Grain size determination conducted using optical micrographs taken from the etched composite samples revealed an average grain size of 59.4 ± 4.9 μm for

TABLE I Results of acid dissolution tests and volume percent porosity determination

Matrix	Condition	Reinforcement size	Weight % SiC	Vol% porosity
Al–Cu	As-cast	23 μm	13.9	7.9
Al–Cu	As-rheocast	23 μm	10.9	0.8

* 1992 Annual Book of ASTM Standards, Vol. 03.01, Section 3, Metals Test Methods and Analytical Procedures, pp. 294–319, ASTM, PA, USA.

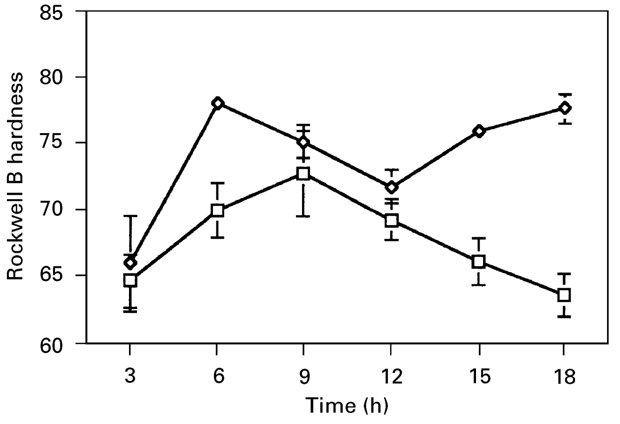

Figure 1 Graphical representation of ageing studies conducted on conventionally cast Al–Cu/SiC samples (□) and rheocast Al–Cu/SiC samples (◇).

the conventionally cast composite samples and 42.6 ± 4.9 μm for the rheocast composite samples.

The results of scanning electron microscopy conducte on conventionally cast Al–Cu/SiC samples revealed a partly dendritic and partly equiaxed matrix microstructure. The interdendritic/intercellular regions were found to be frequently associated with the presence of Cu rich phases (see Fig. 2a). The metallic matrix also revealed the presence of porosity predominantly associated with the individual SiC particulates at the angular locations and with SiC clusters (see Fig. 2b). The distribution of SiC particulates in the cast composite samples can be assessed from Fig. 2a. Predominantly SiC particulates were present in the form of small clusters preferentially located at the grain boundaries. The interfacial integrity between SiC particulates and Al–Cu matrix was found to be poor and in some cases a partially debonded interface was observed in the cast composite samples (see Fig. 2c). In addition, the interface formed between the SiC particulates and Al–Cu matrix also revealed the presence of secondary phases. EDX (energy dispersive analysis by X-ray) analyses carried out at various locations in the intermediate vicinity of: (a) completely integral matrix (Al–Cu) particulate (SiC) interface, (b) partially debonded SiC particulates, and (c) SiC clusters revealed the enrichment of Cu. The extent of segregation of the copper in the interfacial locations was observed to be maximum in the case of the SiC particulates forming a completely integral interface with the metallic matrix (see Fig. 3).

Finally the results of scanning electron microscopy conducted on the rheocast samples revealed the presence of: partially columnar and partially equiaxed matrix microstructure (see Fig. 4a), a minimal amount of porosity, Cu-rich phases at the interdendritic/intercellular regions, relatively more uniform distribution of SiC particulates (see Fig. 4b) and good interfacial integrity between SiC particulates, and the metallic matrix (see Fig. 4c). Microstructural characterization results also revealed a relatively smoother surface of the SiC particulates when compared to that observed in conventionally cast composite samples. In addition, the results of EDX analyses revealed the copper content to decrease with increasing distance from the interface (see Fig. 5).

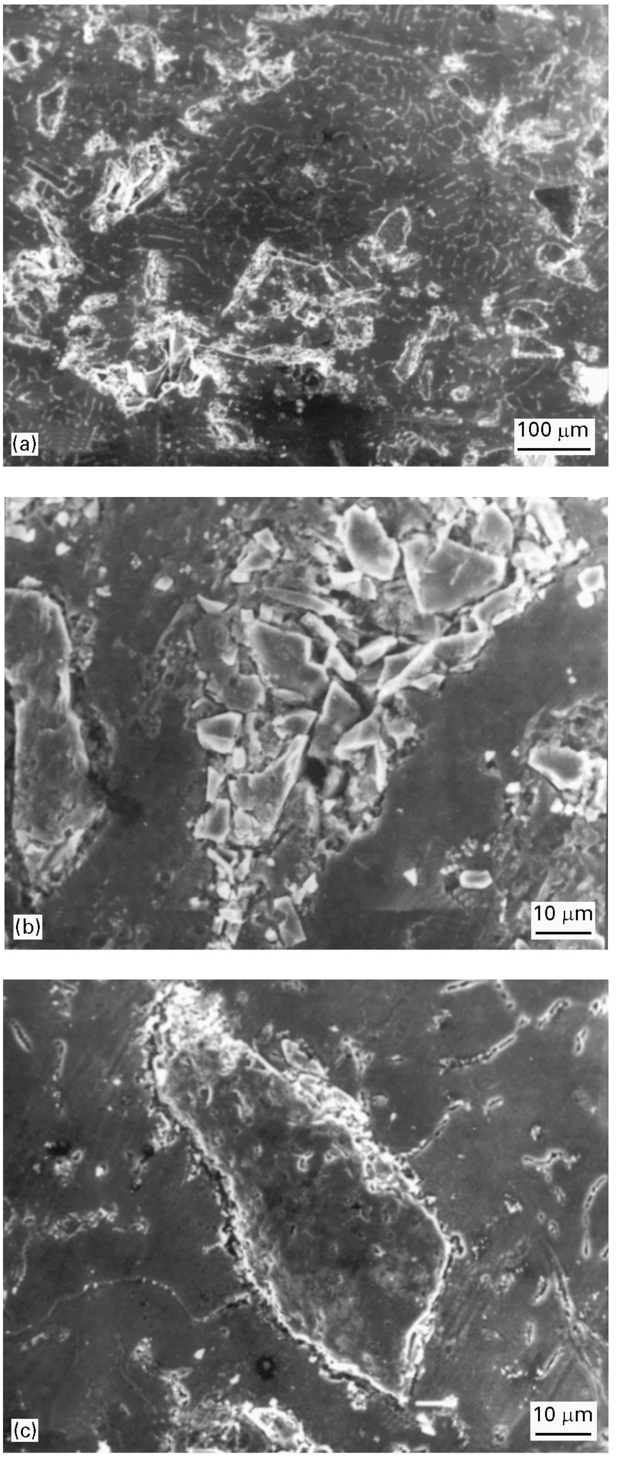

Figure 2 Representative SEM micrographs showing: (a) matrix microstructure and distribution of SiC particulates, (b) SiC particulates associated porosity, and (c) interfacial integrity between a SiC particulate and Al–Cu metallic matrix, in conventionally cast composite samples.

4. Discussion

4.1. Microstructure

The microstructure of the conventionally cast and rheocast samples revealed three common salient features:

(a) presence of partially columnar and partially equiaxed matrix microstructure,
(b) presence of porosity, and
(c) presence of interdendritic Cu-rich phase.

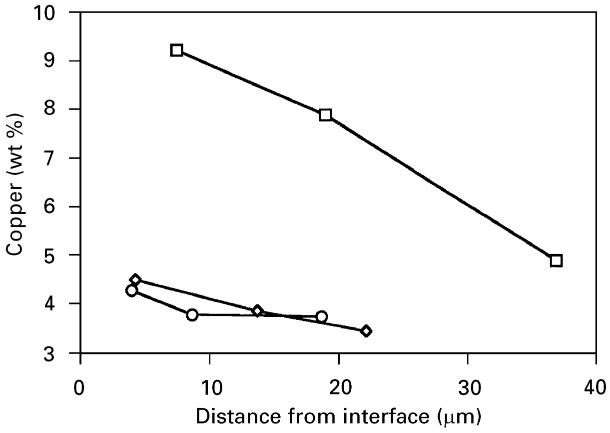

Figure 3 Graphical representation of the segregation pattern of Cu observed in the near vicinity of completely bonded (□), partially debonded (◇) and SiC clusters (○) associated with porosity in case of conventionally cast Al–Cu/SiC samples.

The grain structure of the conventionally cast and rheocast samples was found to be partially columnar and partially equiaxed (see Figs 2a and 4a). The underlying principles behind the development of this type of structure are well established and can be found elsewhere [5]. The decrease in grain size observed in case of rheocast samples may be attributed to:

(i) an increase in the grain nucleation sites during solidification, and
(ii) retardation of grain growth during solid state cooling.

The increase in grain nucleation site can be attributed to the presence of broken dendrite fragments resulting due to stirring of the metallic melt in the partial liquid phase regime. In related studies conducted on Al–7Si–0.6 Mg alloy, investigators reported similar refinement in the grain structure of the rheocast samples when compared to the conventionally cast samples [6]. In addition, the finer grain size observed in case of rheocast samples can also be attributed to the more effective pinning of grain boundaries by uniformly distributed SiC particulates in the matrix during solid state cooling of the rheocast composite samples [4].

Another important microstructural feature observed in case of conventionally cast and rheocast samples investigated in the present study was the presence of non-interconnected and randomly distributed porosity. Two types of porosity were observed in the present study:

(i) microporosity in the bulk metallic matrix in case of rheocast and conventionally cast composite samples, and
(ii) porosity associated with the individual and clustered SiC particulates in case of conventionally cast composite samples (see Fig. 2b).

The mechanisms related to formation of microporosity associated with columnar-equiaxed type of solidification microstructure as observed in case of rheocast and conventionally cast composite samples are well established and will not be reiterated here [5].

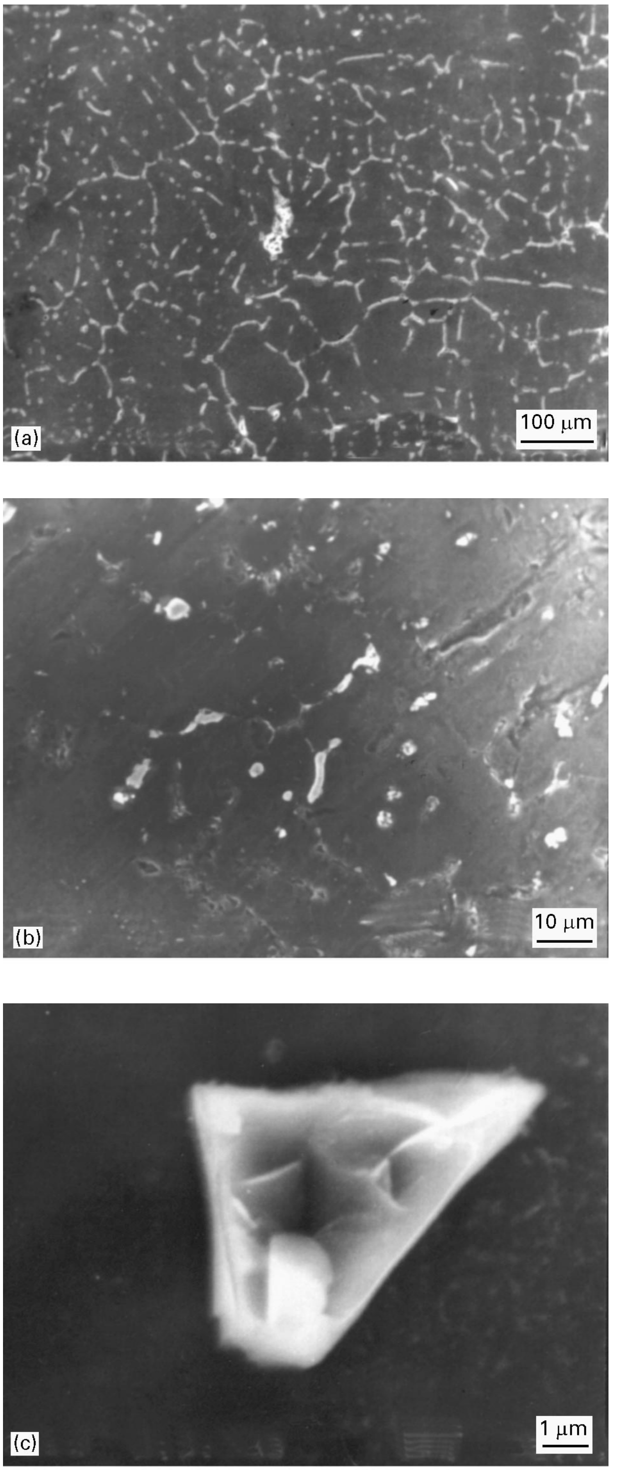

Figure 4 Representative SEM micrographs showing: (a) matrix microstructure, (b) distribution of ceramic particulates and (c) interfacial integrity between a SiC particulate and Al–Cu metallic matrix, in rheocast composite samples.

The presence of porosity at the sharp corners of SiC particulates in the case of conventionally cast composite samples can be primarily attributed to the inability of the high viscosity particulate containing metallic slurry to negotiate sharp corners, while the presence of voids within ceramic particulate clusters can be attributed to the inability of particulate containing liquid metallic slurry to infiltrate the micrometre sized crevices in the inefficiently packed ceramic particulates' clusters which are formed ahead of the

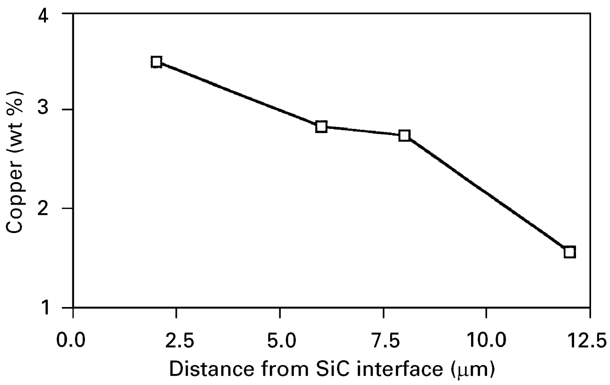

Figure 5 Graphical representation of a typical segregation pattern of Cu observed in the near vicinity of SiC particulates in case of rheocast Al–Cu/SiC samples.

moving solidification front during conventional casting [7].

Regarding the amount of porosity, the results of the present study revealed that the volume percent of porosity was less than 1% in case rheocast composite samples when compared to about 7.9% determined for the conventionally cast composite samples (see Table I). The lower volume percent of porosity observed in the rheocast composite samples can be attributed to the improved fluid flow conditions as a result of more uniform distribution of SiC particulates in the molten metallic matrix during casting when compared to the conventional casting technique. Further work is continuing in this area.

The presence of the interdendritic/intercellular Cu-rich phase observed in case of conventionally cast and rheocast composite samples (see Figs 2a and 4a) can be attributed to the sluggish solidification front velocity achieved during primary processing of materials, rejection of Cu ahead of the moving liquid–solid interface and subsequent solidification when the temperature of the remaining liquid reached the eutectic temperature (548 °C) [3, 5, 8].

4.2. Amount and distribution of SiC particulates

In the present study, 13.9 and 10.9 wt % percent of SiC particulates were successfully incorporated in Al–2 wt% Cu metallic matrix using conventional casting and rheocasting techniques respectively. The successful incorporation of SiC particulates in the limits exceeding 10 wt% using these techniques can be attributed to the preheating of SiC particulates to 900 °C prior to the addition in the liquid metallic melt. Preheating of SiC particulates has been shown to assist in: (i) removing surface impurities, (ii) desorption of gases, and (iii) altering the surface composition due to the formation of a thin oxide layer (SiO_2) on the surface [9]. The ability of the oxide layer to improve the wettability of SiC particulates by alloy melt has previously been suggested by other investigators [10, 11].

Regarding the distribution of SiC particulates, the following comments are important. The segregated distribution of SiC particulates (see Fig. 2a) in the case of cast composite samples is consistent with the results of other investigators and was attributed to the sluggish solidification front velocity commonly associated with the casting route [3, 7]. In the case of rheocast composite samples, the improved distribution of SiC particulates can be attributed to the enhanced mechanical interaction between SiC particulates and the broken dendrite fragments resulting from the stirring of the partial liquid phase melt. The presence of dendrite fragments thus assists in better dispersion of reinforcing particulates and prevents settling of SiC particulates [6, 12, 13].

4.3. Interfacial characteristics

The results of microstructural characterization studies conducted on the particulate/matrix interfacial region revealed a high concentration of Cu in both conventionally cast and rheocast composite samples (see Figs 3 and 5). This phenomenon can primarily be attributed to the presence of enhanced dislocation density in the interfacial region. The enhanced dislocation density results due to the difference in coefficient of thermal expansion between SiC particulates and the aluminium matrix [14] and promotes the dislocation-assisted diffusion of the alloying elements from the adjacent dislocation lean areas of the matrix [4, 14]. In conventionally cast composite samples, a decrease in extent of segregation of Cu in case of partially debonded (or porosity associated) SiC particulates and the clustered SiC particulates indicate the diminishing ability of SiC particulates to generate misfit strains and to punch out dislocations in the matrix. Furthermore, the reduced extent of segregation of copper adjacent to completely bonded SiC particulate in case rheocast samples can be attributed to the presence of other segregation sites in the matrix such as increased grain boundary area besides SiC particulates. The grain size refinement of the rheocast samples when compared to the conventionally cast samples has been previously reported [6] and is also consistent with the experimental results obtained in the present study on the composite samples. Further work is continuing in quantifying the size and distribution of the secondary phases in the matrix.

Another important microstructural characteristic associated with the Al–Cu/SiC interfacial region in the case of conventionally cast composite samples was the presence of Al–Cu based secondary phases (see Fig. 2c). The presence of these secondary phases can be attributed to the heterogeneous nucleation ability of the jagged surface of SiC particulates and the dislocations defect structure in the metallic matrix adjacent to the SiC particulates [15]. Further work is continuing in order to identify the composition and structural aspects of these secondary phases.

4.4. Ageing studies

The results of ageing studies conducted on the conventionally cast and rheocast composite samples revealed:

(a) higher peak hardness achieved by rheocast samples when compared to conventionally cast composite samples, and
(b) an accelerated ageing kinetics exhibited by rheocast samples when compared to conventionally cast composite samples.

The higher peak hardness of the rheocast composite samples containing a lower weight percent of SiC particulates can primarily be attributed to a more refined microstructure and 0.8 vol% porosity when compared to 7.9 vol% determined for conventionally cast composite samples (see Table I).

The accelerated ageing kinetics observed in case of rheocast composite samples aged at an identical ageing temperature and containing less weight percent of SiC particulates when compared with conventionally cast composite samples is strongly indicative of the matrix microstructural variation brought about by the rheocasting technique. Based on the results of microstructural characterization obtained in the present study, the accelerated ageing kinetics exhibited by rheocast composite samples can be attributed to:

(a) decrease in grain size,
(b) relatively non-existent SiC particulates associated porosity, and
(c) minimal presence of SiC particulates' clusters.

The accelerated ageing kinetics of rheocast composite samples can be explained, in part, considering the increase in grain boundary area as a result of the decrease in grain size of the metallic matrix when compared to the conventionally cast composite samples. An increase in grain boundary area assists in increasing the frequency of nucleation of the strengthening phases as a result of the reduced activation barrier for the heterogeneous nucleation [15]. The experimental confirmation of the heterogeneous nucleation of the precipitates at the grain boundaries in case of Al matrices has been established previously [4, 16].

The accelerated ageing kinetics of the rheocast samples can also be attributed to the absence of particulates' associated porosity. An integral matrix–particulate interface ensures the sequential events involving: generation of misfit strains due to difference in coefficient of thermal expansion between SiC particulates and matrix [14, 17]; punching of dislocations in the matrix [14]; and the subsequent heterogeneous nucleation of the strengthening phases in the matrix. The presence of porosity at the particulates' interface as observed in case of conventionally cast composite samples, on the contrary, will diminish the magnitude of misfit strains as a result of the ability of SiC particulate to adjust itself in the metal free space (porosity) during quenching. A reduction in magnitude of misfit strains will be instrumental in reducing the average dislocation density and hence the heterogeneous nucleation volume around the particulates [14, 17, 18] thus retarding the rate of precipitation. This is consistent with the EDX results obtained in the present study showing a relatively lesser extent of segregation of copper in the near vicinity of porosity-associated SiC particulate (see Fig. 3). Finally, the accelerated ageing kinetics exhibited by the rheocast samples can also be attributed to the minimal presence of SiC clusters. In case of conventionally cast composite samples, for example, SiC clusters were observed more frequently and commonly associated with porosity (see Fig. 2b). The presence of porosity in the inefficiently packed SiC clusters enables the individual SiC particulates to readjust themselves when acted upon by the compressive forces during quenching from the solutionizing temperatures. This flexibility associated with the SiC particulates to readjust themselves will reduce the extent of misfit strains and hence the punched out dislocations in the matrix. It may be noted that the diminishing ability of SiC particulates to punch dislocations when present in the form of clusters will relatively reduce the precipitation of strengthening phases as a result of: (a) lowering the segregation of copper in the matrix adjacent to the SiC particulates (see Fig. 3), and (b) reducing the heterogeneous nucleation sites in the form of dislocations [14] thus lowering the ageing kinetics.

The results of the ageing studies thus clearly indicate that the increase in ageing kinetics of the rheocast composite samples can be attributed to an increase in number of heterogeneous nucleation sites as a result of decrease in grain size, uniform distribution of SiC particulates, absence of SiC clusters and a relatively non-existent porosity associated with SiC particulates.

5. Conclusions

The primary conclusions that may be derived from this work are as follows:

1. Aluminium based MMCs containing in excess of 10 wt% SiC particulates can be successfully synthesized using conventional casting and the rheocasting routes used in the present study.
2. Rheocasting procedure used in the present study results in a finer grain size, minimal porosity, relatively uniform distribution of SiC particulates and a superior interfacial integrity between SiC particulates and the metallic matrix when compared to the conventionally cast composite samples.
3. The accelerated ageing kinetics exhibited by the rheocast composite samples can be attributed to an increase in grain boundary area, uniform distribution of SiC particulates. and a relative absence of porosity associated with SiC particulates when compared to the conventionally cast material.

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