

## MICROSTRUCTURE AND INTERFACE CHARACTERISTICS OF B<sub>4</sub>C, SiC AND Al<sub>2</sub>O<sub>3</sub> REINFORCED Al MATRIX COMPOSITES: A COMPARATIVE STUDY

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**Abstract** Three aluminium metal matrix composites containing reinforcing particles of B<sub>4</sub>C, SiC and Al<sub>2</sub>O<sub>3</sub> (0-20 vol. %) were processed. The stir-casting manufacturing route followed by hot extrusion was utilized, being one of the cost-effective industrial methods. In this study, the feasibility of processing B<sub>4</sub>C reinforced Al composite was investigated and a comparison was made with the other two composites. The microstructural distribution of reinforcing particles in all three composites was studied by means of optical and scanning electron microscopy (SEM). The distribution and chemical composition of the phases formed at matrix/particulate interface of the processed composites were also investigated by SEM and energy dispersive x-ray spectroscopy (EDX). A clear interfacial reaction product/layer was found at Al/SiC interface for composites held for a relatively long processing time (> 30 minutes). No reaction product was observed at Al/B<sub>4</sub>C and Al/Al<sub>2</sub>O<sub>3</sub> interfaces at the resolution limit of the SEM used. On the other hand, two secondary phases (alumina and another phase containing aluminium, boron and carbon) were found in the aluminum matrix away from the interface in Al-B<sub>4</sub>C composites. From the fracture surface analysis, B<sub>4</sub>C reinforced Al composite seemed to exhibit a better interfacial bonding compared to the other two composites.

**Keywords:** Metal matrix composites, stir casting, interface, secondary phase.

### INTRODUCTION

Aluminium metal matrix composites (Al MMCs) are being considered as a group of new advanced materials for its light weight, high strength, high specific modulus, low co-efficient of thermal expansion and good wear resistance properties. Combination of these properties are not available in a conventional material [Surappa and Rohatgi, 1981]. The use of Al MMC has been limited in very specific applications such as aerospace and military weapon due to high processing cost. Recently, Al matrix composites have been used for the automobile products such as engine piston, cylinder liner, brake disc/drum etc. [Schumacher, 1991]. Processing techniques for Al MMCs can be classified into (1) liquid state processing, (2) semisolid processing and (3) powder metallurgy [Kozack *et al.*, 1993 and Srivatsan *et al.*, 1991]. Particulate reinforced Al composites can be processed more easily by the liquid state i.e. melt-stirring process. Melt stir casting is an attractive processing method since it is relatively inexpensive and offers a wide selection of materials and processing conditions.

The primary function of the reinforcement in metal matrix composites is to carry most of the applied load, where the matrix binds the reinforcements together, and transmits and distributes the external loads to the individual reinforcement [Rajan *et al.*, 1998]. Good wetting is an essential condition for the generation of a satisfactory bond between particulate reinforcements and liquid Al metal matrix during casting composites, to allow transfer and distribution of load from the matrix to the reinforcements without failure. Strong bonds at the interface are required for good wetting. These bonds may be formed by mutual dissolution or reaction of the particulates and matrix metal. The reaction phenomena are very detrimental to the composite as they bring about a decrease of the mechanical properties [Delannay *et al.*, 1987].

Substantial information is available in literatures on wetting and interface of Al-alloy metal matrix composites reinforced with SiC, Al<sub>2</sub>O<sub>3</sub> particulates [Gui *et al.*, 2000, Laurent *et al.*, 1987, and Kobashi, 1993]. Al/SiC system is a reactive system, as it produces Al<sub>4</sub>SiC<sub>4</sub> or

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**Table-1: Showing the melt temperature during particle addition and before pouring, and total stirring time**

Composites	Volume fraction	Melt temperature during particle addition, °C	Melt temperature before pouring, °C	Stirring time during particle addition, min	stirring time after particle addition, min
Al-SiC	6%	770	710	17	10
	13%	770	710	30	10
	15%	770	710	45	60
	20%	770	710	50	10
Al-Al <sub>2</sub> O <sub>3</sub>	13%	810	670	35	16
	20%	810	670	50	16
Al-B <sub>4</sub> C	13%	770	670	30	16

Al<sub>4</sub>C<sub>3</sub> compound at the interface of particles and metal [Oh et al., 1989]. Al<sub>4</sub>C<sub>3</sub> is detrimental for the composites properties. The formation of Al<sub>4</sub>C<sub>3</sub> can be minimized in several ways such as (1) using a suitable coating on particles, (2) using high silicon content Al alloys or (3) using pre-oxidized silicon carbide particulates [Rajan et al., 1998]. The only reaction at the interface of Al/Al<sub>2</sub>O<sub>3</sub> composites is Al<sub>2</sub>O<sub>3</sub> dissolving into aluminium. Small addition of Mg encourages the formation of MgAl<sub>2</sub>O<sub>4</sub> spinel with Al<sub>2</sub>O<sub>3</sub>. Some studies on the reactivity B<sub>4</sub>C in aluminium processed by infiltration and powder metallurgy techniques reported the formation of different compounds at different processing temperature [Oh et al., 1989, Viala et al., 1997 and Marchi et al. 2000]. Good wettability of B<sub>4</sub>C in aluminium has been found in air due to the formation of boron oxide film around the particles [Oh et al., 1989]. Literature related on the microstructure and interface of Al/B<sub>4</sub>C composites processed by a stir cast method in air is very scarce.

The main aims of the present work were to see the feasibility to produce Al/B<sub>4</sub>C composites by using conventional liquid melt stirring process in air. For the comparison of the microstructure (particles distribution and pores) and interface of Al/B<sub>4</sub>C composites, other two composites Al/SiC and Al/Al<sub>2</sub>O<sub>3</sub> were produced by using the melt stirrer method. The microstructure and interfaces were studied using optical microscopy (OM), scanning electron microscopy (SEM) and energy dispersive x-ray spectroscopy (EDX).

## MATERIALS AND EXPERIMENTAL

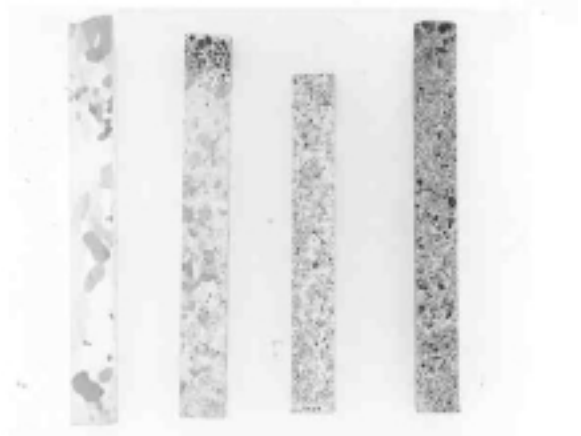
Three different ceramic particles SiC, Al<sub>2</sub>O<sub>3</sub>, B<sub>4</sub>C were used as reinforcement in pure aluminum (99.99) matrix. Commercial β- and α-type silicon carbide particles with an average particle size of 40μm, α-Al<sub>2</sub>O<sub>3</sub> particles of 32μm size and B<sub>4</sub>C particles of 40μm size were used in this study.

An electrical resistance furnace with a stirring assembly (a graphite impeller) was used for the dispersion of the ceramic particles into liquid aluminum. SiC and Al<sub>2</sub>O<sub>3</sub> particles were preheated at 900°C for 1 and 2 hours in air respectively to increase their surface reactivity [Surappa and Rohatgi, 1981]. B<sub>4</sub>C was

preheated at 175°C for 2 hours to remove moisture. The heat-treated particles were then added into the melt through the vortex. Finally the melt was poured into a 52mm internal dia cylindrical graphite mould. The melt temperature during particle addition and before pouring into mold, and total stirring time (stirring during and after particle addition) for some composites are presented in Table 1. Various compositions ranging from 0-20vol% of Al/SiC and Al/Al<sub>2</sub>O<sub>3</sub>, and 0-13vol% Al/B<sub>4</sub>C composites were processed.

To reduce the porosity of the cast composites and to improve the particle distribution within Al matrix, the cast composites were subjected to hot extrusion at 420°C with an extrusion ratio of 27 at a speed of 1m/min. Hot extruded composites of 10mm dia bar were then heat treated at 400°C for 2 hours and cooled in furnace to remove the extrusion effect.

The microstructure was investigated by optical and scanning electron microscope (Philips XL 30 FEG) equipped with energy dispersive x-ray spectroscopy (EDX). Fracture surface of some of the composites were analyzed by the SEM.



**Fig. 1 Macrostructure revealing pores distribution in cast Al-SiC composites (from left: 0, 6,13 and 20Vol% SiC)**

## RESULTS AND DISCUSSION

Fig.1 displays optical micrographs taken at low magnification from pure Al and Al/SiC (6, 13 and 20 vol. %) composites sectioned in the longitudinal direction showing distribution of pores. It is seen that the grain size of aluminium with 0% SiC is quite large. With the introduction of SiC particles, the grain size decreases. At the same time, pores seen as dark spot in Fig. 1 increases in number as more SiC particles are introduced the cast composites. This is due to the fact that the addition of particles while stirring liquid Al introduces air since particles usually enter the melt as a group of particles trapping air in between them. This fact has also been reported elsewhere [Surappa and Rohatgi, 1981].

Therefore as the amount of added SiC particles increases, the amount of trapped air increases thereby increasing the amount of pores. Large pores observed in Al-20SiC contain a cluster of SiC particles which did not get dispersed within the Al matrix during processing. During cutting and grinding, the cluster of particles is pulled out and an empty space appeared as a pore, as shown in Fig. 1. The porosity of the castings during processing can be reduced in several ways (e.g. use of inert atmosphere or vacuum, reduce the vortex motion and turbulent mixing using baffles, use appropriate stirrer speed, size and position) [Surappa and Rohatgi, 1981]. The macrostructure of Al-Al<sub>2</sub>O<sub>3</sub> system looks similar to that of Al-SiC in terms of pores,

with slightly lower amount of overall porosity. The reason of slightly lower porosity in Al/Al<sub>2</sub>O<sub>3</sub> composites as compared to Al/SiC system is not clear. The macrostructure of Al-B<sub>4</sub>C system presented a better result with respect to porosity than both above systems. Since B<sub>4</sub>C is very reactive to air at elevated temperature, the air trapped with B<sub>4</sub>C particles reacts with B<sub>4</sub>C inside the melt [Oh *et al.*, 1989]. This is thought to result in a lower amount of pores.

Fig. 2 shows the microstructure of Al-13SiC, Al-13Al<sub>2</sub>O<sub>3</sub> and Al-13B<sub>4</sub>C cast composites taken from a transverse section cut at about 7mm from the bottom of the cast. The particles are reasonably well distributed within Al matrix with some macropores trapped in the center of a cluster of the particles while micropores are found in the Al matrix. Among the three composites, particle distribution in Al-B<sub>4</sub>C composites is found to be better. This is due to the fact that the wetting of ceramics particles in aluminium controls the quality of Al MMC castings. The wettability of B<sub>4</sub>C with aluminium was found to increase in air. This was attributed to the formation of a layer of liquid B<sub>2</sub>O<sub>3</sub> on the B<sub>4</sub>C particulates. Due to its low melting point, B<sub>2</sub>O<sub>3</sub> exists above 450°C as a liquid on the surface of B<sub>4</sub>C and enhances wettability through a liquid-liquid reaction when contacted with aluminium liquid by forming B<sub>2</sub>O<sub>3</sub>.Al<sub>2</sub>O<sub>3</sub> oxide compound [Oh *et al.*, 1989]. As the composites were processed in air, the distribution of particles in Al-B<sub>4</sub>C is found better than Al-SiC and Al-Al<sub>2</sub>O<sub>3</sub>. A reduction of porosity and a better

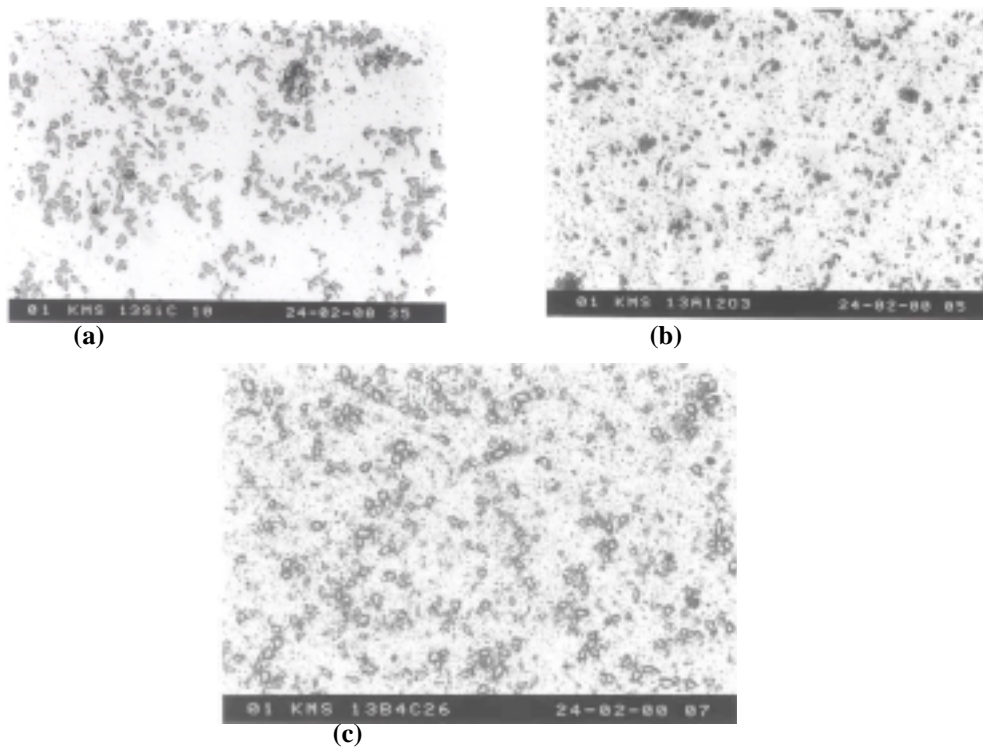


Fig. 2 Optical micrographs showing the distribution particles in as-cast (a) Al-13vol%SiC, (b) Al-13vol%Al<sub>2</sub>O<sub>3</sub> and (c) Al-13vol%B<sub>4</sub>C composites.

distribution of particles were achieved in all composites after hot extrusion.

A interaction layer was found at the interface of Al/SiC composites, none was found at the interface of Al/Al<sub>2</sub>O<sub>3</sub> and Al/B<sub>4</sub>C composites (Fig. 3). Total interaction time (particle mixing time + stirring time) between particles and liquid aluminium before pouring was 60 minutes for Al-SiC (Fig. 3a), 66 minutes for Al-Al<sub>2</sub>O<sub>3</sub> (Fig. 3b) and 30 minutes for Al-B<sub>4</sub>C (Fig. 3c). The thickness of the interaction layer present at Al/SiC interface increases with the interaction time. This is clear in Fig. 4 displaying the micrographs of two composites, Al-6SiC and Al-15SiC with increasing thickness of the interaction layer. While the Al-6SiC composite was stirred for 27 min before pouring into mould, and Al-15SiC composite was stirred for 90 minutes. Although no interaction layer could be observed in Al-6SiC cast composites as shown in Fig. 4a, we may not rule out the presence of a thin layer at the interface that could not be detected at this magnification. The thickness of the interaction layer is up to 3 μm for Al-15SiC (Fig. 4b).

To analyze the phases formed at the Al/SiC interface, a spot analysis was taken from the center of the interaction layer formed in Al-15SiC composite (Fig. 4b). The EDX spectrum reveals the presence of the elements Al, Si, C and O in the interface reaction layer (Fig. 5). It is believed that the small amount of oxygen detected by EDX is probably coming from an oxide layer formed during sample preparation. It is also clear from EDX spectrum that the Al peak is around four times higher than the Si peak. X-ray elemental mapping performed at the Al/SiC interface in Al-15%SiC composites shows both Al and Si changing gradually from matrix to particle and particle to matrix respectively as displayed in Fig. 6. Two probable reactions may take place between aluminium and SiC. Some studies [Indue *et al.*, 1980 and Schoennahl *et al.*, 1984] reported the formation of a ternary compound in a Al/SiC system according to the following reaction.



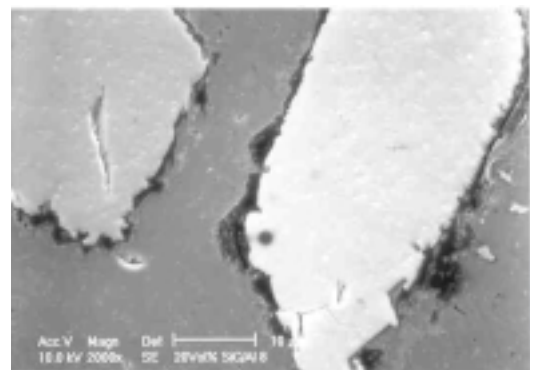
Others [Lloyd, 1989, and Lloyd *et al.*, 1989] suggested the formation of Al<sub>4</sub>C<sub>3</sub> compound at the interface:



From the EDX analysis results and the peak ratio of Al to Si it is suggested that the phase formed at the interface of Al/SiC in the present system is a ternary compound Al<sub>4</sub>SiC<sub>4</sub>. It may be noted that the phase which formed as a reaction product is brittle as evidenced by a few cracks going through this phase in a catastrophic manner (Fig. 4b). It should be mentioned that this secondary phase is not present around all SiC particles in Al-20SiC composite (60 minutes interaction

time), but rather around some SiC particles which were probably added at the beginning and thus had a longer interaction time with the melt.

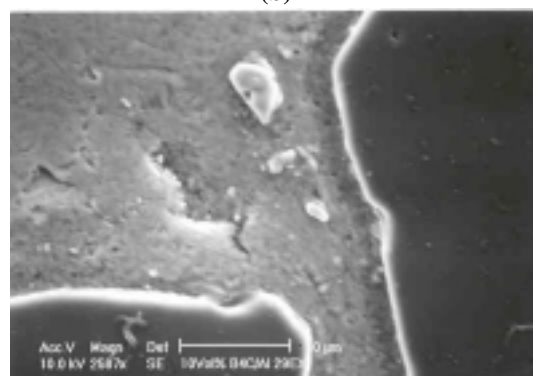
In addition to this ternary compound forming the interaction layer, small white particles, identified as Al<sub>2</sub>O<sub>3</sub>, can be seen in Fig. 4b ahead of this interaction layer. It is expected that the contact between the SiC particles and Al melt would result in an interaction layer which improves wetting between the two constituents. The type of interaction layer depends on the elements present at the interface during processing. In this study, SiC particles were heat-treated at 900°C in air for 1 hour in order to increase their reactivity with Al melt by



(a)

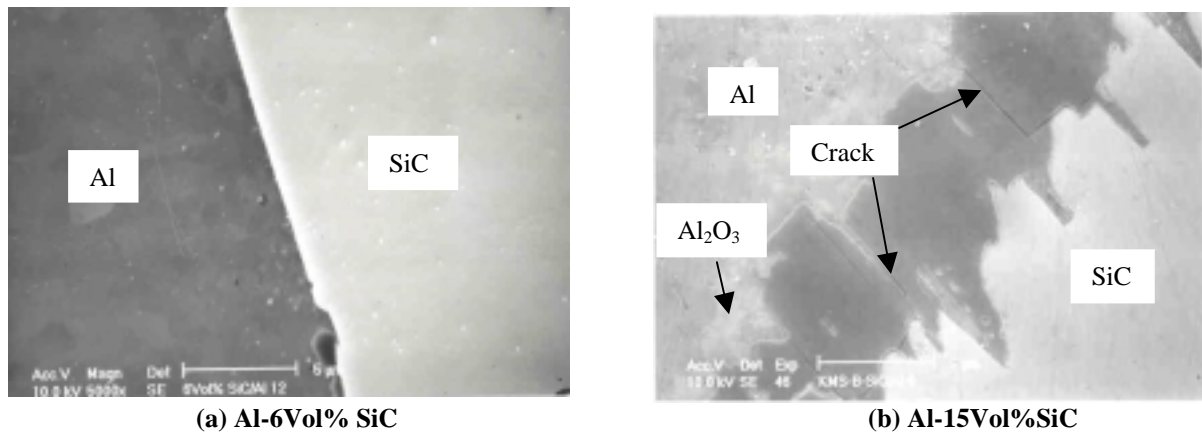


(b)



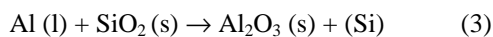
(c)

**Fig. 3 SEM micrographs showing the interfaces of (a) Al-20vol%SiC (interaction time 60 min.) (b) Al-20vol%Al<sub>2</sub>O<sub>3</sub> (interaction time 66 min.) and (c) Al-13vol%B<sub>4</sub>C (interaction time 46 min.).**



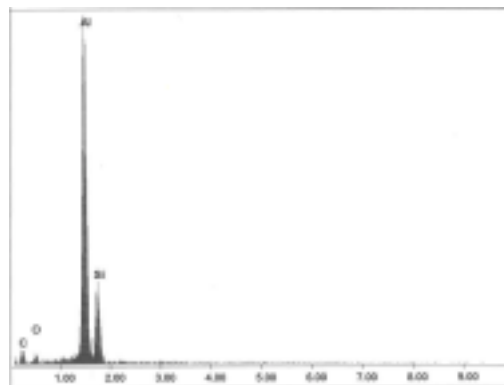
**Fig. 4 SEM micrographs showing the interface of Al/SiC composites at different interaction times: (a) 15 minutes (no interaction layer) and (b) 90 minutes (thick interaction layer)**

increasing their surface energy. The surface energy increase is obtained by removal of the moisture, gas and other contaminants as well as by the formation of a thin layer of SiO<sub>2</sub> on the the surface of SiC particles. This oxide layer reacts with the melt according to the following reaction [Rajan *et al.*, 1998 and Delannay *et al.*, 1987]:

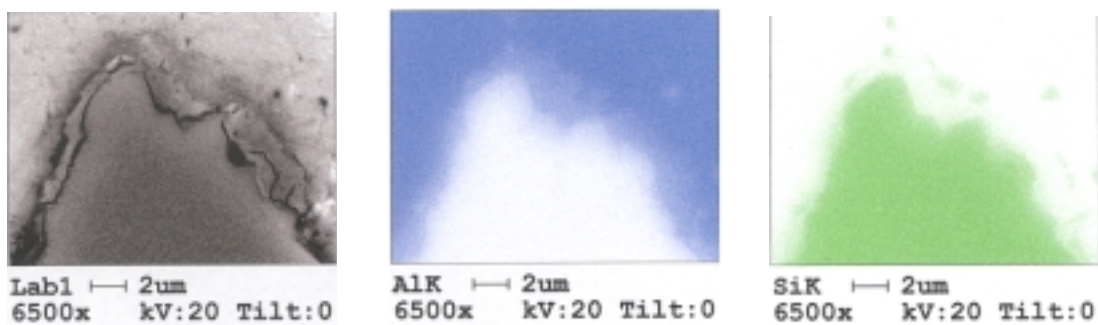


Where l is liquid, s is solid and (Si) is Si dissolved into the melt.

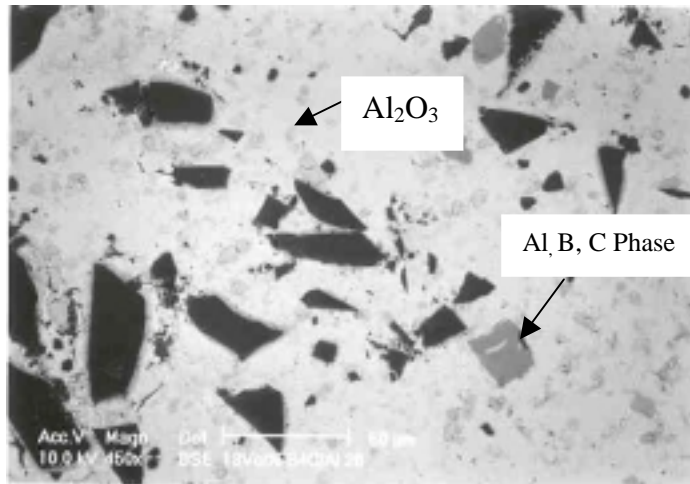
For the Al-Al<sub>2</sub>O<sub>3</sub> composite, no reaction product was found at the interface, which appeared clean in the cast composites as shown in Fig. 3b. Al<sub>2</sub>O<sub>3</sub> particles were heated at 900<sup>o</sup>C for two hours to increase the wettability. Heating aluminium oxide particles to high temperature will lead to dehydroxylation as well as



**Fig. 5 EDX spectra showing the elements present in the interaction layer at the interface of Al-SiC composites**



**Fig. 6 X-ray mapping showing the distribution of Al and Si at the Al/SiC interface (black phase indicates the presence of elements).**

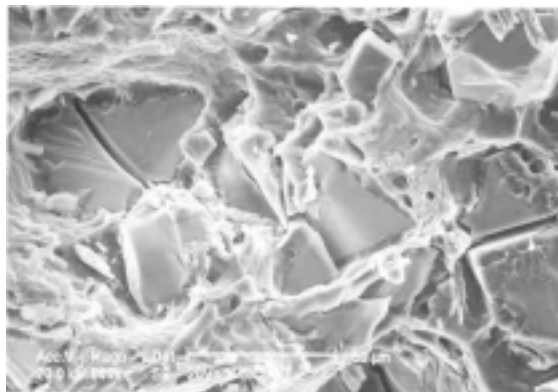


**Fig. 7 SEM micrograph showing the secondary phases in as cast Al-13 vol% B<sub>4</sub>C composites**

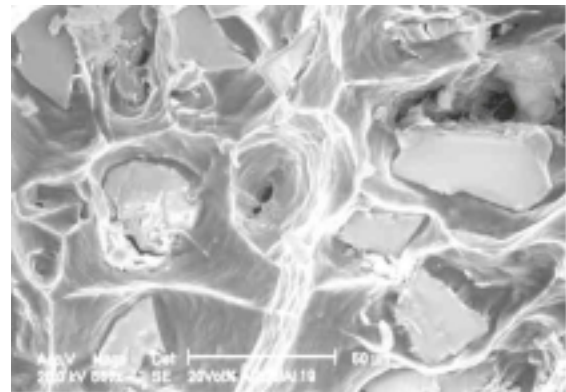
increase of surface energy [Rhee, 1972]. Another study reveal that decrease in contact angle (improved wetting) occur after heat-treatment due to the formation of a oxygen deficient surface containing some AlO in a spinel-type structure on the particle which increase the surface energy [Surappa and Rohatgi, 1981]. In this

investigation, heat-treated alumina particles were probably wetted by molten aluminum around 815°C because of the increase in the surface energy of alumina.

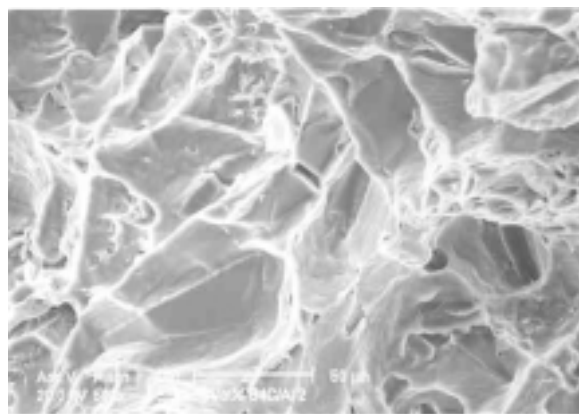
For Al-B<sub>4</sub>C composite, no clear interaction layer was detected at the interface at this resolution level (Fig. 3c).



(a)



(b)



(c)

**Fig. 8 Fracture surfaces of (a) Al-20vol%SiC, (b) Al-20vol%Al<sub>2</sub>O<sub>3</sub> and (c) Al-13 vol% B<sub>4</sub>C.**

The B<sub>4</sub>C particles seemed to adhere well to the Al matrix. Two secondary phases were observed in the matrix away from the interface (Fig. 7). From EDX analysis it is thought that these phases are Al<sub>2</sub>O<sub>3</sub> and aluminum boroncarbide. The presence of alumina particle in Al-B<sub>4</sub>C composites has also been reported elsewhere [Viala *et al.*, 1997]. The alumina may come from B<sub>2</sub>O<sub>3</sub>.Al<sub>2</sub>O<sub>3</sub> in Al-B<sub>4</sub>C composites during processing mentioned earlier. The alumina particles are small and spread throughout the Al matrix while the second phase forms relatively larger particles but less in number. B<sub>4</sub>C decomposes by chemical interaction with solid or liquid aluminium at temperature ranging from 627 to 1000<sup>o</sup>C. The reaction products are ternary carbide Al<sub>3</sub>BC and diboride AlB<sub>2</sub> up to 868<sup>o</sup>C. At temperature higher than 868<sup>o</sup>C, Al<sub>3</sub>BC is still formed while Al<sub>3</sub>B<sub>48</sub>C<sub>2</sub> replaces AlB<sub>2</sub> [Viala *et al.*, 1997]. As the processing temperature is below 868<sup>o</sup>C in the present work, the phases present are expect in the Al-B<sub>4</sub>C composites to be Al<sub>3</sub>BC and AlB<sub>2</sub>. But no aluminium boron containing phase was found. Hence, it is thought that the aluminium boron carbon containing phase are Al<sub>3</sub>BC. No Al<sub>4</sub>C<sub>3</sub> is present in Al-B<sub>4</sub>C composites as it was processed below 1000<sup>o</sup>C. This is a specific feature that differentiates the Al-B<sub>4</sub>C couple from other reactive couple such as Al-SiC, at the interface of which Al<sub>4</sub>C<sub>3</sub> appears as a major reaction product. Moreover, the secondary phases that are produced in Al-B<sub>4</sub>C system are not detrimental for the composites like Al<sub>4</sub>C<sub>3</sub>. These rather improve the properties of the composites [Marchi *et al.*, 2000]

Three composites with the highest volume fractions of the reinforcing particles viz. Al-20SiC, Al-20Al<sub>2</sub>O<sub>3</sub> and Al-13B<sub>4</sub>C were fractured to investigate the nature of the bonding between the reinforcing particles and Al matrix. Fracture surfaces of these three composites are displayed in Fig. 8. Among the three composites, Al-B<sub>4</sub>C system showed the strongest bonding as revealed by the good matrix/particulate adherence. No gap or micro-void was observed at the of particle/matrix interface in the fracture surface. For Al-SiC system, the bonding seems to be weaker than that of Al-B<sub>4</sub>C due to the relatively low matrix alloy adhering to the particulate. The weakest bonding among the three composites was found in the fracture surface of Al-Al<sub>2</sub>O<sub>3</sub> system. Voids and micro-voids were present at the particle/matrix interface indicating a weak bonding.

### CONCLUSIONS

Particles distribution was found to be better in Al-B<sub>4</sub>C composites as compared to Al-SiC and Al-Al<sub>2</sub>O<sub>3</sub> composites. A clear interfacial reaction product was found at Al/SiC interface for composites processed for

long period, while no reaction product was observed at Al/B<sub>4</sub>C and Al/Al<sub>2</sub>O<sub>3</sub> interfaces. Two secondary phases in the aluminium matrix away from the interface in Al-B<sub>4</sub>C composites are thought to be Al<sub>2</sub>O<sub>3</sub> and Al<sub>3</sub>BC. B<sub>4</sub>C reinforced Al composite seemed to exhibit a better interfacial bonding as compared to the other two composites.

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