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The wettability and the reaction for SiC particle/Al alloy system

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The incorporation process and the wettability for an SiC particles/aluminium alloy system were measured. The wettability between SiC particle and liquid aluminium was evaluated by the time required for the particulate incorporation. The incorporation time could be measured from a stirring time–melt temperature chart. Magnesium and titanium shortened the incorporation time of α -SiC particles into liquid aluminium and improved the wettability because of their strong affinity for SiC. Copper and zinc prolonged the incorporation time and no reaction products were found in the matrix. Furthermore, surface active elements with weak affinity for SiC (lead and bismuth) extremely prolonged the incorporation time because these elements prevent the reaction at the interface, whereas lithium shortened the incorporation time remarkably.

1. Introduction

Metal matrix composites (MMCs) have been expected as a new material with various excellent properties. In general, MMCs are classified in three types by the configurations of the reinforcements such as particulate composite, whisker-reinforced metal and fibre-reinforced metal. Although the industrial use of MMCs has been limited by the complexity of a manufacturing process or the high cost of production, particulate composite can be manufactured more easily by the melt-stirring method. In the melt-stirring process, however, good wettability between solid phase and liquid phase is one of the most important factors. The wettability has often been measured by the sessile drop method and estimated quantitatively using the contact angle, θ. Because the surface of the solid particle is activated when the particulate radius is very small (~ 1μm), the wettability for a system of solid particle/liquid metal cannot simply be estimated by the contact angle, θ. Oh *et al.* [1] reported the wettability between silicon carbide (SiC) and zirconium carbide (ZrC) particles and liquid metals (aluminium, Al–Mg alloy and Al–Cu alloy) using a pressure infiltration method. In the present work, the wettability between SiC particles and aluminium alloy has been evaluated from the time necessary for particulate incorporation into liquid aluminium alloy. By using this method, particulate wettability can be evaluated under the same conditions as the manufacturing process of the composite.

2. Experimental procedure

A total amount of 60 g aluminium or aluminium alloy was melted in an MgO crucible (inner diameter 38 mm) in an induction furnace [2]. After the melt had been held at 1023 K, SiC particles wrapped in an aluminium foil were preheated above the melt for 600 s. The average particle size of SiC used in this experiment was 14 μm. Then particles were added to the liquid aluminium and melt stirring was started by using an alumina rod. The time which is necessary for particulate incorporation was measured to estimate the effect of alloying elements on the particulate wettability. The incorporation time was identified from a stirring time – melt temperature chart. A number of alloying elements was experimented with and their respective effects on the incorporation time were measured. A cross-section of the composite was observed by scanning electron microscopy (SEM) and energy probe micro analysis (EPMA).

3. Results and discussion

3.1. Incorporation process of SiC particles

During the melt-stirring process, particles added to the melt were distinguished into incorporated particles and still unincorporated particles. However, further observation enabled unincorporated SiC particles to be classified into two groups; as-powder particles and agglomerates of particles bound by aluminium as shown in Fig. 1. Thus the particles were classified into three groups in all: (a) as-powder particles, (b) agglomerate of particles, (c) incorporated particles. In order to measure the ratio of these three types of particulate forms, as-powder particle and agglomerates were divided by a 200 mesh screen, and incorporated particles were extracted by dissolving the aluminium matrix with NaOH solution. For the SiC/pure aluminium system, the change in the ratio of these three types of particle with stirring time is shown in Fig. 2. According to Fig. 2, the ratio of as-powder SiC decreased

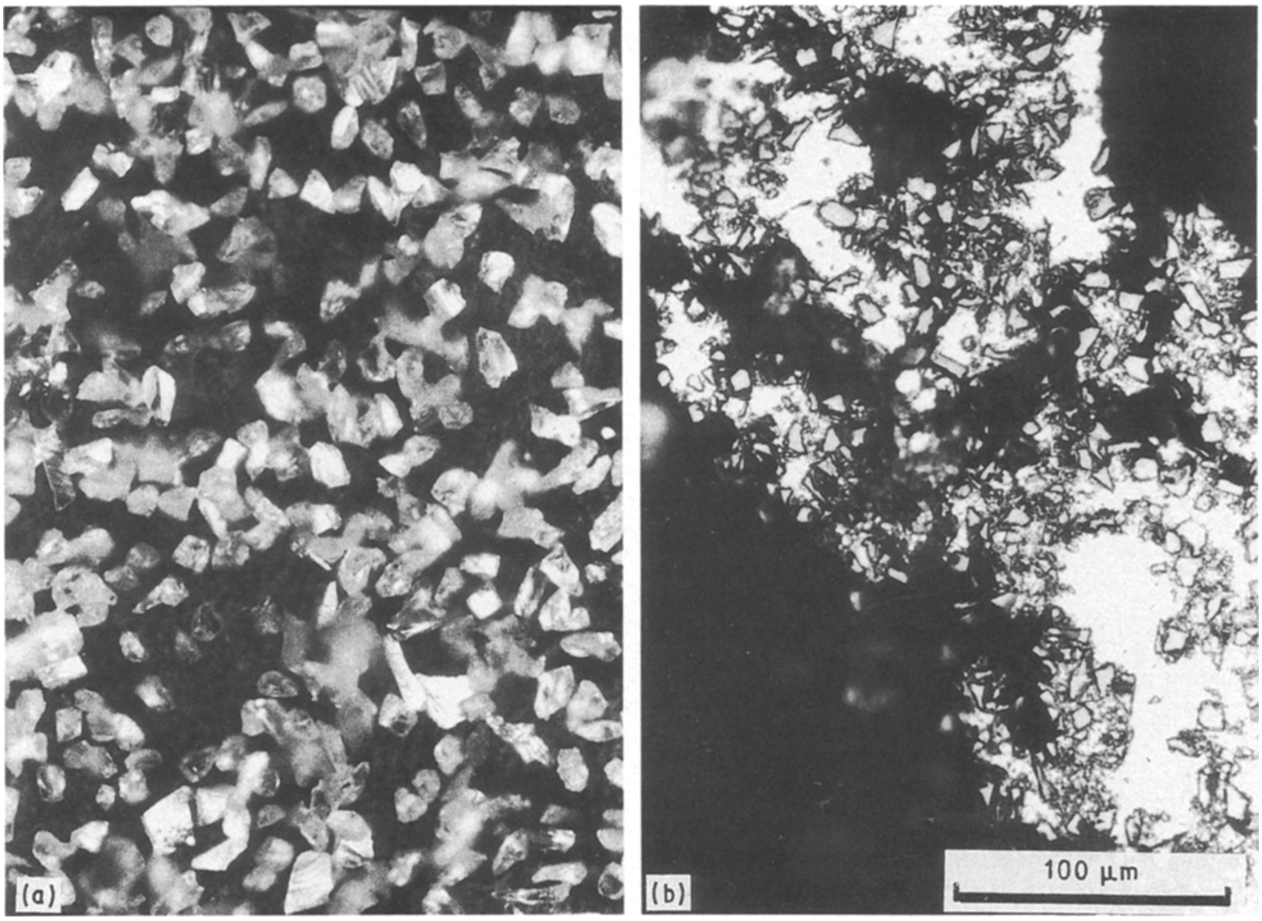


Figure 1 Optical micrographs of α-SiC(20 µm) particles; (a) in powder phase, and (b) in the agglomerate.

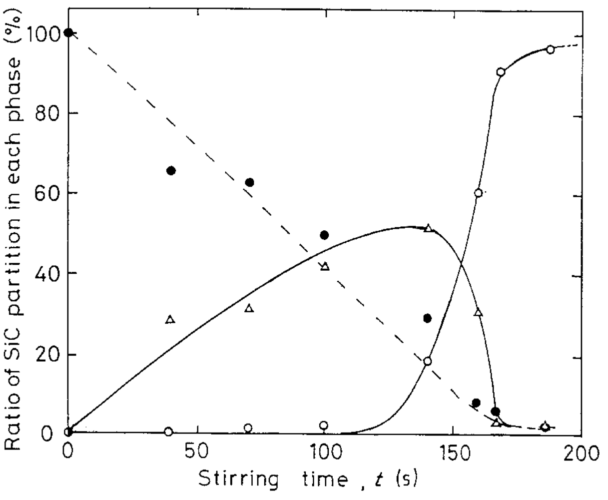


Figure 2 Incorporation process of α-SiC particles into liquid aluminium at 1023 K. (●) SiC in powder phase, (△) SiC in agglomerate, (○) SiC dispersed in melt.

linearly with stirring time, while the ratio of SiC particles in the agglomerate increased and then reached a maximum value at the same time as particulate incorporation started. Therefore, it is clear that SiC particles were not incorporated into the liquid aluminium immediately and an incubation time existed until SiC particles began to be incorporated into the liquid aluminium. This indicates that the SiC particles were gradually wetted by liquid aluminium, and the agglomerates were formed out of partly wetted particles. Thus the incorporation time represents a duration which is necessary for full particulate wetting. The same measurement was conducted on the SiC/Al–0.5% Mg alloy system as shown in Fig. 3. Although the incorporation time and the incubation time were shortened by a magnesium addition, the incorporation process of SiC particles was almost the same as that of the SiC/aluminium system. Furthermore, as shown in Fig. 4, the incorporation process was not affected by the melt temperature, either. So,

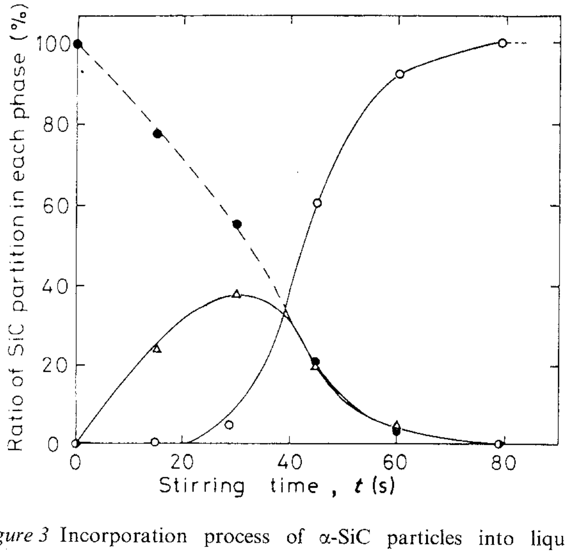


Figure 3 Incorporation process of α-SiC particles into liquid Al–3.0% Mg alloy at 1023 K. For Key, see Fig. 2.

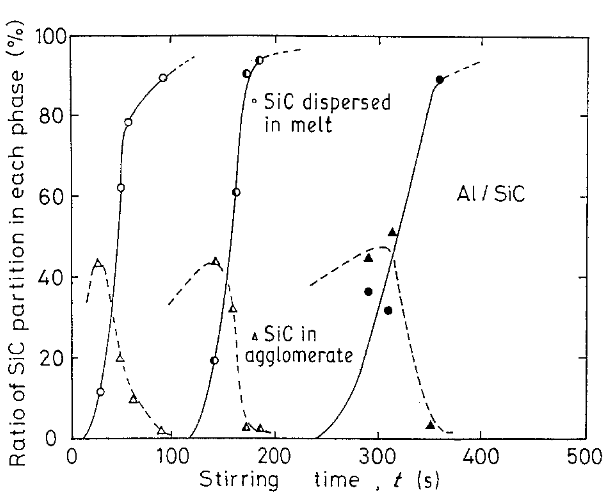


Figure 4 Effect of the temperature on the incorporation process of α-SiC particles into liquid aluminium at (○) 1223 K, (◐) 1073 K, and (●) 993 K.

the incorporation process is considered to be independent of the matrix composition and the melt temperature.

3.2. Measurement of the incorporation time

As reported in Section 3.1, a constant time is required for the particulate incorporation and the wettability is considered to be a dominant factor in this incorporation time. Therefore, the wettability between solid particle and liquid metal can be estimated from the time which is necessary for particulate incorporation (incorporation time). The incorporation time was easily obtained from the stirring time–melt temperature chart (Fig. 5). When liquid metal wets the solid surface and spreads over it, a chemical reaction at the interface may occur in most cases. During the wetting process, a heat generation due to a change in the interfacial energy from solid/vapour to solid/liquid interface ($\sigma_{sl} - \sigma_{sv}$) and due to a reaction at the interface was observed, although, in this case, $\sigma_{sl} - \sigma_{sv}$ is small enough to neglect. Therefore, as shown in Fig. 5, the temperature of the liquid aluminium started to rise as soon as melt stirring began. Finally, the melt temperature returned to the initial value after a constant period. By comparing the change of melt temperature with the incorporation process, incorporation time is ascertained to correspond to the period from the starting point to the end of stirring, as shown by the arrows in Fig. 5. Thus, the incorporation time was decided by using the stirring time–melt temperature chart in this experiment.

3.3. Effect of alloying elements with strong affinity for SiC on the incorporation time

First, the effect of magnesium and titanium on the incorporation time of SiC particles into liquid aluminium was measured. It is clear that the incorporation time was shortened by alloying magnesium and titanium, as shown in Fig. 6. Reaction products were detected in both cases. As shown in Fig. 7, X-ray images of silicon and magnesium overlap each other in the case of SiC/Al–5.0% Mg composite and this indicates the formation of magnesium silicide in the matrix. A cross-section of SiC/Al–5% Ti composite was also observed by SEM and EPMA as shown in Fig. 8 and a titanium-enriched zone was found to be formed around SiC particles. The standard free energies of formation of titanium silicide and titanium carbide are low enough to form TiSi, Ti_2Si, Ti_5Si_3 and TiC, so titanium seems to react with SiC to produce silicide or carbide [3].

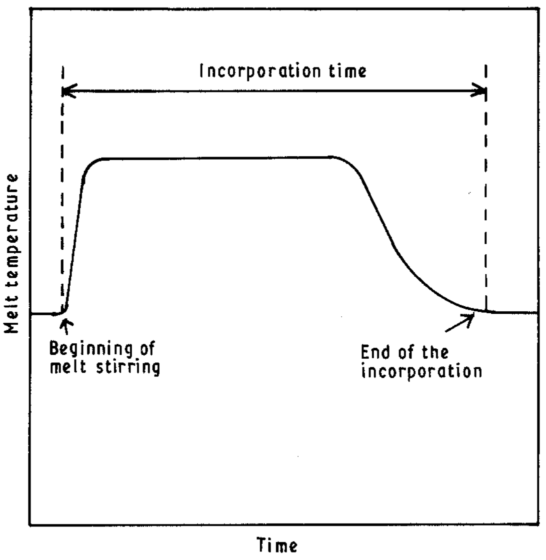


Figure 5 The relation between stirring time and melt temperature.

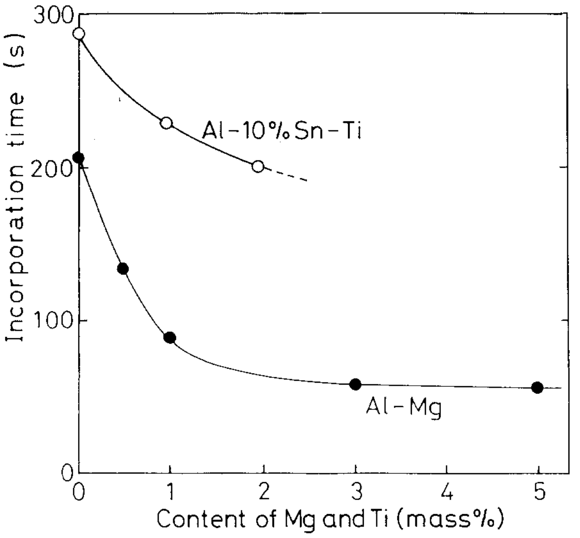


Figure 6 Effect of magnesium and titanium on the incorporation time of α-SiC particles into liquid aluminium.

In these cases, the dissolution of SiC is considered to be a dominant factor of the wetting process.

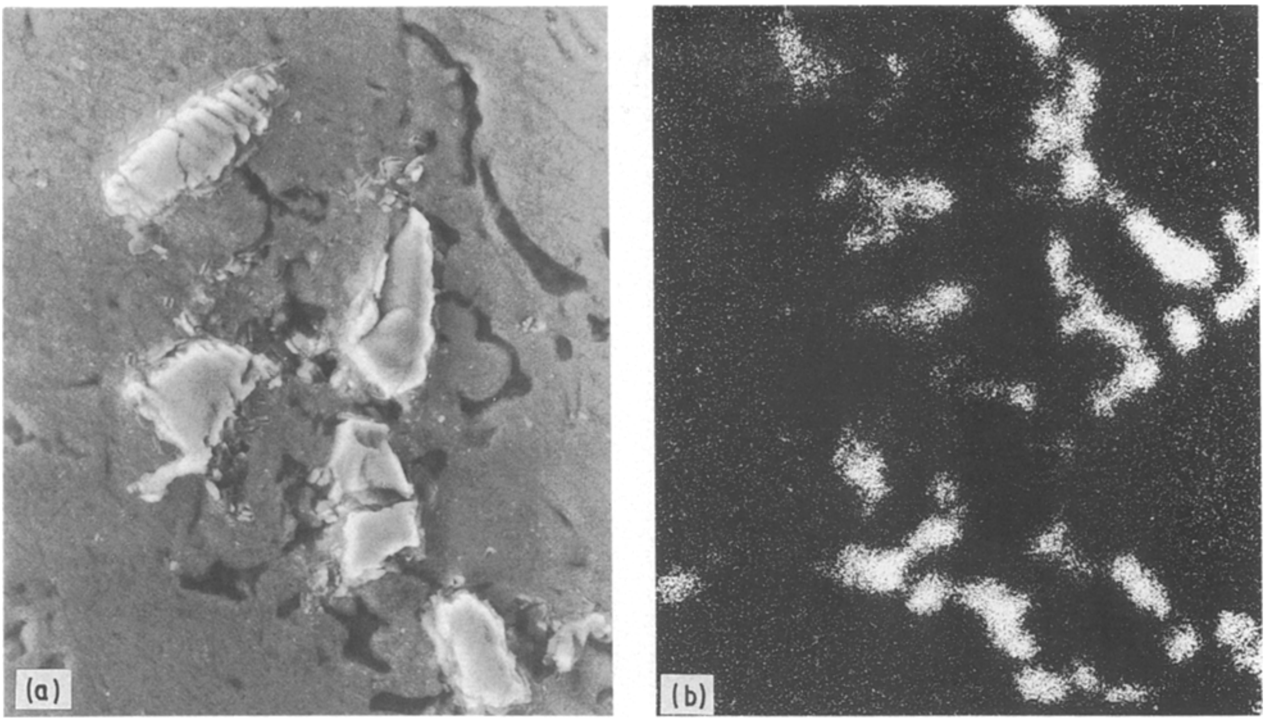
$$\mathrm{SiC} \rightarrow (\mathrm{Si}) + (\mathrm{C}) \tag{1}$$

where (Si) or (C) indicates a dissolved element. Equation 1 is promoted by the reaction between these elements and an alloying element (Me)

$$(\mathrm{Si}) + m\mathrm{Me} = \mathrm{Me}_m\mathrm{Si} \tag{2}$$

$$(\mathrm{C}) + n\mathrm{Me} = \mathrm{Me}_n\mathrm{C} \tag{3}$$

Owing to the reactions given above, a new active



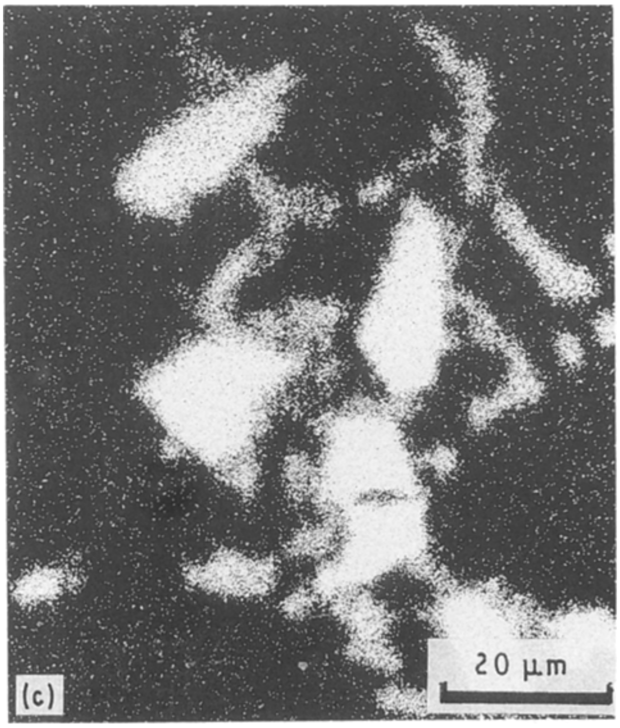


Figure 7 (a) Scanning electron micrograph, and X-ray images of (b) magnesium and (c) silicon for SiC/Al–5.0% Mg composite.

surface appears on the SiC particles and the wettability is improved. From this point of view, it is reasonable to consider that magnesium and titanium contribute to the shortening of the incorporation time through the reaction of silicide or carbide formation.

3.4. Effect of alloying elements with weak affinity for SiC on the incorporation time

The effect of zinc and copper, which have weak affinity for SiC and do not form their silicides, on the incorporation time, was investigated. It is clear from Fig. 9 that the incorporation time was prolonged with an increase in the addition of these elements. From the results of the EPMA analysis on SiC/Al–Zn and SiC/Al–Cu composites, no reaction product was detected in the matrix. Furthermore, they do not show a surface active property which reduces the surface energy of liquid aluminium. Thus, these elements do not contribute to the reactions given in equations 2 and 3. Consequently, an increase in the incorporation time is due to a decrease in the activity of aluminium containing zinc or copper.

3.5. Effect of surface active elements on the incorporation time

Wettability is, generally, improved by a decrease in the surface energy of the liquid surface, γ_{lv}. So a surface active element such as lead, bismuth and lithium, which reduces the surface energy of liquid aluminium [4], was added. From Fig. 10, there is an apparent decrease in the incorporation time by alloying lithium. However, contrary to expectation, it was extremely prolonged by alloying lead or bismuth even at a small amount of addition (Fig. 10). Only a 1% addition of these elements prevents the particles from being incorporated into the liquid aluminium. In these cases, the reaction products formed by the reaction between SiC and lead or SiC and bismuth was not detected by EPMA. On the other hand, with respect to SiC/Al–3% Li specimen, large reaction products were found in the matrix, as shown in Fig. 11. Because lithium could not be detected by EPMA, Auger electron spectroscopy (AES) was used in this case. From the results of AES analysis, it is evident that the reactant consisted of aluminium, silicon and lithium (Fig. 11). According to the experimental results given in Fig. 10, lead, bismuth, and lithium show the opposite effects on the incorporation time, although these

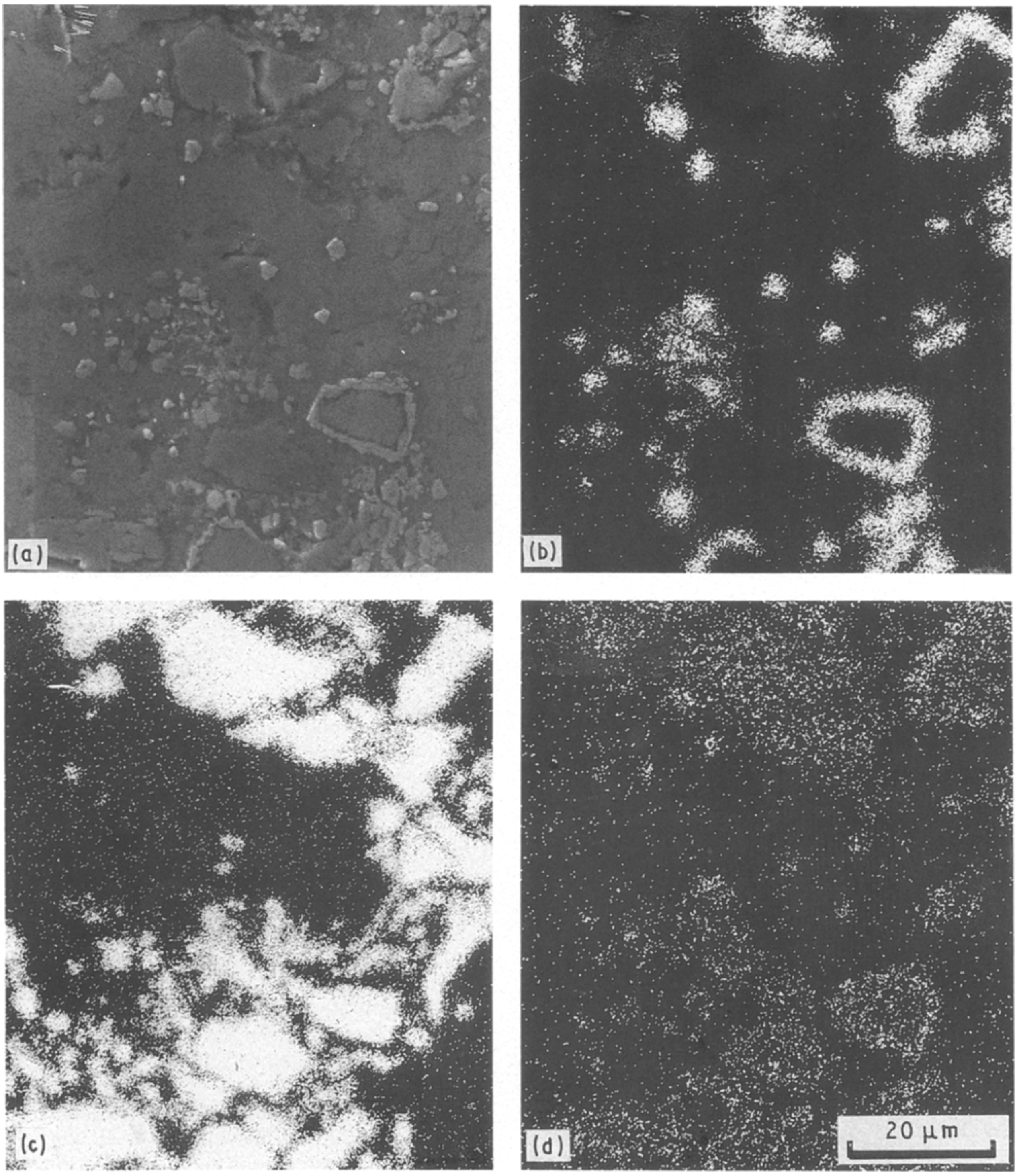


Figure 8 (a) Scanning electron micrograph, and X-ray images of (b) titanium, (c) silicon and (d) carbon for SiC/Al–5.0% Ti composite.

three elements equally indicate the surface activity. This is apparently due to whether each alloying element has an affinity for SiC or not, as reported in Sections 3.3 and 3.4.

To clarify the difference in the effects of lead, bismuth and magnesium, the concentration of those elements at the melt surface (surface excess concentration, Γ) was calculated from the value of the surface tension measured by Lang [5] with the help of Gibbs' adsorption equation

$$\Gamma = -\frac{1}{RT}\frac{d\gamma_{LV}}{d\ln\alpha_j} \qquad (4)$$

Where α_j is the activity of component j, R is the gas constant, T is the temperature (K). Because of the lack of values for lithium activity in aluminium, the surface excess concentration of lithium could not be calculated. Fig. 12 shows the surface excess concentration of bismuth, lead, tin and magnesium calculated from Equation 4. It is apparent that the surface excess concentrations of bismuth and lead are extremely high, even for a small addition. For example, with only a 0.3% addition of lead, the surface excess concentration is near to the value of the atomic density of the (1 0 0) plane of lead, shown in Fig. 12. Thus the surface of the aluminium melt is almost occupied by lead

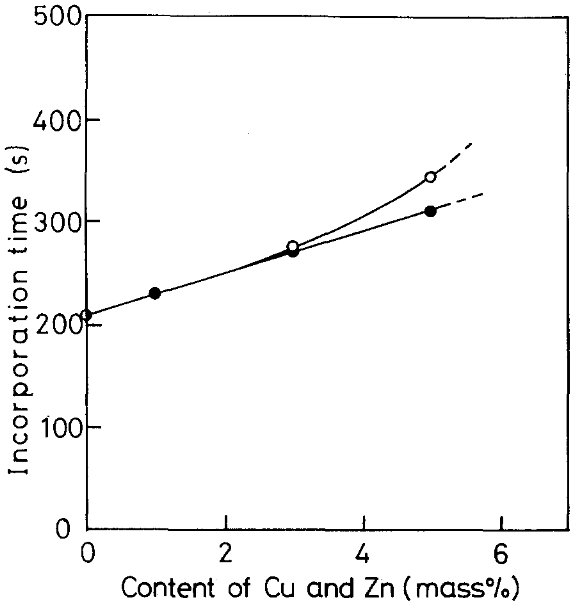


Figure 9 Effect of (●) zinc and (○) copper on the incorporation time of α-SiC particles into liquid aluminium.

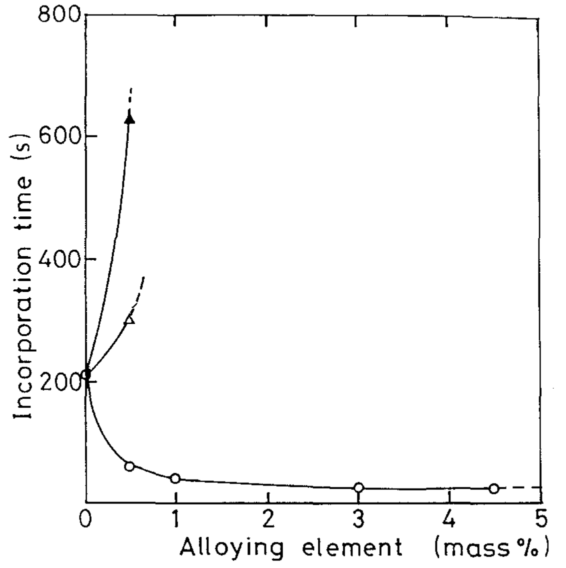


Figure 10 Effect of (○) lithium, (△) lead and (▲) bismuth on the incorporation time of α-SiC particles into liquid aluminium.

atoms. So the affinity of these surface active elements for SiC is a more important factor than the degree of surface activity. Lead or bismuth, which has weak affinity for SiC, adsorbs at the surface of the liquid aluminium and behaves as an obstacle to the progress of SiC dissolution or reaction given in Equations 2 and 3.

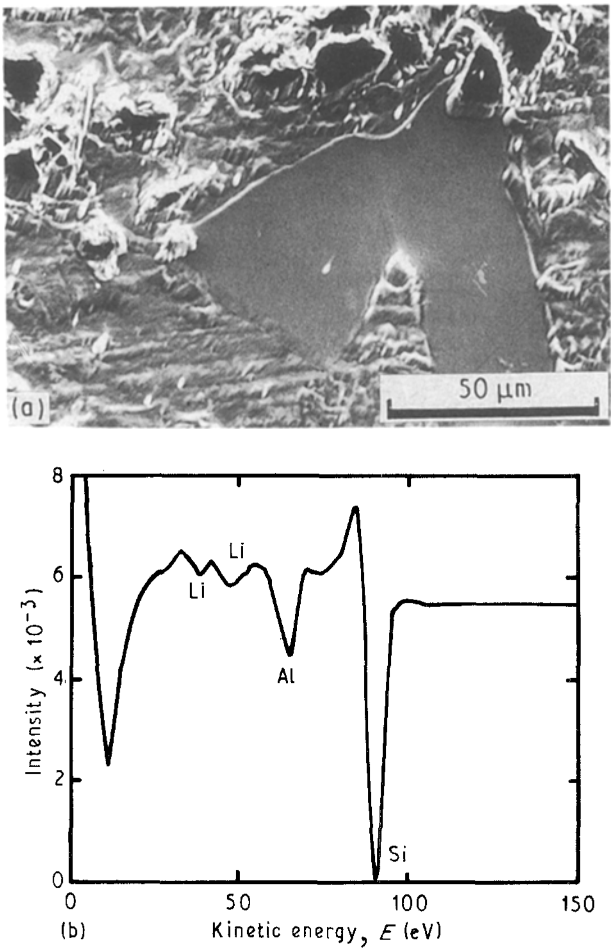


Figure 11 (a) Scanning electron micrograph and (b) the results of AES analysis on the reaction product for SiC/Al–3.0% Li composite.

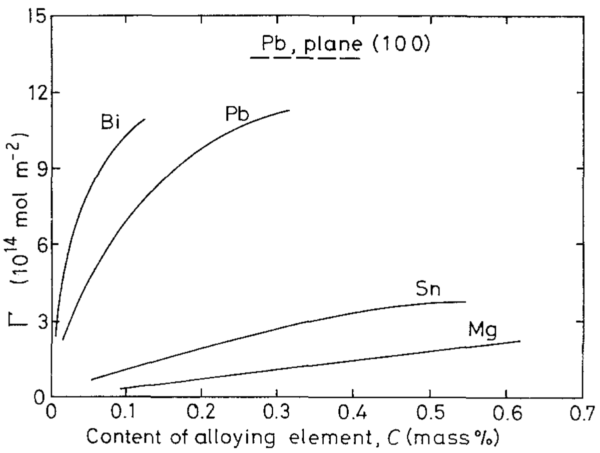


Figure 12 Surface excess concentration of magnesium, lead and bismuth in liquid aluminium, calculated from the data of Lang [5] at 973 K.

4. Conclusions

The incorporation process and incorporation time of SiC particles into liquid aluminium were measured in order to evaluate particulate wettability.

1. During the particulate incorporation process, the agglomerates of SiC particles were formed until particulate incorporation began.

2. The incorporation time was identified from the stirring time–melt temperature chart.

3. Magnesium and titanium, which have affinity for SiC, decreased the incorporation time of SiC particles into liquid aluminium, while copper and zinc prolonged it.

4. Although lead and bismuth decrease the surface energy of liquid aluminium, they strongly prevented SiC particulate incorporation because they both have weak affinity for SiC.

5. Lithium shortened the incorporation time remarkably, and reaction products including lithium were observed.

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