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Production of rapidly solidified Al/SiC composites

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Rapidly solidified metal matrix composites have been produced on a laboratory scale either by (1) melt spinning a composite after introduction of the ceramic phase and extrusion of the flakes obtained, or (2) blending melt-spun powder (basic alloy) with the ceramic phase and subsequent extrusion. AlMg(Si) alloys were used as matrix material while SiC particles with diameters of 3 or 20 μm were used as the ceramic phase. For the composites prepared by route 1 it was found that the basic alloy was reinforced by the addition of 3 μm particles whereas for the 20 μm particles reinforcement was observed only for very ductile matrices. The bond between SiC particles and matrix was good. A diffusion and wetting bond was formed. For the composites prepared by route 2 it was found that reinforcement did not occur and that the bond between particles and matrix was weak. Debonding of the particles took place in the case of tensile fracture. The advantage of a rapidly solidified matrix for these composites is that relatively high ductilities are combined with good reinforcement effects. Prior contact of the ceramic phase and the aluminium melt is needed for a good bond between SiC and the matrix material. It is concluded that route 1 should be preferred for the production of rapidly solidified aluminium matrix composites.

1. Introduction

The introduction of a ceramic phase in an aluminium alloy matrix has proved to be a very promising way to improve a number of properties of these alloys, such as strength at room temperature or higher temperatures, Young's modulus, wear resistance and thermal expansion coefficient. Yet these materials usually show bad ductility and toughness [1, 2].

SiC has been one of the most successful reinforcement materials for aluminium alloys to date, because it has very high strength, stiffness and hardness [3] and it is chemically compatible with aluminium alloys up to at least 500° C [4, 5]. The wetting of SiC by aluminium, however, is bad at temperatures below 1000° C, which causes difficulties in the production of SiC/Al composites [6]. Production of these composites, with either short fibres or particles as reinforcement, is possible via both the powder metallurgy route (P/M) and the foundry route.

Powder metallurgy involves mechanical mixing of aluminium powders with the ceramic phase and subsequent consolidation of the powder mixture. The consolidation of the powder mixture is achieved by hot pressing at temperatures which are often above solidus temperatures and/or by applying a hot deformation process such as extrusion.

Two techniques are frequently used in the foundry route, namely liquid infiltration into a preform of the ceramic phase or introduction of the ceramic phase into the molten metal. For this latter method, generally referred to as compocasting, it is necessary either to coat the ceramic particles for easy wetting [7–9] or to introduce uncoated particles into a semi-solid slurry during stirring [10]. By introduction into a semi-solid slurry, the particles are mechanically entrapped and bonding between the ceramic phase and the alloy is promoted by a long contact time. It has also been observed that the presence of certain alloying elements, such as magnesium in the case of SiC and aluminium, promotes wetting and bonding [11].

There are few publications concerning the rapid solidification processing (RSP) of metal matrix composites [12–16] and to our knowledge none about RSP of aluminium matrix composites. Slaughter and Das [12] produced Fe–Al alloys with TiB_2 particles by gas atomization. The TiB_2 particles acted as a stabilizer of the RSP structure. Kimura *et al.* [13] prepared, by melt spinning, nickel-based metallic glasses with WC particles which showed improved yield strength compared to the matrix. Zielinski and Ast [14–16] incorporated other kinds of particles and fibres (SiC, TiC, B, C) in the same matrix. They reported that the introduction of the particles in the melt before melt spinning did not give a good repartition of these particles in the ribbon, compared with blowing the particles into the puddle during melt spinning.

In the present research the combination of the good characteristics of a ceramic phase and a rapidly

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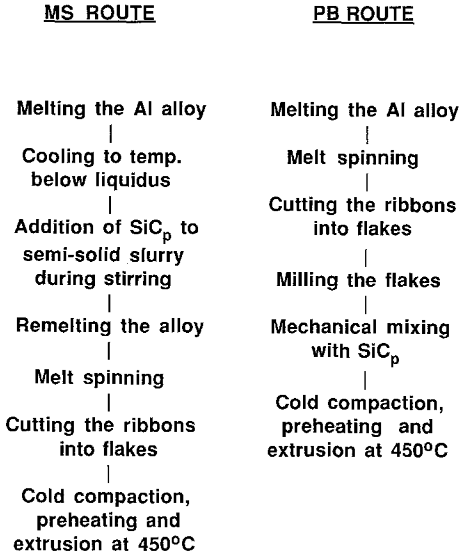

Figure 1 Processing routes used for the production of the composite materials.

solidified matrix structure was investigated. The better ductility and higher strength of a rapidly solidified matrix in comparison with a conventional one, should be profitable when used for a composite material.

2. Experimental procedure

Aluminium alloys of three compositions (AlMg5, AlMg5Si1 and AlSi7Mg5) were made starting from pure aluminium (99.99%), pure magnesium and AlSi13. SiC particles of 20 and 3 μm average diameter were used as-delivered. In Fig. 1 the two processing routes applied are shown schematically.

In the MS route the aluminium alloy is melted and then cooled some 20 to 30° C below liquidus temperature while stirring. Preheated SiC particles were introduced into the semi-solid slurry through the hollow stirrer shaft with an argon flow. After the introduction, which took about 5 to 10 min, the semi-solid composite was heated into the liquidus region. Rapid solidification was performed by melt spinning with a copper wheel rotating at a circumferential velocity of $25\,\mathrm{m\,s^{-1}}$. After melt spinning, the rapidly solidifed ribbons were cut into flakes of about 2 mm length using a chopper, cold compacted, preheated and finally extruded at a temperature of 450° C in a laboratory extrusion press with a capacity of 60 ton. The extrusion ratio was 35:1. The final product was strip 2 mm thick, 10 mm wide and about 1 m long. No heat treatment was applied in most cases. Only a relaxation treatment (10 min at 345° C) was applied to some of the AlMg5Si1 composites.

The powder blending (PB) route consisted of melt spinning the aluminium matrix alloy, chopping the ribbons into flakes and milling the flakes into smaller particles with diameters of 100 to 600 μm. The matrix powder and the SiC particles were then mechanically mixed, compacted and hot extruded under the same conditions as the corresponding MS composites. From each strip three tensile specimens were obtained by spark-erosion. Table I summarizes the compositions of the materials produced. Microstructural investigation of the ribbons and the extruded products was performed using optical microscopy and transmisson electron microscopy (TEM). The SiC concentrations were determined using a Quantimet system. Examination of fracture surfaces of both ribbons and tensile specimens was performed with a scanning electron microscope (SEM).

3. Results

3.1. Composite ribbons

The thickness of the composite ribbons was in the range 50 to 100 μm. Their appearance was not very different from that of conventional ribbons. The concentration of particles varied along the longitudinal direction, but this variation was not periodic, as reported by Zielinski and Ast [16]. Almost no clustering of the particles was observed in the ribbons after melt spinning. The particles had a tendency to segregate towards the upper surface of the ribbon, although in general they were not at the surface or in contact with air. It was observed that the interface between SiC particles and the matrix differed with the type of matrix. For the matrix without silicon, the interface was less straight and less sharply defined than for the matrices containing silicon (Fig. 2), which indicates that the SiC particles were chemically attacked. In composites with silicon in the matrix no crystals resulting from a chemical reaction at the interface and no phases with a characteristic diffraction pattern were found (TEM). This means that if present they would be smaller than 10 nm. In composites without silicon in the matrix we could not see crystals at the interface either, but as the interface is less sharply defined, it would be more difficult to detect them. X-ray energy dispersive spectrometry measurements on the TEM using an electron beam diameter of 100 nm indicated some magnesium enrichment in the matrix at the SiC/Al interface. However, this enrichment occurred within a distance somewhat smaller than the beam spot diameter.

Tensile fracture of the composite ribbons showed that fracture occurred in the particles themselves and not at the interface between particles and matrix.

TABLE I Materials produced by MS or PB processing and their concentrations (wt %)

Matrix	20 μm particles		3 μm particles		no particles	
	MS	PB	MS	PB	MS	PB
AlMg5	10	10			x	
AlMg5Si1	5	5–10	2–5	5	x	x
AlSi7Mg5	10	10			x	

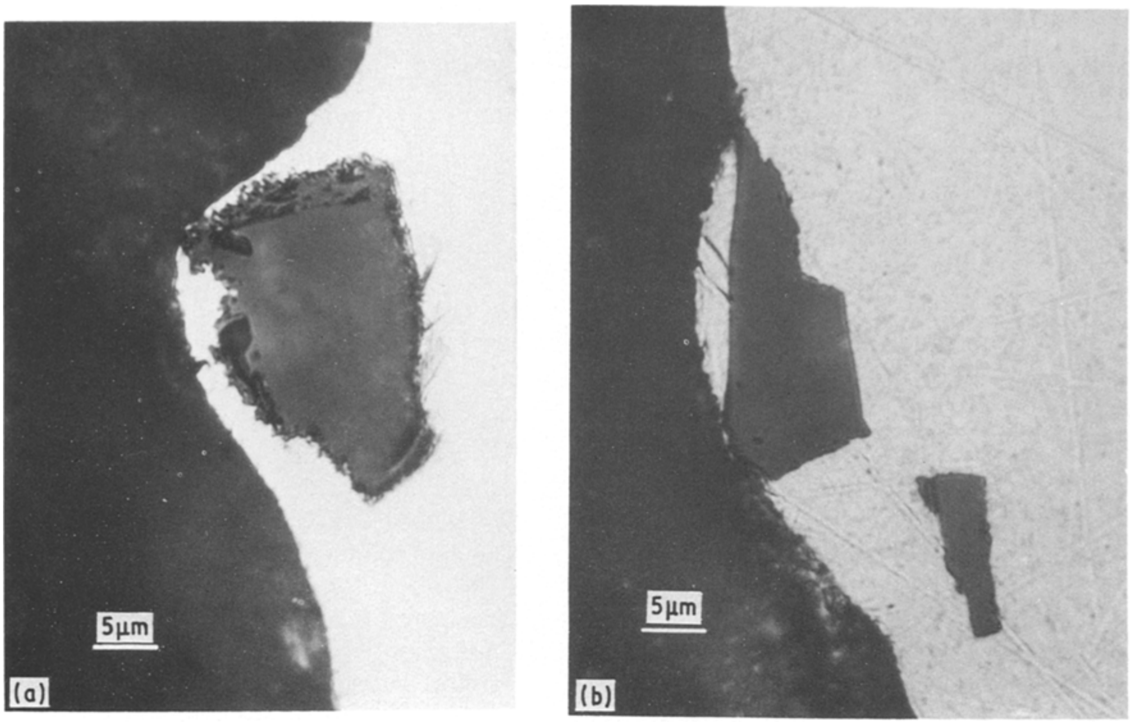

Figure 2 Optical micrographs showing SiC particles in the melt-spun ribbon. Matrices are (a) AlMg5 and (b) AlSi7Mg5.

Fig. 3 shows some broken particles which are embedded in the ribbon.

3.2. Extruded composites

3.2.1. Processing effects

At the same extrusion temperature, MS AlMg5 and AlMg5Si1 composites could be extruded more easily than the corresponding PB composites, this effect being most pronounced for the most ductile alloy (AlMg5). For this alloy the pressure needed for extrusion of the PB composite was 20% higher while at the same time the extrusion speed was 50% lower. The surface quality of the extruded products was much better for MS composites than for PB composites extruded under the same conditions. For both PB and MS composites, the surface quality improved with higher temperatures and higher extrusion speeds. Whenever a bad surface quality was obtained, we could observe that an irregular layer of material had developed on the edges of the extrusion die. However, the tensile properties of the material were not influenced by this surface defect. For the AlSi7Mg5 composites the difference in both extrudability and surface quality between PB and MS processing was not significant. So, the effects were more pronounced with the more ductile matrices.

On a macroscopic scale, the repartition of the particles was generally homogeneous. The heterogeneity in the longitudinal direction of the ribbons disappeared after cutting the ribbons into flakes and mixing the flakes. Yet, for percentages lower than 5% of 20 μm SiC particles, a remarkable macroscopic segregation effect occurred which led to the formation of bands with higher or lower concentrations of particles. This extrusion effect is described and discussed in another paper [17]. For the results presented here, the conditions were such that this effect could be neglected. On a microscopic scale the repartition of the particles in the extruded product was also homogeneous (Fig. 4). No clustering was observed with 20 μm particles and very little with 3 μm particles.

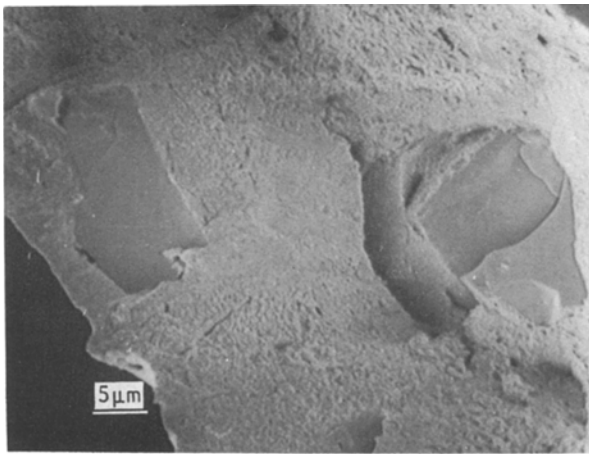

Figure 3 Scanning electron micrograph of the fracture surface of the composite ribbon (transverse section).

3.2.2. Tensile tests

The results of the tensile tests are schematically presented in Figs 5 and 6. Values are given in Table II. The materials were tested as-extruded. A relaxation heat treatment (10 min at 345° C) was applied to some AlMg5Si1 composites but it did not affect the tensile properties. The non-reinforced matrix materials yielded similar tensile properties when produced by either the MS or the PB route (the difference between the two routes being finer milling of the flakes). For 20 μm SiC particles, significant reinforcement resulted

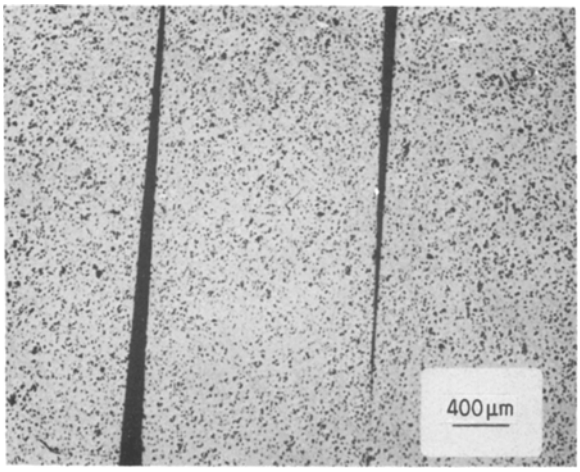

Figure 4 Optical micrograph showing the SiC particle (20 μm) distribution of three extruded strips. Extrusion direction is normal to the paper.

only with the AlMg5 matrix, which is the most ductile, after MS processing. The reinforcement effect was coupled with a decrease of elongation. However, when using the PB route, both a lower ultimate tensile strength (UTS), and a strongly reduced ductility were measured for 10% particle concentration. For the matrices of AlMg5Si1 and AlSi7Mg5 the results were less clear. Reinforcement of the matrices was not apparent whereas, in general, the values found for the elongation were considerably lower for the PB route as compared to the MS route. The situation was different for the composites containing 3 μm particles. For the MS composites we observed a strong increase of both UTS and yield strength (YS) with increasing particle concentration, whereas the elongation did not change appreciably. It was found that with 3.5% SiC particles the UTS and the YS increased by about 75 and 55 MPa, respectively, whereas the reduction in elongation was about 1%. For the PB composites the UTS and YS remained approximately the same, while the value of the elongation was lowered to about 10%. In general, when comparing the two processing routes applied, the results obtained after PB processing were inferior to those of the MS route.

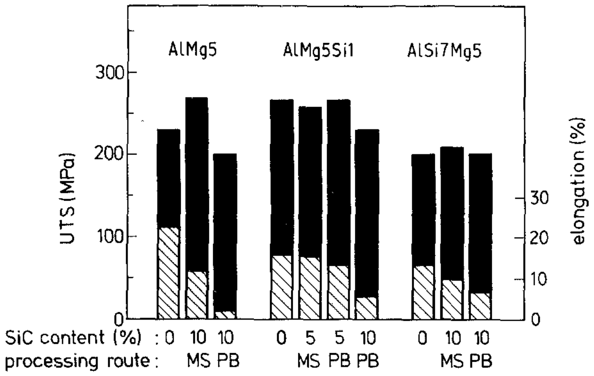

Figure 5 Schematic representation of the results of the tensile tests for various matrices and concentrations of 20 μm SiC particles and for MS and PB processing.

3.2.3. Fracture

The observation of the fracture surface by SEM showed that, for the MS composites, the 20 μm particles were broken under tensile loading, see Fig. 7. This is confirmed by Fig. 8, which shows the separated parts of individual particles after traction. On the fracture surfaces of MS composites reinforced with 3 μm particles, no broken particles were seen. Except for very few debonded particles, almost no SiC particles were recognized at the fracture surface. We ascribed certain protrusions to particles which were coated with matrix material. All fracture surfaces of PB composites showed debonding of the 20 or 3 μm SiC particles, which is illustrated in Fig. 9.

4. Discussion

The MS process permits the production of composite materials with relatively low volume fractions of reinforcement. Maximum percentages are ~15% with 20 μm particles and ~5% with 3 μm particles at present. Ast and co-workers [13, 14] introduced 2 to 7 wt % reinforcement in amorphous alloys using melt spinning. The volume fraction in our experiments can certainly be increased by further improvements of the experimental set-up, but will remain below the classical percentages of reinforcement used in metal matrix composites. Difficulties arise from the tendency of the ceramic phase to segregate towards the surface of the

TABLE II Tensile properties of the materials investigated

Matrix	SiC particles		MS/PB	UTS (MPa)	YS (MPa)	Elong. (%)
	Conc. (%)	Diam. (μm)				
AlMg5Si7	–	–	MS, PB	266	148	16
	2	3	MS	299	178	15
	3	3	MS	330	205	15
	3.5	3	MS	340	203	15
	5	3	PB	259	154	10
	5	20	MS	258		15–16
	5	20	PB	265		13–14
	10	20	PB	230		6
AlMg5	–	–	MS, PB	229		22
	10	20	MS	268		12
	10	20	PB	200		2
AlSi7Mg5	–	–	MS, PB	199		13
	10	20	MS	209		10
	10	20	PB	201		7

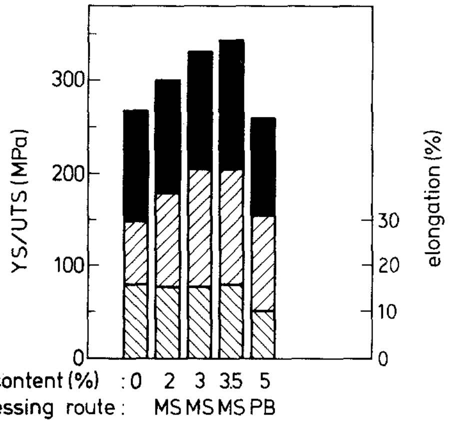

Figure 6 Schematic representation of the results of the tensile tests for AlMg5Si1, various concentrations of 3 μm SiC particles and MS or PB processing.

melt and from the higher viscosity of the liquid composite. These difficulties are more severe for the smaller particles which are the most efficient for reinforcement.

In Fig. 10 the present results on the tensile properties are compared with those of various discontinuous SiC/Al matrix composites taken from the literature [1, 18–21]. The graph shows that the highest tensile strengths are obtained with whisker-reinforced composites. It also shows, that the strength of particulate composites prepared by P/M is usually higher than the strength obtained by the MS method. However, the elongation of both particulate and whisker composites mentioned in the literature is very much lower (ranging from 1 to 8%) than the elongation of the MS composites (ranging from 10 to 16%). At equivalent strength, the ductility of MS composites reinforced with 3 μm particles is the highest. As our results are still confined to non-high strength matrices and low particle concentrations, further research will be needed to establish whether this combination of reinforcement and low loss of ductility will be maintained for high strength matrices and higher particle concentrations.

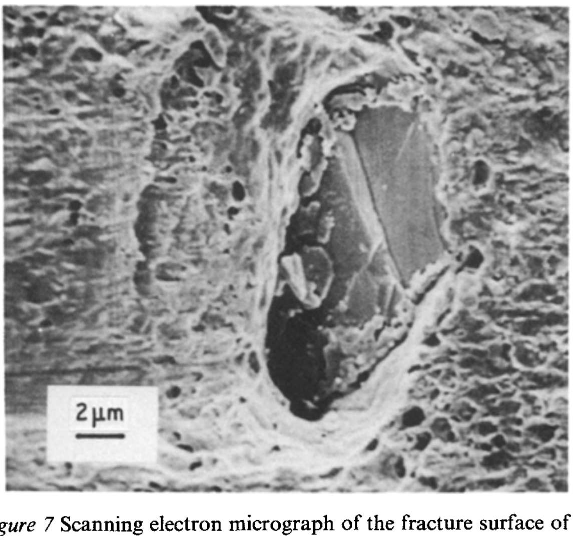

Figure 7 Scanning electron micrograph of the fracture surface of a MS composite showing a fractured 20 μm SiC particle embedded in the matrix.

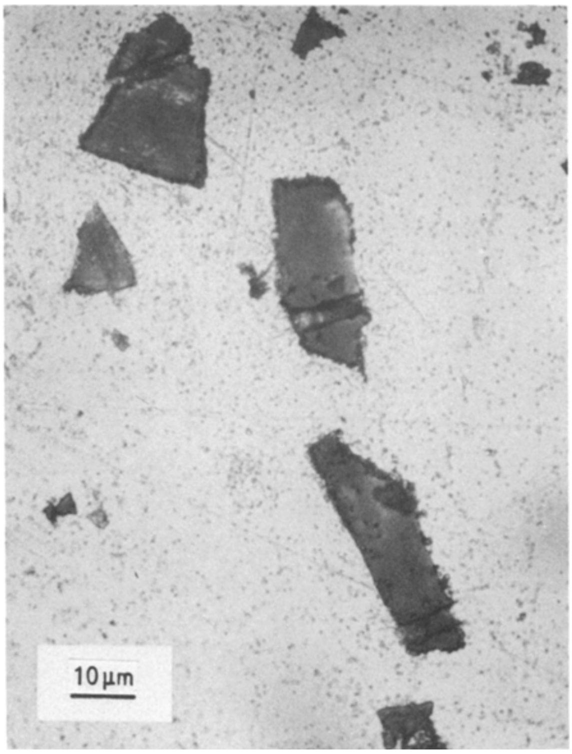

Figure 8 Optical micrograph of fractured SiC particles present in the bulk material of a MS composite after a tensile load was applied in vertical direction.

In Fig. 11 the difference between the tensile strength of a composite and its matrix is plotted against the difference between the elongation of this composite and its matrix. Again we notice that for the same increase in strength due to the introduction of SiC, the decrease in elongation is less for the 3 μm MS

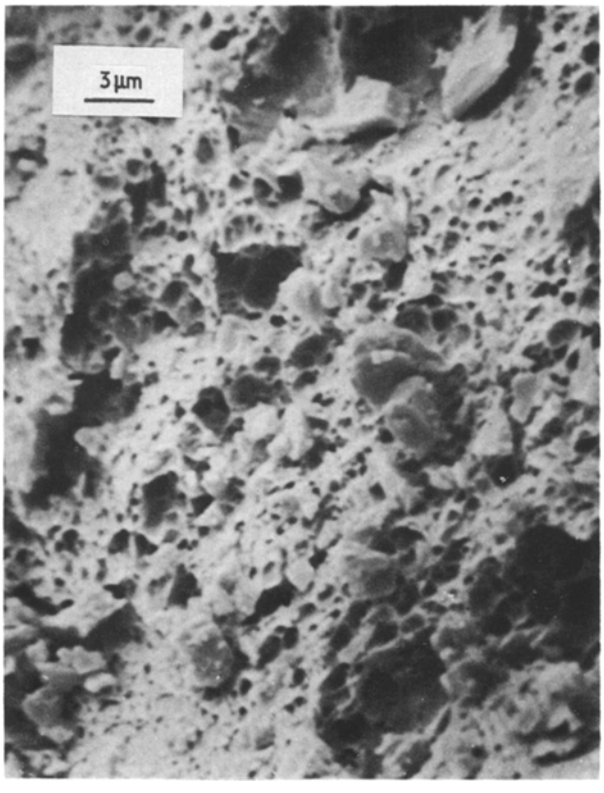

Figure 9 Scanning electron micrograph of the fracture surface of a PB composite showing debonded 3 μm SiC particles.

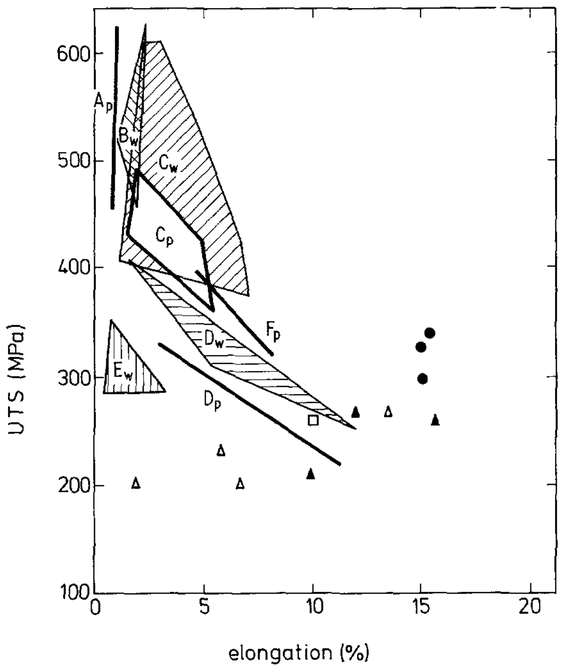

Figure 10 Comparison of present results with the literature [1, 18–21]. Literature data: subscripts p and w denote particle and whisker reinforced, respectively. (A) 2024 T6; (B) 2024 T4; (C) 6061 T6; (D) 6061 F; (E) 5083 F; (F) AlCu4Mg0.75, compocast. Present data: (△) PB, 20 μm SiC; (□) Pb, 3 μm SiC; (▲) MS, 20 μm SiC; (●) MS, 3 μm SiC.

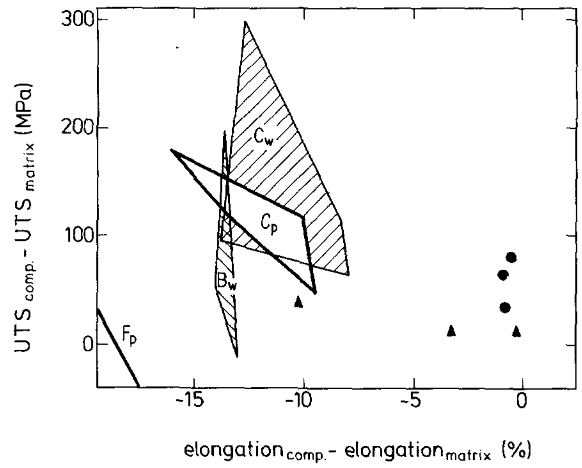

Figure 11 A plot of reinforcement effect against ductility loss. Literature data [1, 18–21]: subscripts p and w denote particle and whisker reinforced, respectively. (B) 2024 T4; (C) 6061 T6; (F) AlCu4Mg0.75, compocast. Present data: (▲) MS, 20 μm SiC; (●) MS, 3 μm SiC.

composites than for composites produced by other methods. The situation is less advantageous for 20 μm MS composites. After Sato and Mehrabian [18], SiC particles of 5 to 10 and 20 μm in compocast materials have a still smaller reinforcement effect: drastic decrease in ductility and negative to slightly positive increase in tensile strength. This confirms that 20 μm particles are better suited for improving properties such as wear resistance than for improving tensile properties. No increase of the tensile strength was found for the PB composites. The elongation always reduced considerably. Lederich and Sastry [22] produced whisker-reinforced composites using a PB-type route and obtained only a small improvement of the tensile strength with a drastic decrease of the elongation. The PB composites have quite inferior properties to those given in the literature for P/M composites. These P/M composites are produced via more sophisticated (expensive) routes, generally involving pressing above the solidus temperature of the matrix, HIPing or other steps leading to the destruction of the rapidly solidified structure.

The bond between the SiC particles and the matrix in MS composites is strong as is shown by the tensile fracture of ribbons and extruded products containing 20 μm particles. In both cases the SiC particles were broken transversely. Examination of the 20 μm particles before traction indicated that some have defects which probably initiate fracture. Only a very ductile matrix such as AlMg5 can partly eliminate the generated stress concentrations. During traction the 20 μm particles are broken and subsequently crack extension takes place between the broken particles. Defects in 3 μm particles are less critical which explains why practically none of them are broken. Fracture runs through the matrix material, leading to a fracture surface where SiC is almost absent. These observations are consistent with those of Flom and Arsenault [1], who report that the SiC particles are not broken if they are less than 10 μm in size, and that the quantity of SiC visible at the fracture surface is, in general, less than expected from the average SiC content of the material. It appears that bonding between particles (both 3 and 20 μm) and matrix was not achieved in PB products. This suggests that a strong bond can only be formed by putting the SiC particles into contact with the liquid or semi-liquid matrix.

As is shown in Section 3.1, a chemical attack of the SiC is observed in aluminium matrices containing no silicon. This occurs during contact between the SiC particles and the (partially) liquid alloy. Interaction of the SiC with aluminium above 700° C has been reported [4, 5, 23–26]. The interaction is attributed to a chemical reaction forming Al_4C_3 [23, 25, 26], to dissolution of SiC [4], or to diffusion [24]. As we did, Kohara [4] and Iseki *et al.* [26] also found that the presence of silicon in the matrix reduces the reaction. As TEM examination did not show any reaction products, it seems that the interaction occurring in the MS process is a dissolution of the SiC. Thus, the type of bond corresponds to the "dissolution and wetting bond" of Metcalfe [27]. The relatively high concentration of magnesium at the interface confirms that this element promotes wetting.

There are several advantages when applying the MS route for the preparation of composite materials. The first is that the bonding between the ceramic phase and the aluminium matrix is controlled by the compocasting step. In this step the optimum conditions for bonding can be chosen. After the melt-spinning process the composite ribbon is cut into flakes. Compared with compocasting there is an additional mixing step which will improve the homogeneity of the billet on a macroscale. A third advantage is that, compared with the PB route, mechanical mixing of the aluminium powders and the SiC particles is not needed. Large flakes can easily be handled further, whereas good mixing of aluminium powders and small SiC particles requires at least finer milling of the aluminium powders, which will give rise to a higher oxygen content in the material. Hot pressing of the aluminium powders and the SiC particles at temperatures above solidus temperatures, as is usually done with P/M processing, will destroy the good characteristics of the rapidly solidified aluminium powder particles and will lead to a cast microstructure in which intermetallic phases nucleate at the SiC particles. These brittle intermetallic phases will initiate voids during traction which will induce failure. The rapid solidification process will hinder the formation of these brittle intermetallics at the SiC interface and will give rise to a strongly refined microstructure. The small grain size should lead to a better repartition of the matrix deformation around the particles, thereby improving the ductility.

5. Conclusion

The production and properties of rapidly solidified aluminium matrix/SiC particle composites have been studied. It is shown that:

1. The combination of compocasting and melt spinning permits the production of composites with a homogeneous and strongly refined microstructure.
2. This route provides strong bonding between SiC and the aluminium matrix.
3. In most cases reinforcement effects are not found for the 20 μm SiC/Al matrix composites as the particles did fracture first under tensile load.
4. Reinforcement effects are found for the 3 μm SiC/Al matrix composites, which was coupled with a very low loss of ductility.
5. The composites prepared by powder blending and extrusion also have a homogeneous and refined microstructure but the properties are inferior due to the bad adhesion between particles and matrix. Good bonding requires a prior wetting contact with the liquid phase.

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