

## Microstructure and electrical conductivity of Al–SiC<sub>p</sub> composites produced by spray forming process

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MS received 30 January 2004

**Abstract.** Al–SiC<sub>p</sub> composites have been synthesized by spray forming process with variation in particle flow rate, size of reinforcement particles and their volume fraction. The microstructure of composites and their electrical conductivity have been investigated. The results showed a uniform dispersion of large size particulate phase in the matrix of the primary  $\alpha$ -phase with its equiaxed grain morphology. However, clustering of small size particles was observed at the grain boundary and grain junctions. The grain size of the composite materials was observed to be lower than that of the base Al-alloy. The composite materials invariably indicated their lower electrical conductivity compared to that of the monolithic Al-alloy. The electrical conductivity of composites decreased with increase in the volume fraction and decrease in size of the reinforcement particles. A high flow rate of particles during spray deposition resulted in a decrease in its conductivity. These results are explained in the light of thermal mismatch between the matrix and the reinforcement phases resulting in generation of high dislocation density. The droplet-particle interaction and resulting microstructure evolution during the spray deposition of the composites are discussed.

**Keywords.** Composites; spray forming; microstructure; electrical conductivity; grain size.

### 1. Introduction

The particulate reinforced composites have been considered to provide an attractive alternative to the traditional un-reinforced monolithic alloys. A large number of composite materials have metallic matrices reinforced with high strength, high modulus and often brittle ceramic phase particles (Lloyd 1994). A uniform dispersion of reinforcement materials in the metal matrix offers improvement in strength, elastic modulus, corrosion and wear resistance of resultant composites. Due to these properties, a particulate reinforced composite finds a wide range of applications in automotive and aerospace industries compared to their counterpart monolithic alloys. There are several reviews available on processing, properties and interfacial phenomenon in composites (Rohatgi *et al* 1986; Asthana and Tewari 1993; Srivatsan *et al* 1995; Lavernia and Wu 1996). Although there are several techniques currently existing for processing of composite materials, on a large scale these are produced by liquid metallurgy (LM) or powder metallurgy (PM) routes. In the former, the particulate phases are mechanically dispersed in the liquid before solidification of the melt. Whereas in the latter, either elemental or pre-alloyed powders are blended with particulates and compacted by hot pressing and hot extrusion processes. Relatively low solid-

liquid interface velocity associated with slow cooling rate of the melt and poor wettability of the melt with the second phase particles generate non-uniform distribution of second phase particles in the matrix phase. In addition, slow cooling rate of the melt provides dendritic solidification structure of the cast composites. Consequently, secondary processing methods are adopted to achieve their microstructural refinement as well as chemical homogeneity. Several microstructural as well as processing benefits of liquid metallurgy methods have been widely discussed (Rohatgi *et al* 1986). However, a high degree of melt superheat often used in the liquid metallurgy method results in reaction interfaces between the matrix and particulates. This effect deteriorates mechanical properties of composites. Alternatively, the reaction interfaces are although significantly reduced in powder metallurgy processing route, this processing methodology requires expensive equipments in powder consolidation by hot extrusion or hot-isostatic pressing. In addition, different processing methods also result in a composite material having properties that are different from each other (Srivatsan *et al* 1995). The powder metallurgy method is still preferred since it offers a number of product advantages.

Several shortcomings of conventional LM and PM methods of synthesizing composite materials are overcome in spray forming. In this processing technique, high energy gas jets interact with the stream of metallic melt to generate a spray of micron size droplets. These are subsequently deposited on a stationary or movable sub-

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strate to achieve a near-net shape preform. The reinforcement particles are injected in the spray of droplets at a suitable distance from the atomizer which are simultaneously deposited on the substrate to generate a preform of the composite material (Gupta *et al* 1992; Lawley and Apelian 1994; Zhang *et al* 1994). A high heat extraction rate achievable during solidification of droplets as well as on the deposition surface provides several beneficial effects to the composites produced by this technique. Rapid solidification inherent in spray deposition generates a considerable refinement in the grain size with equiaxed grain morphology and chemical homogeneity of the matrix phase. Incorporation of small volume fraction of a reactive gas in the atomization chamber results in formation of oxide particles finely dispersed in the matrix phase (Zeng *et al* 1992; Lu *et al* 1993). Composite materials produced by this technique are reported to exhibit their superior physical and mechanical properties (Srivatsan and Lavernia 1994).

In the present investigation spray deposition processing of Al-base composite materials reinforced with SiC<sub>p</sub> particles is described. The droplet-particle interaction in the spray and the resulting microstructural features of composites are discussed. The variation in electrical conductivity of the composites arising from non-equilibrium solidification of the spray-deposit is reported.

## 2. Experimental

### 2.1 Process description

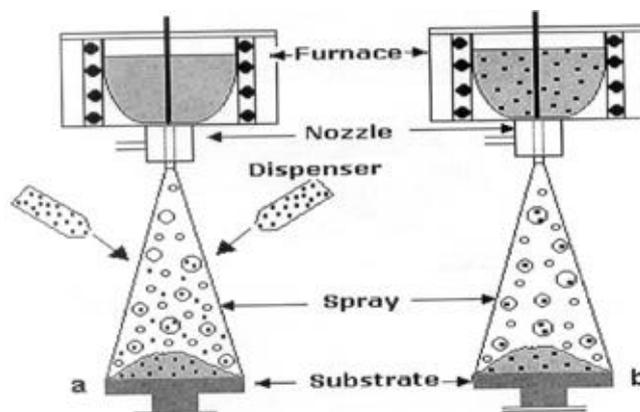
The spray deposition system basically consists of (i) a spray assembly to produce spray of fine droplets, (ii) an atomization chamber wherein spray atomization takes place in an inert gas atmosphere and (iii) a control system for the gas and metal flow and manipulation of the deposition substrate. The alloy is melted by induction heating in a graphite crucible placed on the top of the atomizer. A continuous supply of nitrogen gas is ensured during the melting process to protect the melt from oxidation. The liquid metal from a desired degree of melt superheat is poured through a flow tube of refractory material in the bottom of the crucible. A stream of molten metal, the diameter of which is the same as the internal diameter of the flow tube, then reaches into the atomization zone. In the atomization region, the molten metal stream is disintegrated into spray of droplets by gas jets in the pressure range of 0.8–2 MPa. The droplets are cooled by the gas stream and accelerated towards a deposition substrate on which they impinge and consolidate to form a coherent deposit. There are basically two ways by which second phase particulates are incorporated in the metal matrix. The first method involves pre-mixing of a desired quantity of second phase particulates in the melt in the crucible and allowing the mixture to atomize and deposit onto a

substrate. The second method features incorporation of particulates from outside i.e. second phase particulates are supplied to the matrix materials in the spray cone at some distance from the atomization zone as shown in figure 1. This results in co-deposition of particulates with the matrix metal to give rise to a coherent preform of composite material.

This is the most frequently used method for the synthesis of composite materials. The reinforcing particles are injected in the spray using a fluidized bed under the effect of high pressure inert gas. The particles interact with the liquid droplets in the spray and travel towards the substrate. These are dragged by the droplets or pull under gravity. One or more than one injectors can be used in each run. The choice of point of injection of second phase particles is governed by the consideration of the following: (i) the yield of reinforcing agents should be high, (ii) the interfacial reaction between particles and the matrix should be minimum as they come into intimate contact with each other and (iii) a selected distance of particles injection from the spray nozzle facilitates proper dispersion of the reinforcement phases in the matrix.

### 2.2 Experimental procedure

The base alloy composition used in the present work consisted of Al–4.5%Cu–1%Si–0.5%Mg (composition in wt%). The alloy was melted in a graphite crucible using an induction furnace. The melt temperature was continuously monitored using a chromel-alumel thermocouple connected to a temperature recorder. A gas regulator controlled the flow rate of N<sub>2</sub> gas for atomization. The atomization was carried out at a N<sub>2</sub> gas pressure of 1 MPa. SiC<sub>p</sub> particles of different sizes with median particle diameters varying between 6 and 45 µm were injected in the spray closer to the atomization zone of the melt. The melt to particle flow ratio was estimated to vary from 3 to 5 for different volume fractions of SiC<sub>p</sub> particles. The



**Figure 1.** Mode of addition of particles **a.** through dispenser and **b.** pre-mixing in the crucible.

spray was subsequently deposited over a copper substrate centred along the spray axis to achieve a cylindrical shape preform of 150 mm diameter with a height of 200 mm. A nozzle to substrate distance of 450 mm was invariably used in all the experimental runs. Several samples were machined from different regions of the spray-deposits for microstructural examination and measurement of electrical conductivity. The over-spray powders were also collected for further examination.

### 2.3 Materials characterization

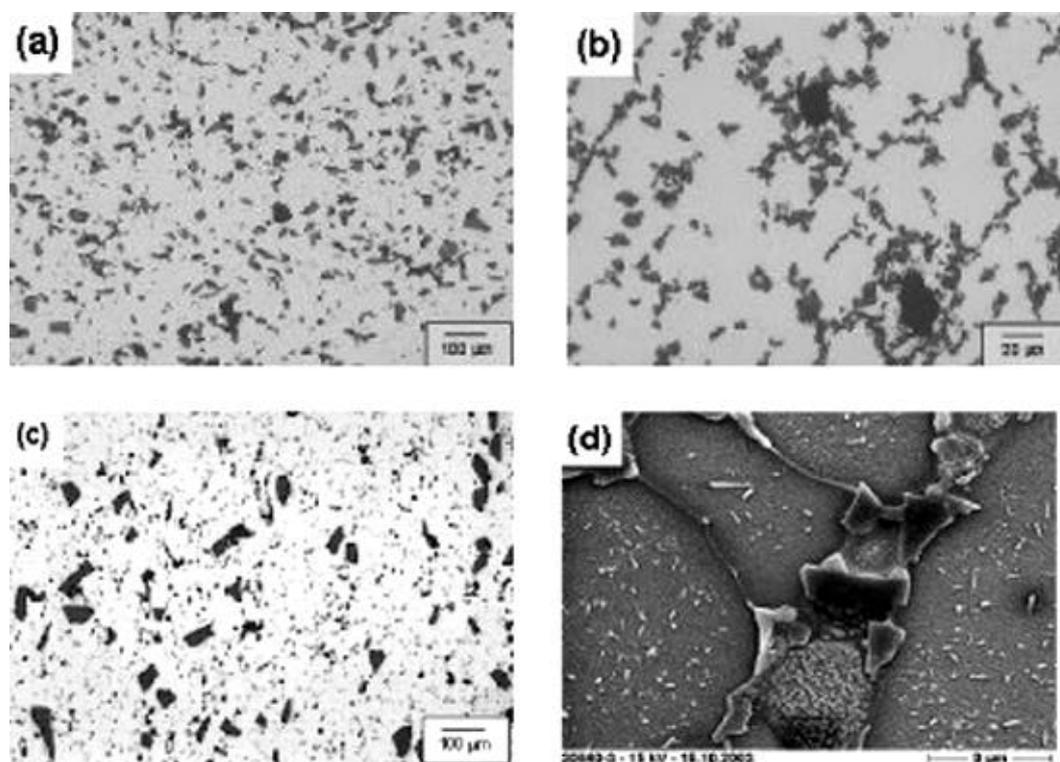
The samples for the microstructural investigation were prepared following the standard metallographic procedure of grinding and polishing. Over sprayed powder particles were mounted with a cold setting acrylic resin to facilitate metallographic preparation. All the samples were etched in a Keller's reagent. The microstructural examination was carried out using a Metallux-3 Optical Metallograph. Several samples were also examined in a Jeol 840 A Scanning Electron Microscope operating at 15 kV. The phases present in the spray formed alloys were examined by EDAX analysis technique. The electrical resistivity of the spray formed composites were measured on the samples extracted from cross-section of the deposit. These measurements were carried out as a function

of the size, flow rate and volume fraction of the reinforcement phase.

## 3. Results and discussion

### 3.1 Microstructural features

The microstructure of the spray deposited Al-SiC<sub>p</sub> composite is shown in figure 2. A uniform dispersion of second phase particles is observed in composites consisting of large size particles of the reinforcement phase. The grain boundary of the primary matrix phase is not clearly discernible. This feature is due to presence of large volume fraction of the particulate phase (figure 2(a)). However, small size particles are observed to be present along the grain boundary area in some of the regions. The grain size of the primary phase is estimated to vary from 15–25 μm in different regions of the preform and also with variation in size and volume fraction of the reinforcement phase. A considerable difference in the microstructure of the composites is observed with decrease in size of the particulate phase. This includes clustering of particles at the grain junctions and their continuous network along the grain boundaries (figure 2(b)). The microstructure of the composites also indicated formation of coarse precipitate phase (figure 2(c)). Similar results were observed as a



**Figure 2.** Microstructural features of spray deposited Al-SiC<sub>p</sub> composites showing (a) a uniform dispersion of second phase particles, (b) clustering of fine particles, (c) precipitation of CuAl<sub>2</sub> particles and (d) irregular shape of precipitate phase.

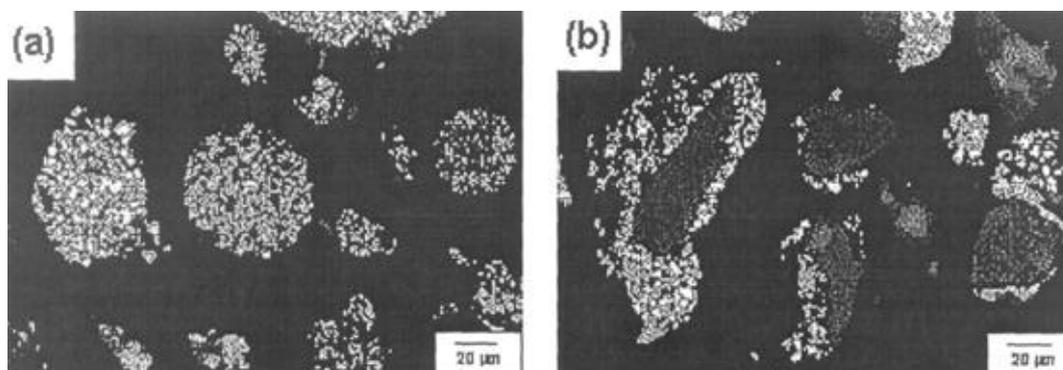
result of increasing the volume fraction of the particulate phase. The white phases as shown in figure 2(d) have been identified to be  $\text{CuAl}_2$  phases which show a tendency to nucleate at  $\text{SiC}_p$  surface. However, more often they are observed to basically decorate the grain boundaries. The grain size of the primary  $\alpha$ -Al phase varies with volume fraction and the size of the  $\text{SiC}_p$  particulates. The average grain size without reinforcement is  $45 \mu\text{m}$ . This decreases to  $15 \mu\text{m}$  for reinforcement particle size of  $6 \mu\text{m}$  with 8.5 vol%. Whereas, the grain size was estimated to be  $24 \mu\text{m}$  for a particle size of the reinforcement phase of  $16 \mu\text{m}$  with the same volume fraction as in the previous processing condition.

The particle–droplet interaction in the spray and also on the deposition surface plays a crucial role in evolution of microstructure during spray deposition. Several interesting features were observed in the microstructure of the over-spray powder particles. These are shown in figure 3 for both small and large size  $\text{SiC}_p$ . The particles of the base Al-powder show a typical cellular–dendritic morphology in contrast to an equiaxed grain morphology of the particles consisting of fine particulate phase. This result indicates considerable microstructural modification of powder particles associated with heterogeneous nucleation of droplets in presence of particulate phase. A distinct nucleation site is observed on surface of  $\text{SiC}_p$  particles for solidification of droplets in the spray. The small size particulates are observed to be entrapped in the droplets prior to their solidification. In addition, the droplets are observed to be splashed on the surface of the large size particles of  $\text{SiC}_p$  as shown in figure 3(b). This effect arises due to different velocities of droplets vis-à-vis that of the  $\text{SiC}_p$  in the spray.

The microstructural features of spray formed particulate composite materials have been widely reported in literature (Wu and Lavernia 1991). The microstructure of  $\text{SiC}_p$  reinforced 6061-Al alloy composites revealed the presence of an equiaxed grain morphology consistent with the observations made in the present investigation.

However, the average grain size was reported to be  $20 \mu\text{m}$  with a variation of  $\pm 6 \mu\text{m}$ . The result showed a decrease in the as spray deposited grain size with an increase in volume fraction of  $\text{SiC}_p$  particles. The grain size of un-reinforced material was reported to be  $34 \mu\text{m}$  compared to a small grain size of  $22 \mu\text{m}$  of a composite material containing 5.2 vol%  $\text{SiC}_p$ . The microstructure of both the reinforced and un-reinforced materials was observed to have partially recrystallized grain structure with well defined subgrain boundaries. This result is consistent with the observations made on Al– $\text{SiC}_p$  composite in the present work. It is well known that there exists a certain amount of porosity in the spray deposited MMCs that result in a lower density compared to the theoretical density of the alloy. Except for the common reasons for the formation of porosity in spray deposited preform, addition of reinforcements during spray deposition cause some further effect on the formation of porosity. For example, an increase in the viscosity of the melt would cause insufficient flow of the melt. In addition, the difference in coefficient of thermal expansion between the ceramic phase and the matrix will cause contraction misfit between the components during the solidification. This effect has been reported to generate interfacial micro-porosity. The larger the reinforcement particulate, the more such micro-porosity generated. These porosities will lower the bonding of interface between the reinforcement and matrix phase. Finally, the residual gas on the rough surface of the reinforcement particles may enter into the inner part of the droplets along with the particles. The gas is not easy to be eliminated and will agglomerate to form the gas filled porosity. In case the gas does not interact with the matrix alloy, such porosities will not easily be closed during cold or hot working of the composites.

The mechanisms involving particle distribution during spray deposition processing of particulate reinforced composites have been widely investigated (Chou and Oki 1987; Gupta *et al* 1991). The results of the published literature reveal that the kinetic and thermodynamic factors



**Figure 3.** Micrographs showing droplet–particle interaction during atomization stage giving rise to (a) cellular–dendritic solidification structure and (b) nucleation on surface of  $\text{SiC}_p$  particles.

govern the entrapment or rejection of particles by moving solid-liquid interface. The criteria considered for particle entrapment are based on either the thermal conductivity differences or thermal diffusivity ratio of the reinforcement particles and the matrix phase. In addition, a critical velocity criterion has been used to describe the engulfment of the particle by the moving solidification front. The above criteria were originally for the steady state conditions present in equilibrium solidification processes. However, the condition present during the spray deposition processes are far from being at equilibrium and none of the above criteria can fully describe the nature of particle distribution in spray deposition processing. However, in this process the second phase particles are incorporated into the matrix in various stages. Initially, the particles penetrate the atomized droplets during co-injection and remain entrapped in the matrix during subsequent deposition. This feature is clearly observed in the microstructure of powder particles shown in figure 3(a). Alternatively, particles can remain on surface of the atomized droplets and are entrapped by the matrix on impact. In the mechanism of mechanically driven entrapment, a particle is approached by multiple solidification fronts. As the solidification front converge, a capillary region forms around the reinforcing particles. At this point, the particles experience repulsive forces from the adjacent solidification fronts, capillary forces and convective forces of the fluid arising from the impact of the droplets (Lavernia and Wu 1996). All the three forces push the particles out of the capillary region. As the particle is displaced, subsequent impact by high velocity droplets leads to a mechanically stimulated engulfment of the second phase particles. This behaviour of the spray deposition process results in a uniform distribution of fine ceramic particles in metal matrix of composites.

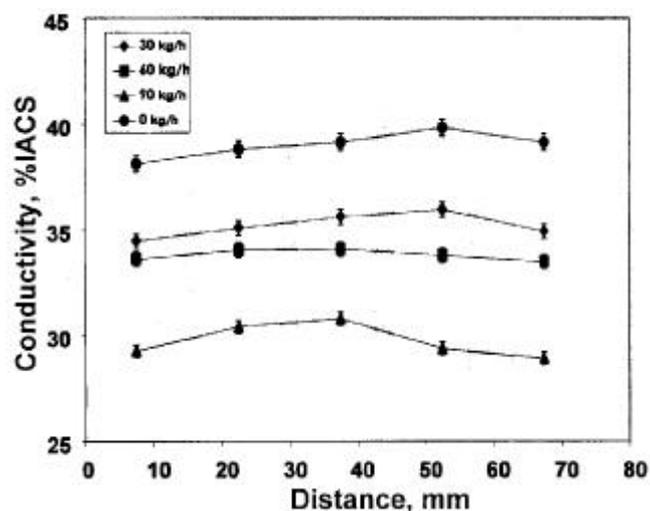


Figure 4. Variation in electrical conductivity of the composites along deposit section as a function of particle flow rate.

### 3.2 Electrical conductivity

Variation in electrical conductivity of the composite materials along their cross-section is shown in figure 4. These measurements were made in composite materials produced employing three different flow rates of silicon carbide particles in the spray. The electrical conductivity of the base alloy spray-deposit is also shown for comparison. The results indicate a relatively high electrical conductivity throughout the cross-section of the deposit of the base alloy compared to that of the composite materials. This result corresponds to different flow rates of powder injection at different distances from the centre of the preform. The electrical conductivity decreases as the flow rate of SiC<sub>p</sub> increases in the spray. The conductivity is observed to be minimum for a highest flow rate of 90 kg/h of the particulates. The variation in the surface and centre of the deposit is observed to be maximum for the highest flow rate of particles. This variation is attributed to the uneven distribution of SiC<sub>p</sub> particles in the peripheral and central regions of the spray-deposit. Such features were also observed in the microstructural features of the deposit examined in the samples extracted from different locations of the deposit. The variation in electrical conductivity of the composite material was also examined as a function of particle size of silicon carbide. The results show that the conductivity of the composite increases with increase in size of the particle. This effect of SiC<sub>p</sub> size on the electrical conductivity of the spray formed billets at a constant volume fraction of 8.5% is shown in figure 5. It is interesting to note that there is a rapid rise in conductivity of the composite as the particle size increases from 6 to 18 μm. With subsequent increase in size of the particle the conductivity increases slowly. The highest value is observed in the composite materials made using SiC<sub>p</sub> of 60 μm. The volume fraction of SiC<sub>p</sub> is also observed to influence the electrical conductivity of the composite material. The result presented in figure 6 exhibits a linear relationship between electrical conductivity and volume fraction of the SiC<sub>p</sub> particles.

The electrical property of the Al-SiC<sub>p</sub> composite is an important parameter because of its application in electrical circuits and support structures in optical devices. SiC<sub>p</sub> possesses very low electrical conductivity and therefore its incorporation in Al alloys leads to a drastic decrease in its electrical conductivity. The reduction in conductivity of composite materials is basically attributed to high dislocation density near the interface and also the elastic discontinuity at the interface. In addition there is a considerable difference in thermal expansion coefficient of the matrix and the reinforcement phase. This mismatch in properties of composite materials creates a high dislocation density at the interface due to internal stresses. Such crystal defects play a major role in acting as scattering sites to the electrons. Therefore, the electrical conductivity is affected by volume fraction and also by the size of

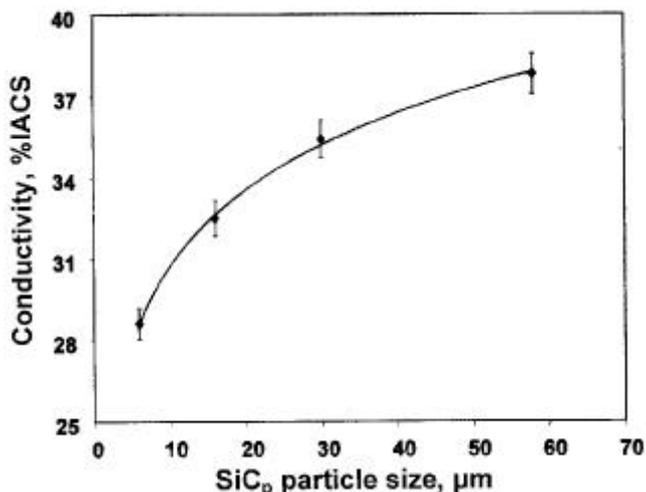


Figure 5. Variation in electrical conductivity of composites as a function of particle size.

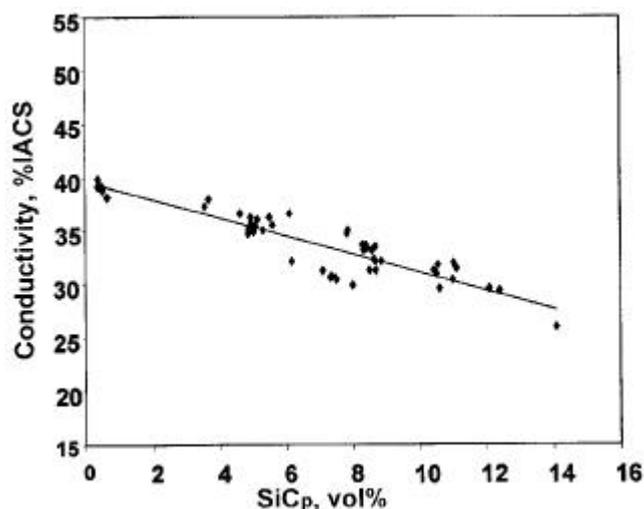


Figure 6. Variation in electrical conductivity of composites as a function of volume percent of reinforcement particles.

reinforcement phases in composite materials. A small size of the reinforcement phase results in large interfacial area and thus a high internal stress and resulting dislocation density. The variation of conductivity at different distances in the billets is also a measure of the variation in SiC<sub>p</sub> content along the cross sectional area of the deposit other than variation in porosity of the deposit.

#### 4. Conclusions

The spray deposition process has been used to synthesize Al-SiC<sub>p</sub> composites. The volume fraction and particle size of SiC<sub>p</sub> have been varied. The microstructural features of composites and their electrical conductivity have been

investigated. The following conclusions are derived from the results of the present work.

(I) The microstructure of the spray deposits reveals equiaxed grain microstructure of the primary  $\alpha$ -phase. The large size SiC<sub>p</sub> particles are shown to be uniformly dispersed in the matrix phase. However, clustering of fine particles along the grain junctions and grain boundary areas is observed. The microstructure of over-spray powder particles exhibits cellular-dendritic morphology.

(II) The grain size of composites is observed to vary from 15 to 25  $\mu\text{m}$  compared to the grain size of 45  $\mu\text{m}$  of the spray deposits of the monolithic Al-alloy. A considerable refinement in grain size of the matrix phase is observed with an increase in flow rate of SiC<sub>p</sub> particles in the spray.

(III) The electrical conductivity of composite materials is observed to be invariably lower than that of the monolithic Al-alloy. The electrical conductivity of composite materials decreases with increase in particle flow rate and the volume percent of the reinforcement phase during spray deposition. However, the electrical conductivity is observed to increase with increase in particle size for the same volume fraction of SiC<sub>p</sub>. This effect is attributed to the large mismatch of the thermal expansion coefficient between the Al-alloy and reinforcement SiC<sub>p</sub> phase.

#### Acknowledgements

The authors gratefully acknowledge the Centre for Advanced Study, Department of Metallurgical Engineering, Banaras Hindu University, Varanasi and the National Metallurgical Laboratory, Jamshedpur, for providing the necessary facilities to carry out the present work.

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