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Cast aluminium alloy composites containing copper-coated ground mica particles

DEONATH
Institute of Technology, Banaras Hindu University, Varanasi 221 005, India

P. K. ROHATGI
Regional Research Laboratory, Council of Scientific and Industrial Research, Trivandrum 695 019, India

An electroless method of coating copper on ground mica particles using copper sulphate solution is described. The effects of time of sensitization, $PdCl_2$ concentration and time of stirring the activated particles in electroless solution, on the extent of copper coating on mica particles are reported. Using this method it is possible to deposit up to about 35 wt% copper on mica particles. A process for making cast aluminium alloy–mica particle composite alloys using these coated particles is also described. The process involves stirring the copper-coated mica particles into liquid alloys using an impeller, and casting the melts containing suspended mica particles in suitable permanent moulds. Coating of copper on mica particles makes possible the dispersion of ground mica particles in molten aluminium alloys with high recoveries which is otherwise difficult, even when magnesium is added to the surface of the melts. Copper coating on ground mica particles masks the basal planes, and apparently increases their wettability with aluminium alloy melts. Recoveries of ground mica particles in composite castings made using copper-coated mica particles are as high as 80%, which is three times higher than the corresponding recoveries in the castings made using uncoated particles. The mechanical properties of cast aluminium mica composites made using copper-coated ground mica powders are adequate for a variety of bearing and antifriction applications.

1. Introduction

Solid lubricants dispersed in metallic matrices result in self-lubrication composite materials suitable for antifriction applications. Bearings of cast aluminium alloy–graphite [1–9] and aluminium alloy–mica [10] composites made by dispersing these particles in liquid aluminium alloy have been shown to work satisfactorily under boundary lubrication conditions. Aluminium alloy–mica composite bearings made using cut mica powders work even under semi-dry and dry friction conditions [11].

It has been possible to disperse uncoated cut mica powder (mica powder produced by milling mica sheets across their cleavage planes) in molten aluminium alloys in the presence of magnesium. However, appreciable amounts of uncoated ground mica particles (Fig. 1), produced by grinding mica sheets in a ball mill, could not be dispersed under similar conditions. Ground mica particles have a much higher aspect ratio (ratio of diameter to thickness) compared to cut mica powder, and a much greater fraction of their surface is bound by basal planes which apparently are not wetted as readily by molten aluminium as the prismatic non-basal planes exposed by milling. In clays which have a layered structure like mica, it is known that adsorption is much easier on non-basal planes due to unbalanced electrical charges. As a parallel example it is shown that yttrium is more effective [12] as a nucleant in liquid metals when the prismatic plane is exposed than when the basal plane is exposed, indicating a greater wettability between prismatic planes and metals in general. It has been

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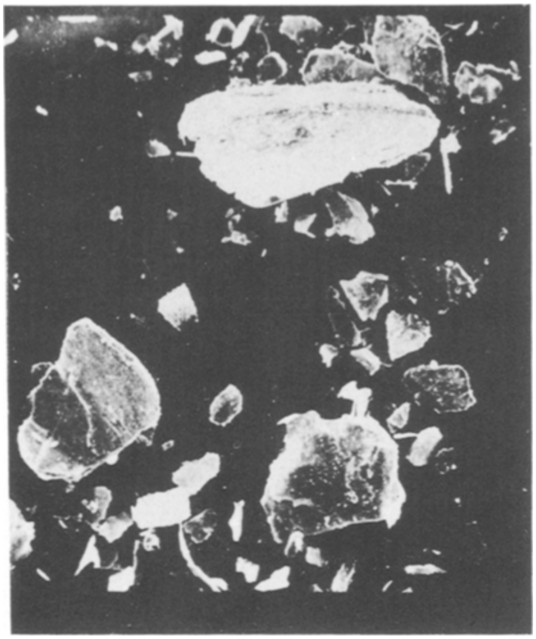


Figure 1 Scanning electron microphotograph of ground mica powder (× 230).

reported that nickel coating [2, 7] and copper coating [5, 8] on graphite particles helped in their dispersion in aluminium alloy melts. Therefore, in this paper the use of copper coatings on ground mica particles to improve their dispersability in aluminium alloy melts has been explored. Ground mica powder is cheaper than cut mica powder and is also likely to impart better lubrication properties when dispersed in aluminium alloys than the cut mica particles. In the case of graphite it is well known that flake graphite is a better lubricant than granular graphite.

Dispersion of uncoated cut mica particles in aluminium alloy has been described elsewhere [10]. There is no report in the literature on the process of coating copper on mica particles, and on dispersion of copper-coated ground mica particles in liquid aluminium alloys. The present paper describes a process for coating copper on ground mica particles, and making cast aluminium alloy ground mica particle composites using copper-coated mica particles. Limited experiments were also carried out on copper coating of cut mica particles and dispersion of copper-coated cut mica particles in aluminium alloys. Tensile strength and ductility of the resulting composites were also measured and compared with the properties of alloys made using uncoated cut mica particles.

TABLE I Chemical composition of ground mica powder

Constituents	Percentage
SiO_2	48.00
Loss on ignition	4.91
Fe_2O_3	3.20
Al_2O_3	33.17
CuO	1.65
MgO	1.20
K_2O	6.83
Na_2O	0.72
Li_2O	0.32

2. Experimental procedure

2.1. Materials

Aluminium–copper alloys with the following chemical compositions were used as the base alloys for making the composites.

Al: Balance, Cu: 3.90 to 4.50 wt%,
Si: 0.31 to 0.35 wt%, Mg: 0.42 to 0.45 wt%,
Fe: 0.10 to 0.15 wt%.

This alloy is similar to LM-11 casting alloy except for the higher magnesium content of about 0.4% to increase the dispersability of mica. This modified alloy composition is similar to Duralumin type alloy.

2.2. Mica powder

(1) Ground mica powder was obtained from Mysors Micanite Industries, Bangalore. This powder was produced by heating muscovite mica sheets to 150° C and grinding them in water. The chemical composition of ground mica powder is given in Table I. Most of the experiments were done with as-received ground mica powder ranging in size from 15 to 150 μm (referred to as 65 μm average size mica for convenience in the text).

(2) Mica powder was produced by cutting muscovite mica sheets across the cleavage plane using a milling cutter on a milling machine. This mica powder is referred to as cut mica powder in the present paper and its composition was similar to the composition of the ground mica powder.

2.3. Chemicals

BDH LR grade chemicals used for copper coating on mica particles included $SnCl_2$, NaOH, $PdCl_2 \cdot 5H_2O$, $NaK\,C_4H_4O_6 \cdot 4H_2O$, HCHO (37 to 41% w/v).

2.4. Copper coating on mica particles

Both ground and cut mica powders described above were coated using the electroless method. About

50 g of mica particles were stirred in one litre of aqueous solution containing $10\,g\,l^{-1}$ of $SnCl_2$ and $40\,m\,l^{-1}$ of concentrated hydrochloric acid. The solution containing mica particles was filtered, and the mica powder obtained as residue was activated by stirring it for 10 min in a 250 ml solution containing $PdCl_2$ ($0.5\,g\,l^{-1}$) and concentrated hydrochloric acid ($1\,m\,l^{-1}$). The solution was decanted and the powder washed throughly with distilled water to remove any traces of $PdCl_2$ solution adhering to the particles. The powder was then stirred in a solution for electroless plating containing $10\,g\,l^{-1}$ $CuSO_4 \cdot 5H_2O$, $10\,g\,l^{-1}$ NaOH, $50\,g\,l^{-1}$ sodium potassium tartarate and $10\,ml\,l^{-1}$ HCHO (37 to 41% w/v) for 30 min or till the blue colour of the solution disappeared. The copper-coated mica particles were filtered and dried in a hot oven maintained at 150° C.

2.5. Preparation of the composite

The experimental set-up used for making cast composites consisted of a mechanical stirrer and a holding furnace and is described elsewhere [10]. About 3 kg of aluminium alloy of the composition given above was melted in a super salamander (a class of clay graphite) crucible using an oil fired furnace. The metal was super-heated to 800° C. The melt was then de-gassed by bubbling nitrogen through it at a rate of $10\,l\,min^{-1}$. De-gassing was carried out while the molten metal was kept in an electrical resistance holding furnace at about 700° C.

The mechanical stirrer was then inserted into the molten metal and rotated at 1000 rpm to create the necessary vortex. Copper-coated mica-particle (containing about 30% copper as coating) were added onto the vortex at an approximate rate of $50\,g\,min^{-1}$. Metal was stirred for at least half a minute after the addition of powder was complete, and then de-gassed by bubbling nitrogen at a rate slower (about 2 to $3\,l\,min^{-1}$) than that used for de-gassing before dispersion. The de-gassing was accompanied by further stirring of the melt at about 500 rpm for a further 1 min. Stirring was then stopped and the stirrer was taken out of the crucible. The metal was taken out of the furnace, stirred with a graphite rod and poured into permanent moulds.

Limited attempts were made to prepare composites to which magnesium was added to the surfaces of the melt along with copper-coated mica particles.

2.6. Chemical analysis

Copper-coated mica particles and cast aluminium alloy–mica particle composites were analysed to determine the amount of copper coated on mica, and the amount of mica present in the composite. The amount of copper in copper-coated mica was determined by dissolving the particles in nitric acid solution. For determination of mica in composites, the drillings of composites were first dissolved in hydrochloric acid. The solution was filtered and the residue dissolved in nitric acid. The residue of this solution gives mica powder.

3. Results and discussions

The various factors affecting the extent of copper coating on mica are described below.

3.1. Effect of time of sensitization

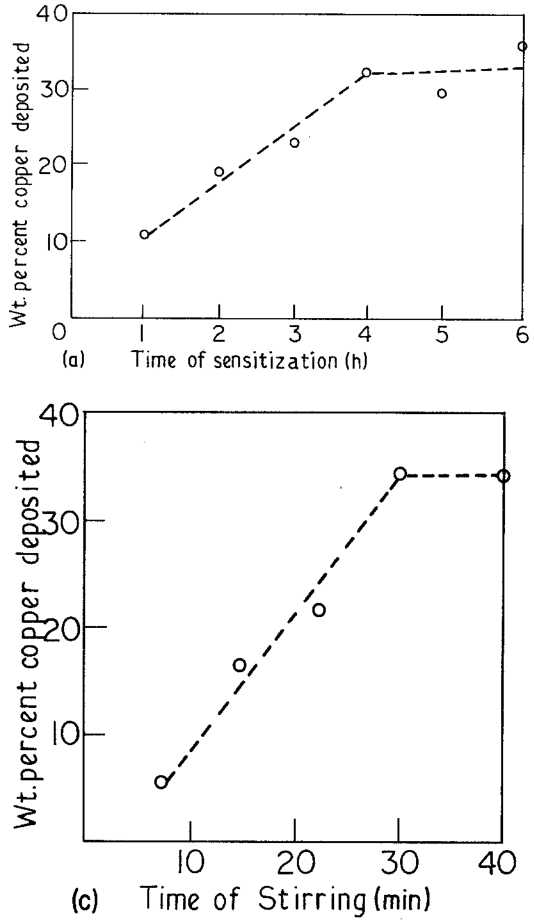
The effect of time of sensitization on the weight per cent of copper deposited is shown in Fig. 2a. Apparently, as the time of sensitization increases, the surface becomes more and more suitable to receive a coating. However, after about 4 h of sensitization, there appears to be no significant increase in the amount of copper deposited with further increase in sensitization time.

3.2. Effect of concentration of $PdCl_2$

The effect of $PdCl_2$ concentration on the weight per cent of copper deposited is shown in Fig. 2b. It can be seen that the change in weight per cent of copper deposited with a change in $PdCl_2$ concentration from 0.2 to $0.5\,g\,l^{-1}$ of $PdCl_2$ is from 28 to 36.

3.3. Effect of stirring time in electroless bath

Fig. 2c shows that as the stirring time of mica particles in the electroless bath increases, the percent of copper deposited on the mica particles also increases up to a stirring time of about 30 min. The coating reaction starts as soon as the mica particles activated by $PdCl_2$ are dispersed in the coating solution. Palladium metal at the surface of mica particle acts as a catalyst. After the start of the coating reaction it proceeds autocatalytically. After putting the activated powder in the electroless bath, the bath should be stirred continuously to obtain a uniform coating of copper on the mica particles. The colour of the electroless solution changes progressively as the reaction progresses due to depletion of copper. The reaction is gener-



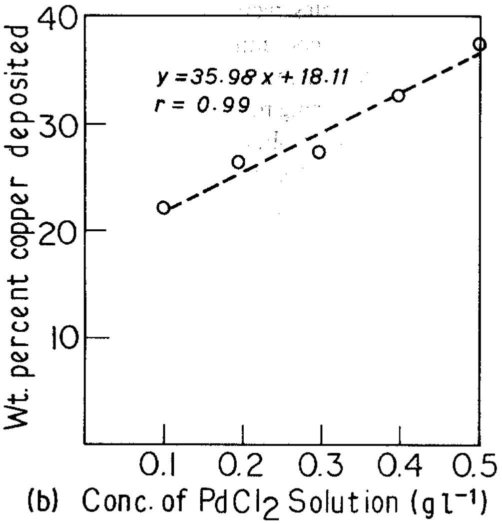


Figure 2 Weight per cent copper deposited as a function of: (a) time of sensitization in $SnCl_2$ solution, (b) concentration of $PdCl_2$ solution and (c) time of stirring in electroless bath.

ally completed within about 30 min during which time about 35 to 36 wt% of copper is deposited. After this time period there is no significant increase in the copper deposited on the particles due to (a) depletion of copper in the solution and (b) the tendency for precipitation of free copper in the solution instead of a further build up of the coating.

3.4. Re-use of the electroless bath

In the coating reaction, formaldehyde (HCHO) reduces $CuSO_4$, and copper is deposited on activated mica particles. It was found that after completion of the coating reaction, the pH of the solution had decreased to 9. After adding sodium hydroxide in an amount sufficient to maintain the pH at 12.5, it was possible to coat copper on to mica by replenishing the solution with $CuSO_4$ and formaldehyde.

Copper coating was visible on mica particles since it changed the colour of mica particles from white to copper brown, and this effect was readily documented in several coloured pictures. These pictures show that there are a few regions on the surfaces of some mica particles on which no copper is deposited. As will be described later, even this patched coating of copper was adequate to impart requisite wettability to ground mica particles.

3.5. Preparation of composites

Fig. 3 is a typical macrophotograph of a machine cut surface of a bearing machined from a casting

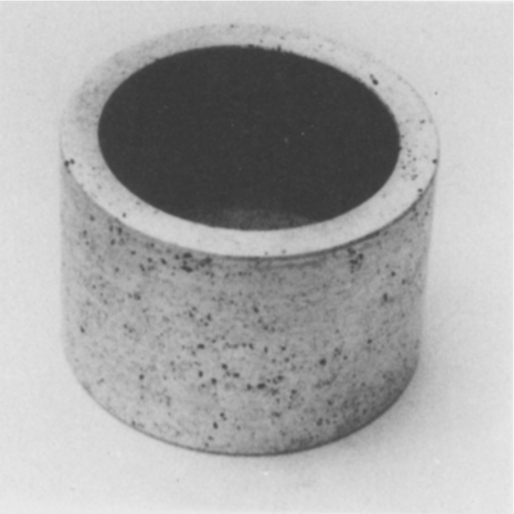


Figure 3 Macrophotograph of a machine cut surface of a bearing machined from a casting made using copper-coated ground mica particles (× 1).

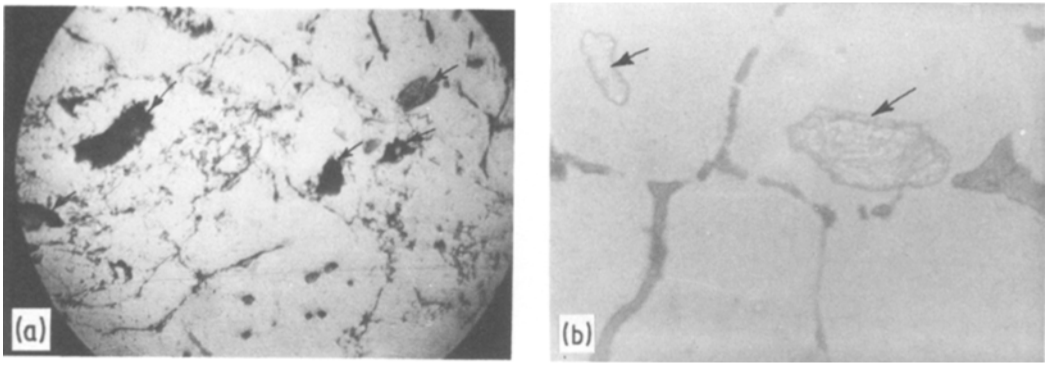


Figure 4 Microphotograph of castings containing ground mica particles (a) (× 230) and (b) (× 350).

made using copper-coated ground mica particles. It shows a uniform dispersion of dots due to mica particles in composite castings. A few mica particles have apparently agglomerated or have voids around them giving rise to large-sized dots. The microscopic distribution of ground mica particles in the matrix of alpha-aluminium and second-phase particles is shown in Fig. 4. Fig. 5 shows the secondary-electron image and X-ray images of aluminium, potassium and copper of a region of the casting and mica particle. The potassium image clearly outlines the section through the flake-shaped mica (Fig. 5b). This is also confirmed by the aluminium image (Fig. 5c) showing the absence of aluminium in the region where mica is present. The copper image (Fig. 5d) shows some residual copper coating in a few places around the mica particles. This picture also shows the presence of copper in second-phase particles near the mica particle.

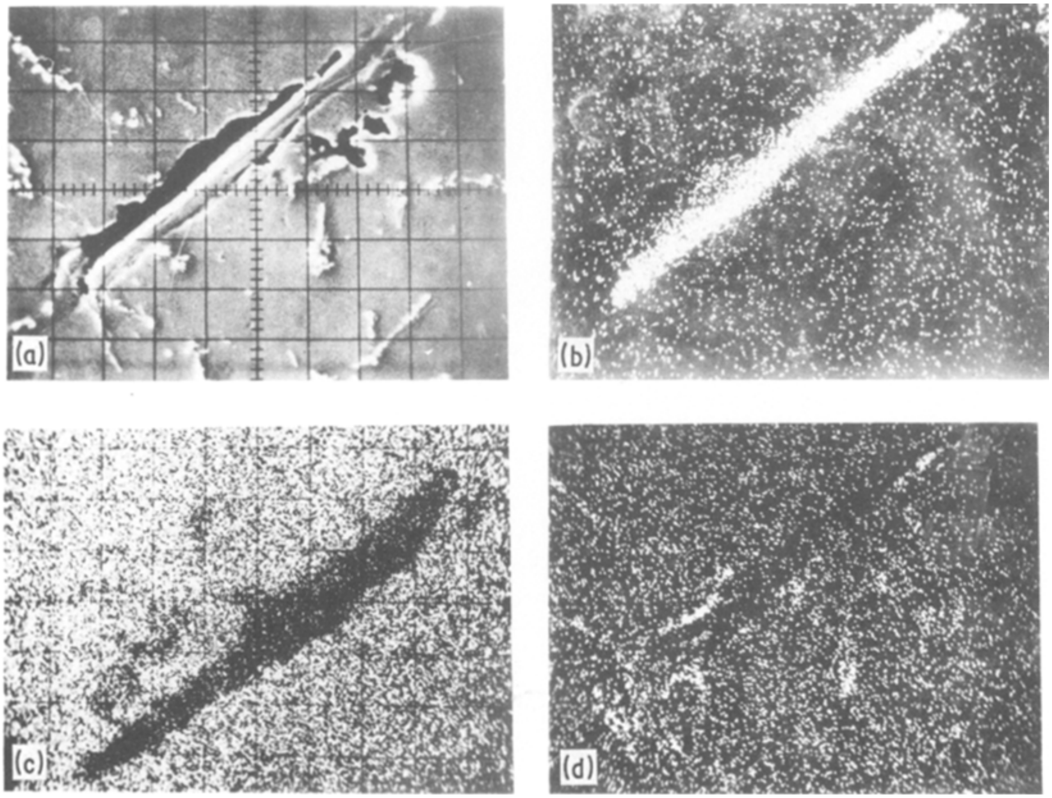


Figure 5 Scanning X-ray micrographs of a typical region in the casting containing a ground mica particles (× 360): (a) secondary-electron image, (b) X-ray potassium K-image, (c) X-ray aluminium K-image and (d) X-ray copper K-image.

TABLE II Recoveries of mica particles in composite castings

Sample number	Type of mica powder	Percentage magnesium added to the surface of the melt	Percentage of mica particles added to surface of the melt (a)	Percentage of mica particles retained in casting (b)	Percentage recovery $\left(\frac{a}{b} \times 100\right)$
1	Uncoated cut mica	3	2	1.41	70.5
2	Copper coated cut mica	Nil	2	1.65	82.5
3	Copper coated cut mica	3	2	1.80	90
4	Uncoated ground mica	3	1	0.06	6
5	Uncoated ground mica	3	2	0.56	25
6	Copper coated ground mica	Nil	2	1.60	80

3.6. Recovery of mica particle in the composite casting

The recovery of mica particles in the aluminium–mica particle composite castings made using different types of mica particles with different treatments are listed in Table II.

Table II shows that the recoveries of uncoated ground mica particles were quite low (25%) compared to uncoated cut mica (70%) under similar conditions. Table II also shows that in the case of ground mica the recoveries of mica in the castings increase from 25 to 80% as a result of copper coating. Even in the case of cut mica particles, copper coatings on mica increase their recoveries from 70 to 90%. Copper coatings on both ground and cut mica particles improve their wettability with liquid aluminium alloys, thereby facilitating their introduction in the melt, and increase their density, thereby reducing the speed of flotation of the particles. This appears to be the reason for higher recoveries of coated particles in the castings compared with uncoated particles. The recoveries of coated cut mica particles and coated ground mica particles are similar (about 80%) under identical conditions. This indicates that in the presence of copper coatings the wettability of ground and cut mica particles with aluminium alloy melts are similar. Apparently copper masks the relatively unwettable basal planes in the case of ground mica particles, and the molten aluminium comes in contact mainly with the copper coating during the dispersion of both cut and ground mica particles.

The recovery of the magnesium, added as pieces to the surface of the melt while adding mica, is generally less than 40% in the castings. These low recoveries are due to (a) no flux or sulphurous atmosphere being used and (b) due to very high transient concentrations of magnesium built in the upper layer of stirred liquid immediately following the melting of magnesium pieces.

3.7. Optimum conditions for making the composites using copper-coated mica powders

When copper-coated mica particles are dispersed into the melt, the melt temperatures should be kept between 730 and 750° C to avoid quick dissolution of the copper coating. Likewise, the rates of addition of coated mica powder and mixing with the melt should be greater (compared with uncoated mica particles), otherwise premature dissolution of the coating, leading to the rejection of the mica particles, takes place. This phenomenon has also been reported by French *et al.* [13] for Ag–Ti–MgO composites. Addition of magnesium to the surface of the aluminium alloy melts along with copper-coated mica particles helps in dispersion of such particles, and results in higher recoveries of mica in the castings (than the recoveries obtained using only copper-coated mica particles). Magnesium apparently maintains the increased wettability of mica particles with molten aluminium alloys even after the copper coating begins to dissolve.

TABLE III Results of tensile tests on aluminium alloy–mica particle composites made using copper-coated and uncoated mica particles

Sample number	Composition	Whether coated or uncoated mica particle	Tensile strength (kg mm^{-2})	Elongation (%)
1	Al–3.9 wt% Cu–0.3 wt% Si–o.45 Mg	No mica	24.30	10.70
2	Al–5 wt% Cu–1.5 wt% Mg–0.3 wt% Si–1.38 wt% mica	Copper-coated cut mica	20.10	3.50
3	Al–3,9 wt% Cu–0.3 wt% Si–1.5 wt% Mg–1.38 wt% mica	Uncoated cut mica	20.80	1.65
4	Al–5 wt% Cu–0.3 wt% Si–0.45 wt% Mg–1.8 wt% mica	Copper-coated cut mica	12.8	4.82
5	Al–5 wt% Cu–0.3 wt% Si–0.45 wt% Mg–1.8 wt% mica	Copper-coated ground mica	11.90	4.90

3.8. Mechanical properties

The results of tensile tests on aluminium alloy–mica particle composites made using copper-coated mica particles are listed in Table III. This table also contains the results on composites made using uncoated mica powder under similar conditions for comparison.

Comparison of tensile strength and ductility of the composite made using copper-coated cut mica powder and uncoated cut mica powder (given in Table III) reveals that the ductilities are higher in the case of the former than the latter, whereas there is no appreciable difference in tensile strength. It can be inferred from Table III that the copper coating has been effective in improving the ductility of the composite. This suggests that high ductilities obtained in castings made from ground mica can also be attributed to some extent to copper coating on these particles. The tensile properties of Al–3.9 wt% Cu–1.8 wt% mica composite made using copper-coated ground mica particles are similar to the properties of composites made using cut mica particles which have already been shown to successfully run as bearings under dry and semi-dry conditions [10]. This suggests that cast aluminium–mica particle composites made using copper-coated ground mica particles have adequate strength to be used as bearings in several applications.

4. Conclusions

(1) After suitable sensitization and activation treatment, both cut mica particles and ground mica particles can be coated with up to 40 wt% copper using a copper sulphate solution of electroless plating.

(2) It is possible to disperse copper-coated ground mica particles in Al–4 wt% Cu alloy melts and retain above 80% of the mica added to the melt, in permanent mould castings of sizes up to 150 mm height and 62.5 mm diameter. It is difficult to obtain recoveries above 25% mica when uncoated ground mica particles are dispersed.

(3) Recoveries of mica in the composite castings made using copper-coated mica particles (even without magnesium additon to the melt) were better than the recoveries of uncoated mica particles which were added along with magnesium addition. A combination of copper coating and magnesium addition to the surfaces of the melts gave the best recoveries of mica particles in castings.

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