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Liquid Metallurgy Synthesis and Thermo–electrical Characterization of Copper–aluminum Metal Matrix Composite

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Abstract. Samples of copper-aluminum reinforced metal matrix composite (MMC) were prepared by liquid metallurgy method using micron–sized silicon carbide (SiC) particulate. The resultant MMC samples were characterized to determine their thermal, electrical and mechanical properties in respect to varying particle sizes (212, 425, 710 and 1200 μm) of the SiC. The analyses of the results obtained showed that the thermal conductivity of the composite increased with decrease in particle size and volume fraction of the SiC. Also with decrease in grains size, high thermal conductivity was achieved by increasing the volume fraction. The results obtained in this study showed that alloying Cu matrix with aluminium is effective in reducing the interfacial reactions of a typical Cu-SiC matrix composite. The synthesized MMC samples also possessed a combination of high thermal and electrical conductivities with a low coefficient of thermal expansions which is synonymous to a low tensile strain at a maximum load. These properties were achieved for a 60%Cu/Al(40%SiC) at 212 μm, 50%Cu/Al(50%SiC at 12 μm and 70%Cu/Al(30%SiC) at 710 μm. The microstructural evaluation using optical microscopy (OM) indicated good dispersion of the SiC particles in all the samples which consequently enhanced the microhardness obtained in the MMC samples.

Introduction

Lightweight constructions are widely established in the field of high-tech applications such as automobile and aerospace industries. Copper and aluminum based materials are of high interest in these applications due to their properties, availability and relatively low cost. In recent years, a great interest is being directed to the metal-matrix composites (MMCs) due to their superior mechanical, thermal and electrical properties, weight saving, increased component life, and improved recyclability [1 – 2]. These properties of MMCs make them respond to unprecedented demands from technology due to rapidly advancing activities in aircrafts, aerospace, automotive industries and electronic devices. However, the fabrication cost of these materials has made them slightly more expensive than their conventional alloy competitors. Therefore, reinforcements can considerably reduce the costs of the MMCs. While powder metallurgy processing has been described very expensive, casting has been recognized as the most economical route [3 – 4].

Silicon carbide (SiC) particles which are currently being used as MMCs reinforcement due to their availability and cost [5], offer enhanced improvement of properties in MMCs [6 – 7]. Copper MMCs reinforced with silicon carbide (SiC) possess higher modulus of elasticity, yield strength and wear resistance than the corresponding non–reinforced matrix alloy system and are currently being used for products such as break discs and bicycle frames [8]. They have also found applications in microelectronics for thermal management and packaging purposes due to their high thermal conductivity and low thermal coefficient of expansion [9].

Some important properties of composites, such as elastic modulus in various directions, tensile strength, coefficient of thermal expansion, and thermal and electrical conductivity, can be estimated from the reinforcement arrangement and the volume fraction of the matrix and reinforcing phase. Although most applications of composites are in high-performance structures, their use is growing rapidly in the electrical and electronics industries. The continuous advancements have led to the use of composite materials in more diversified applications. The importance of composites as engineering materials is reflected by the fact that out of over 1600 engineering materials available in the market today more than 200 are composites [10]. Electrical packaging involves interconnecting, powering, protecting and cooling of semiconductor circuits for use in a variety of microelectronic applications. For microelectronic circuits; the main type of failure is thermal fatigue, owing to the different thermal expansion coefficients of semiconductor chips and packaging materials. Furthermore, because the power density increases rapidly, the ability to dissipate heat becomes a very important factor. Therefore, materials having reduced coefficient of thermal expansion (CTE) in combination with a high thermal conductivity are required for heat sinks and heat spreaders [11 – 12].

A silicon carbide reinforced MMC has been investigated by some researchers [11,13]. It is known that silicon carbide is not stable in contact with copper at elevated temperatures but silicon itself is partially soluble in copper to form a copper–silicon solid solution while pure carbon remain at the Cu/SiC interface. The soluble silicon however reduces the thermal and electrical conductivities of Cu/SiC composites, hence diffusion barriers are needed to prevent the detrimental interfacial reaction between copper and SiC [14]. One of the ways of enhancing the properties of MMCs is to alloy the copper matrix with a strong carbide former such as aluminum, chromium etc [14]. Copper is one of the most important materials for thermal and electronic applications. It has higher electrical and thermal conductivities and lower co-efficient of thermal expansion (CTE) than aluminium. Unfortunately, the thermal expansion of copper is about four times higher than that of the semiconductor silicon. SiC combines a thermal conductivity of about 200-300 W/mK with a co-efficient of thermal expansion of 4.5×10^{-6} K^{-1}. Therefore the use of SiC particles as reinforcement in copper based composites is considered very attractive to meet the increasing demands for high performance MMCs for applications such as heat sink materials and packages.

Therefore the aims of this study are to synthesize and characterize the thermo-physical properties of 10% aluminum bronze matrix composite, reinforced with varying particle sizes (212, 425, 710 and 1200 μm) of SiC, suitable as a heat sink material in the electronic industry. The thermo-electrical properties will be discussed as a function of the reinforcement. The relationship between microstructures, particle sizes and volume fractions of the SiC reinforcement will also be studied.

Materials and methods

Pure commercial samples of copper and aluminum were obtained from Cable metal Nigeria limited and Nigeria aluminum extrusion (NIGALEX) Lagos, Nigeria respectively. Their chemical compositions as determined by the suppliers are shown in Tables 1 and 2.

Table 1: Percentage chemical composition of copper sample

Element	Cu	Pb	Zn	Si	Mn	Fe	Cr	Sb
Composition	98.800	0.490	0.073	0.032	0.013	0.450	0.043	0.012

Table 2: Percentage chemical composition of aluminum sample

Element	Al	Si	Fe	Cu	Mn	Mg	Cr	Zn	Ti	Bal.
Composition	99.04	0.25	0.40	0.05	0.05	0.05	0.05	0.05	0.03	0.03

The SiC used for this investigation was purchased from a local commercial supplier in Lagos, Nigeria. This SiC was milled to obtain different particles sizes of 212, 425, 710 and 1200 μm. The matrix of the composite was a copper–aluminum (called aluminum bronze) alloy with composition 90% Cu and 10% Al. The SiC was added to the matrix in varying percentages by volume of the mould cavity (i.e. 5, 10, 20, 30, 40 and 50%).

The composite samples were synthesized by liquid metallurgy method in a 1 kg oil-fired crucible furnace. The furnace charges were prepared as shown on Table 3. The copper samples were properly cleaned alcohol and dried prior to melting to eliminate any surface impurities. The measured quantities of Cu and Al were charged into the furnace and heated to melting temperature. The SiC particulates were added to the molten metals at melting point and the mixture was continuously stirred before casting. The melt was cast in a prepared CO_2 sand mould, allowed to cool and solidified in the mould. The as-cast samples were heated and homogenized at 510^{o}C for 1 hour before they were cooled in still air. The normalized samples were characterized to determine their electrical conductivity, resistivity, tensile strength, hardness and metallographic structures.

The electrical resistivity of the samples was conducted on a DV power micro-ohmmeter RM0600 at 10 and 20 A using an average of three values. The thermal conductivity of the samples was conducted using thermal transport sample station (TTSS) P670. The specimens for metallographic examination were prepared using standard techniques [15]. The etchant used was a mixture of 10 g of ferric chloride in 100 ml of water and 30 ml of dilute hydrochloric acid. The microstructures of the as-cast and the heat treated samples were examined using a digital metallurgical microscope at 20X magnification.

Table 3: Mass of materials weighed for melting

Particle Size	212µm			425µm			710µm			1200µm					
	SiC	Cu	Al	SiC	Cu	Al	SiC	Cu	Al	SiC	Cu	Al	SiC	Cu	Al
5%-95%	6.39g	303.5g	10.2g	6.39g	303.5g	10.2g	6.39g	303.5g	10.2g	6.39g	303.5g	10.2g	25.56g	1214g	40.8g
10%-90%	12.7g	287.5g	9.65g	12.7g	287.5g	9.65g	12.7g	287.5g	9.65g	12.7g	287.5g	9.65g	50.8g	1150g	38.6g
20%-80%	25.6g	255.5g	8.58g	25.6g	255.5g	8.58g	25.6g	255.5g	8.58g	25.6g	255.5g	8.58g	102.4g	1022g	34.2g
30%-70%	38.4g	223.6g	7.5g	38.4g	223.6g	7.5g	38.4g	223.6g	7.5g	38.4g	223.6g	7.5g	153.6g	894.4g	30g
40%-60%	51.1g	191.7g	6.43g	51.1g	191.7g	6.43g	51.1g	191.7g	6.43g	51.1g	191.7g	6.43g	204.4g	766.8g	25.7g
50%-50%	63.9g	159.7g	5.36g	63.9g	159.7g	5.36g	63.9g	159.7g	5.36g	63.9g	159.7g	5.36g	255.6g	638.8g	21.2g
											TOTAL		792.3g	5686g	190.5g

Results and discussion

Copper and aluminium are widely used as industrial and functional metals for various thermal and electrical applications. This is due to their good thermal and electrical conductivities, high plasticity and excellent resistance to corrosion and oxidation. They however, possess low mechanical strength and wear resistance which limits their structural applications [16–17]. Synthesis of copper-aluminium reinforced metal matrix composite by liquid metallurgy is one method that can be used to enhance the deficient properties of these metals to make them suitable for structural applications. In this study, copper-aluminium reinforced MMC samples were prepared by liquid metallurgy method using micron–sized SiC particles and they were characterized to determine their thermal and electrical properties.

Thermal conductivity of the synthesized MMC samples

Table 4 shows the effect of the particle size of the SiC on thermal conductivity of the synthesized MMC samples. It can be seen from the table that thermal conductivity is greatly influenced by the particle sizes of the SiC. The thermal conductivity generally increases with decrease in measured particle sizes of the SiC. For instance at SiC particle size of 212 μm, the thermal conductivity is 7691.11 W/mk for volume fraction of 50 SiC, while the value is 9428.27 W/mk for SiC particle size 1200 μm at the same volume fraction of the particulate. This effect is probably due to the fact that at lower particle sizes of the SiC, there was thorough distribution of these particles within the matrix of the metals. This might have enhanced the perfect mixing of the two metals which are very good conductors of heat and enhanced by the SiC particles which equally have high thermal conductivity.

Table 4: Thermal conductivity of copper-aluminum/SiC composite

Particle size	212 μm	425 μm	710 μm	1200 μm
Volume fraction of SiC (%)	W/mk	W/mk	W/mk	W/mk
5	466.51	1289.41	1068.98	964.96
10	1234.20	810.57	2994.21	3097.64
20	6698.93	3727.16	1770.95	3287.31
30	703.55	1264.15	5702.43	2134.26
40	1440.67	4863.28	3451.96	13676.48
50	7691.11	3379.66	2737.51	9428.27

Electrical properties of the MMC sample

Electrical conductivity is the ability of a material to conduct electric current, as measured by the current per unit of applied voltage. The good conductivity of a material is due to the high mobility of the outer shell, or valence electrons throughout the crystal lattice. Conversely, electrical resistivity is an intrinsic property of a material that depicts its ability to resist the flow of current through it. It is defined as the ratio of the magnitudes of electric field and current density, in Ωm (ohm-metres);

$$\rho = \frac{E}{J}$$ (i) where; ρ is the resistivity in ohm-metre, E is the electric field strength V/m, and J is the current density in A/m^2. The greater the resistivity, the greater the field required to cause a given current density. The reciprocal of resistivity determines the electrical conductivity $(\Omega.m)^{-1}$ of the material. A perfect conductor would have zero resistivity, and a perfect insulator would have an infinite resistivity. The value of resistivity of a material can be evaluated from the expression;

Resistivity, $\rho = \frac{RA}{L}$ (ii) where; R is the resistance of the material in Ohms, A is the cross section area in square metre, and L is the length of the material in metre.

Table 5 shows the electrical conductivity of the MMS samples at different volume fractions of SiC particles.

Table 5: Electrical conductivity of the copper-aluminium/SiC matrix composite

Particle size	212 μm	425 μm	710 μm	1200 μm
Volume fraction of SiC (%)	Siemens/metre ($x10^7$)	Siemens/metre ($x10^7$)	Siemens/metre ($x10^7$)	Siemens/metre ($x10^7$)
5	11.51	12.65	11.88	9.75
10	9.56	11.81	8.34	13.56
20	3.89	5.38	6.99	4.73
30	7.19	4.55	2.60	8.75
40	4.14	2.32	3.37	1.32
50	1.86	3.55	3.88	1.43

The data present in this Table show that the electrical conductivity of these samples is greatly affected by the volume fraction of the SiC particles. The response of these samples to electrical conductor is similar to that of their behaviour to thermal conductivity. The electrical property of all the samples appear to decrease with increase in volume fraction of the silicon carbide particles, except for the 1200 μm sample which displays no definite behaviour in response to the presence of SiC particles.

A close look at the Table also reveals that the last three rows of all the samples with 30, 40 and 50% volume fraction of SiC are inconsistent in their response to the particles and these inconsistent values are typically low. It can therefore be inferred that at higher percentages of volume fraction of SiC particles, there appear to be a low and unreliable behaviour of the conductivity of these samples. This can be as a result of the fact that SiC particles are poor conductor of electricity, and the more contents that are present in these samples, the more they reduce their electrical conductivity. This is similar to the behaviour of the samples in response to thermal conductivity as previously explained.

From the behaviour of the samples of both thermal and electrical conductivities, there is an expected correlation between them since the free electrons that carry charge in electric conduction in metals also provide the principal mechanism for heat conduction. The ratio of the thermal conductivity to the electrical conductivity of a metal is proportional to the temperature. Qualitatively, this relationship is based upon the fact that the heat and electrical transport both involve the free electrons in the metal. The thermal conductivity increases with the average particle velocity since that increases the forward transport of energy. However, the electrical conductivity decreases with increase in particle velocity because the collisions divert the electrons from forward transport of charge. This means that the ratio of thermal to electrical conductivity depends on the average velocity squared, which is proportional to the kinetic temperature. The ratio of thermal to electrical conductivity multiply by the temperature forms a constant in Wiedemann-Franz law shown in the equation below. This law is also independent of the particle mass and the number density of the particles [18].

Microstructural analysis of the MMC samples

Fig. 1 shows a typical low magnification of the microstructure of a Cu–Al metal matrix composite sample at various particle sizes of silicon carbide. It can be observed from the Figure that the SiC particulates are evenly distributed within the matrices of the metals. The microstructures of the MMC samples as observed in the Figure appear to contain primary phase and the two metallic phases. It can therefore be inferred that the SiC particulates acted as nucleation sites for copper and aluminium phases during solidification. The two metallic phases were formed on the SiC particles. The lower the grain sizes of these particles the more the distribution within the matrices of these metals as evident in the micrographs (Figure 1 a – d). Segregation of other metallic particles is virtually not visible in these images due to low contents of other metals in the composition of copper and aluminium (Tables 1 and 2).

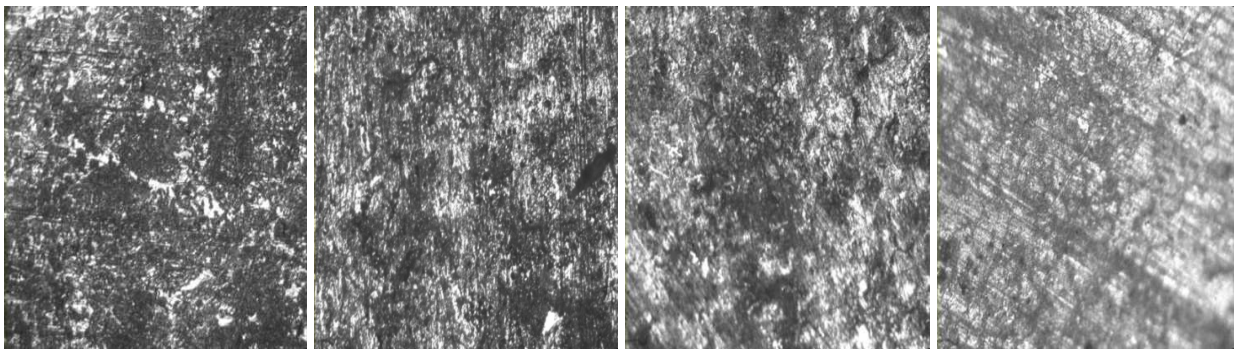

Fig. 1: The micrograph images of 50% Cu-Al composite samples at (a) 212 (b) 425 (c) 710 and (d) 1200 μm

It can also be observed that the no interfacial layers of constituent metals are visible in throughout the micrograph images at all the particulate sizes of the samples. When in contact with copper at elevated temperature, SiC particulates have the tendency to decompose to Si and C, while Si could also dissolve in Cu to lead to a decrease in the thermal conductivity of the matrix. Since the affinity of Cu for C and its solubility in the former is negligible, it is believed that the remaining carbon was responsible for the prevention of interfacial bonding which could also reduce the values of the CTE.

Conclusions

In this study, a low cost of aluminium-copper composite was synthesized using liquid metallurgy method. The synthesized MMC samples containing different micro-size silicon carbide particulates were characterized to determine their thermal and electrical properties. The analyses of the results obtained showed that thermal conductivity of the composite increased with decrease in particle sizes of the SiC. This property also has great influence on the volume fraction of the SiC, such that with a decrease in particle grain size, high thermal conductivity was achieved by increasing the volume fraction. The results obtained in this study showed that alloying copper matrix with aluminum is effective in reducing the interfacial particle matrix reactions of a typical MMC. The MMC samples also showed the high thermal conductivities with a low coefficient of thermal expansion which is synonymous to a low tensile strain at a maximum load applied.

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