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Synthesis and Characterisation of Silicon Carbide and Graphıte Partıculate Reınforced Hybrıd Copper Matrıx Composıtes

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Abstract

Effects of graphite (C) and silicon carbide particulates (SiC_p) on the morphology, electrical and mechanical properties of C10200 copper (Cu) alloy have been investigated. The Cu alloy was reinforced with blended C and SiC_p in varied wt. % by mould casting method to produce Cu-SiC_p/C composites. Mechanical, electrical and microstructural properties of the hybrid Cu-SiC_p/C composites were investigated. The developed composites exhibited acceptable electrical conductivity despite the addition of SiC/C particulates. The electrical conductivity of the Cu-SiC_p/C composites is within the bound limits required in literature. The hybrid composite with 30/50/20 % formulation of Cu/SiC_p/C exhibited the highest ultimate tensile strength of 90 N/mm^2 and 92 N/mm^2 for the 212 µm and 710 µm particle sizes respectively. The composite with 30/50/20 % formulation of Cu/SiC_p/C exhibited hardness values of 28.7 HB and 30 HB for the 212 µm and 710 µm particle sizes respectively. The addition of SiC_p/C appreciably increased the tensile strength and hardness of the composites.

Keywords: Graphite, Silicon Carbide, Copper Alloy, Mould Casting, Hybrid Composites.

1. INTRODUCTION

In this era of advanced technology, desirable engineering materials that will exhibit unique or exceptional properties such as corrosion resistance, electrical conductivity, thermal conductivity and stability, weight reduction and improved strength are required for a wide range of applications in aerospace, medical, power sector, automobiles and other fields. Engineering materials are used in various applications, which have made them to be valuable materials because of their desirable properties. Hence, their importance for optimum performance cannot be over emphasized.

Copper (Cu) and its alloys are among the groups of engineering materials used for various engineering applications. Among the desirable characteristics of Cu are low density, high ductility, good electrical and thermal conductivities, high corrosion resistance and it is non-magnetic in nature. Copper also exhibits high machinability and can easily be joined. Among its numerous areas of application are electrical wiring, heating system, cladding, roofing, water pipes and fittings, etc. However, pure copper is not suitable for some applications because of its low strength. Hence, improvement in the properties of copper has become imperative to make it suitable for applications in many fields.

In order to improve the properties of copper for suitability for wider applications, copper-based metal matrix composites (CuMMCs) were developed. Generally, metal matrix composites combine both metallic properties, such as toughness and ductility, and ceramic properties, such as elastic modulus and high compressive and shear strengths with stability at high temperatures. There has been considerable interest in the use of CuMMCs. Composite materials are composed of discrete reinforcement distributed in a continuous phase of matrix. In copper matrix composites, one constituent is copper which forms network i.e. matrix phase and other constituents serve as reinforcements, which are generally ceramics or non-metallic hard materials. Composite materials play vital roles in the present modern industrial sectors. They offer several applications in the aerospace, automotive and ship building industries as they have certain advantages over conventional metals.

Hybrid metal matrix composites (HMMCs) are produced by incorporating two or more reinforcement phases into a single metal matrix phase [1]. Often, HMMCs possess unique properties that may be difficult to find in a single reinforcement. Studies have

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been carried out on how to enhance the mechanical and structural properties of HMMCs with various types of matrix phases. Copper based metal matrix composites were synthesized using different reinforcements such as E-glass fibre and silicon carbide (SiC) particles by powder metallurgy route. Scanning electron microscopy (SEM) and EDS were used for microstructural analysis. Microstructure showed a random distribution of reinforcements in the matrix and very good bonding of the reinforcement with the matrix. The hardness of the composite increased with the vol. % of E-glass fibre and SiC in the Cu matrix [2]. Production by stir casting method of hybrid copper matrix composites using silicon carbide (SiC) and aluminium oxide (Al_2O_3) as reinforcements was also carried out. Mechanical characterisation of the composites indicated that ultimate tensile strength and yield strength were enhanced. The Young's modulus and shear modulus improved with increasing reinforcement. Poisson's ratio and density of the composites reduced with increasing reinforcement percentage. There was tremendous improvement in hardness of the component. There was improved electrical and thermal conductivities because of presence of graphite. The composites also exhibited increased wear resistance and machinability [3].

Also, the physical, mechanical and wear resistance properties of copper matrix composites prepared by stir casting using steel machining chips as reinforcement were examined. The results showed that addition of steel machining chips in copper resulted in significantly low porosity levels in the composites. The mechanical properties (hardness and tensile strength) and wear resistance improved with the use of the steel machining chips as reinforcement [4]. Production by powder metallurgy of copper matrix composites using different weight percentages (wt. %) of alumina (Al_2O_3) as reinforcement was also carried out and different properties of the composites were evaluated. Hardness of the composites increased with increasing content of Al_2O_3. There was a slight decrease in the compressive strength with increase in Al_2O_3. There was a decrease in the density of the composites with increase in alumina wt. %. The porosity of the composites increased with increase in reinforcement. Microstructure revealed voids with increase in wt. % of alumina [5].

In this study, Silicon carbide and graphite were used as reinforcements. Silicon carbide is an excellent abrasive used as a material in grinding wheels and other abrasive products. In air, SiC can be used up to 1600 °C and it forms a protective silicon oxide coating at 1200 °C. SiC has high thermal conductivity coupled with low thermal expansion, which provides thermal shock resistance. Silicon carbide with little or no grain boundary impurities maintain their strength to very high temperatures, approaching 1600 °C. Graphite is a crystalline form of carbon and it improves the machinability of composites and exhibits excellent thermal and electrical conductivity, thereby improving the conducting capability of copper composites. Hence, the objective of this study is to examine the effects of addition of silicon carbide and graphite particulates to copper alloy with respect to the morphology, electrical and mechanical properties of the composites for wider engineering application.

2. MATERIALS AND METHODS

2.1. Materials

The materials used are commercial copper wires C10200, silicon carbide (SiC) lump and 150 µm-graphite (C). The copper wires were obtained from Aalco Metal, Lagos, Nigeria and the chemical composition is presented (Table 1). Both SiC and C were obtained from Cermatics Nigeria Ltd. Copper wires were used as the matrix while SiC and C were used as reinforcements.

2.2. Materials Preparation and Production of Composite Specimens

The copper wires were washed with detergent, rinsed in distilled water and were allowed to dry. This was done to remove any unwanted surface impurity. Thereafter, they were ground using a pulverizer and sieved to 50 µm using British standardised sieves (BSS). The as-received SiC lump was ground and sieved to obtain two-grain sizes of 212 µm and 710 µm while the 150 µm-graphite was used as received. Measured quantity of copper alloy matrix was placed in a crucible pot, charged into a L02-MAN crucible furnace (Figure 1) and heated to temperature of 1100 ^{0}C until molten form was obtained. Prior to pouring of the molten copper, the reinforcements were pre-heated at 200 ^{0}C for 30 minutes to enhance the wettability of the composite [6]. Thereafter, the reinforcements were added to the molten copper alloy according to the mixing composition (Table 2). The mixture was thoroughly stirred using a stirrer for 10 mins to avoid clustering and to achieve good dispersion of the particles in the matrix. Thereafter, the slurry was steadily poured into cylindrical metallic moulds of diameter 1.5 cm and length 15 cm and allowed to cure

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for one hour after which they were removed from the moulds. In each of the mixtures (Table 2), the mass of the materials needed was calculated using Equation (1) and Table 3.

Table 1. Chemical composition of the as-received copper wire

Elements	Cu	Pb	Zn	Si	Mn	Fe	Cr	Sb
Weight (%)	98.88	0.49	0.073	0.032	0.013	0.45	0.050	0.012

Figure 1. Melting of copper alloy in the crucible furnace

Table 2. Weight % of the Cu-SiC_p/C composite constituents

S/N	Cu-SiC_p (%)		Cu-SiC_p-C (%) (varied graphite)			Cu-SiC_p-C (%) (varied SiC_p)		
	Cu	SiC_p	Cu	SiC_p	C	Cu	SiC_p	C
1	80	20	75	20	5	80	15	5
2	70	30	60	30	10	70	20	10
3	60	40	45	40	15	60	25	15
4	50	50	30	50	20	50	30	20

$$\mathbf{m = \rho \times v} \qquad (1)$$

where, m is the mass (g), ρ is the density (g/cm^3) and v is the volume (cm^3) of the material.

Table 3. Density of the materials used

Materials	Copper alloy	Silicon carbide	Graphite
Density (g/cm^3)	6.940	3.100	2.266

2.3. Microstructural Examination

Microstructural examination of the specimens was done using an optical microscope. The specimens for the optical microscopy were smoothened using a series of emery papers of grit sizes ranging from 500 – 1500 µm while fine polishing was done afterwards with diamond paste. They were etched using 10 % H_3PO_4 solution in 10 seconds [7] before examination in an optical microscope.

2.4. Mechanical Tests

Specimens for mechanical tests were machined to specifications using a lathe machine and were cut with a hacksaw. Tensile test was conducted on the specimens (Figure 2) using Instron electro-mechanical tester model 3369 (Figure 3) in accordance with the ASTM E-8 standard [8]. From the tensile tests carried out, ultimate tensile strength, true stress and true strain

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were determined. The hardness of the composites was determined using a Brinell hardness tester in accordance with the ASTM E10-01E1 standard [9].

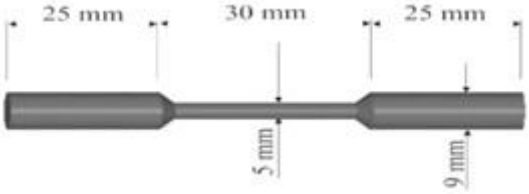

Figure 2. Cylindrical tensile strength test specimen.

Figure 3. Instron electro-mechanical tester.

2.5. Electrical Conductivity and Resistivity Tests

Resistivity test was conducted on specimens of gauge length 15 cm and diameter 1.5 cm. Resistivity test was conducted using the DV power micro-ohmmeter RM0600 machine (Figure 4) at 10A and 20A, using Equation (2) [10]. In each test, an average of three different readings were taken. Electrical conductivity ($\boldsymbol{C}$) of the samples was obtained according to Equation (3) [10]. The experimental set-up for the conduction of the electrical resistivity experiment is illustrated (Figure 5).

$$\boldsymbol{\rho = R\frac{A}{L}} \qquad (2)$$

$\boldsymbol{\rho}$ = **resistivity** (Ω)
$\boldsymbol{R}$ = **resistance** (Ω)
$\boldsymbol{A}$ = **cross − sectional area of the sample** ($\boldsymbol{cm^2}$)
$\boldsymbol{L}$ = **distance between 2 leads of voltmeter**($\boldsymbol{V}$)

$$\boldsymbol{C = \frac{1}{\rho}} \qquad (3)$$

Figure 4. DV power micro-ohmmeter RM0600 machine.

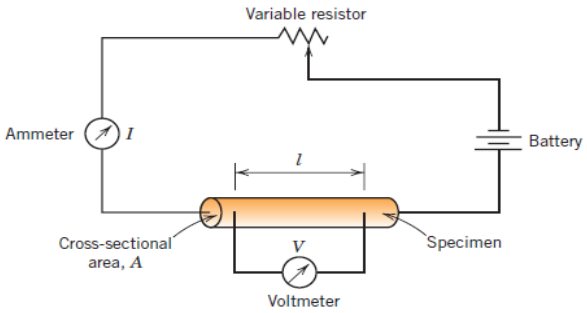

Figure 5. Experimental set-up for the electrical resistivity test.

2.6. Theoretical Analysis of the Electrical Properties of the Composites

Table 4 shows the theoretical electrical properties of the copper alloy and silicon carbide used for this study. The rule of mixtures for the upper and lower bounds are presented in Equations (4) and (5) [11, **12**] respectively. These two equations were combined with electrical properties (Table 4) and mixing fractions (Table 2) to obtain the theoretical bound limits for the composites.

$$\boldsymbol{C_{c(u)} = V_m C_m + V_p C_p} \qquad (4)$$

$$\boldsymbol{C_{c(l)} = \frac{C_m C_p}{V_m C_p + V_p C_m}} \qquad (5)$$

Where,
$\boldsymbol{C_{c(u,l)}}$ = electrical conductivity of the composite
U = upper bound
I = lower bound
C_m = electrical conductivity of the copper alloy
C_p = electrical conductivity of the silicon carbide particulate
V_m = volume of the copper alloy

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V_p = volume of the silicon carbide particulate

Table 4: Theoretical electrical properties of the materials [13]

Electrical properties	Copper alloy	Silicon carbide	Graphite
Conductivity (Ohm.m)$^{-1}$	6.00 x 10^7	1.63 x 10^7	2 x 10^5
Resistivity (Ohm.m)	1.67 x 10^{-8}	6.13 x 10^{-8}	5 x 10^{-6}

3. RESULTS AND DISCUSSION

3.1. Electrical Properties of the Composites

The responses of electrical conductivity of the copper alloy reinforced with only SiC_p is illustrated (Figure 6). The electrical conductivity of the copper alloy decreased as the SiC_p is increased. The same trend was observed in the two particle sizes investigated. As expected, the electrical conductivity of copper alloy decreased due to the addition of semiconductor SiC_p that has lower electrical conductivity or higher electrical resistivity compared to the copper alloy (Table 4). The electrical conductivity of the fabricated composites ($\mathbf{C_c}$) is within the bound limits (Figure 7). The lowest electrical conductivity of 3.0 Ωm obtained is close to that of aluminum alloy 3.8 Ωm [13] despite the addition of SiC_p. Figures 8 and 9 show the electrical conductivity of Cu-SiC_p/C composites at higher and lower copper content respectively. The electrical conductivity decreased with increasing graphite addition. The two figures also indicate that larger particle size of 710 µm exhibited better electrical conductivity than smaller particle size of 212 µm. However, at higher copper alloy content with addition of graphite, the conductivity of the composites improved significantly as illustrated (Figure 10).

The electrical conductivity and resistivity of the composites developed depend on the electron concentration, charge and mobility presence [14]. In this study, the electron mobility played the most significant roles since limited chemical reaction was expected between the reinforcement phases and matrix phase. When silicon carbide (SiC_p) was introduced to the melt pool, the vacant sites in the copper alloy was occupied. The presence of SiC_p opposed the free mobile copper electrons by reducing their drift velocity. The free electrons also experienced scattering phenomenon that reduced the flow of electric current and thus causing reduction in electrical conductivity (Figure 6 to 9). The improvement in the electrical conductivity (Figure 10) was due to higher electrical conductivity of the graphite compared to SiC_p (Table 4).

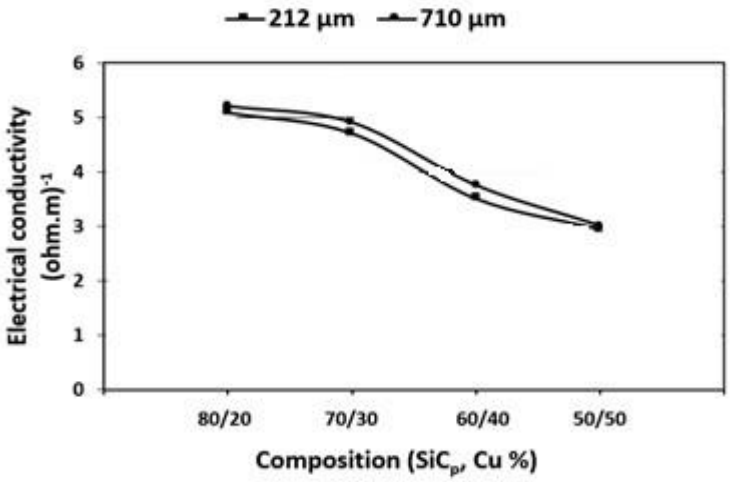

Figure 6. Response of electrical conductivity to Cu alloy reinforced with SiC_p of 212 µm and 710 µm.

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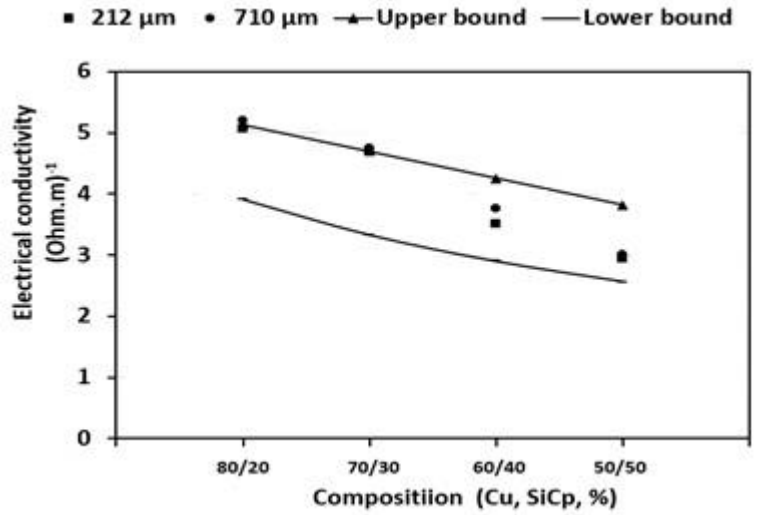

Figure 7. Comparison of electrical conductivity of fabricated and theoretical composites.

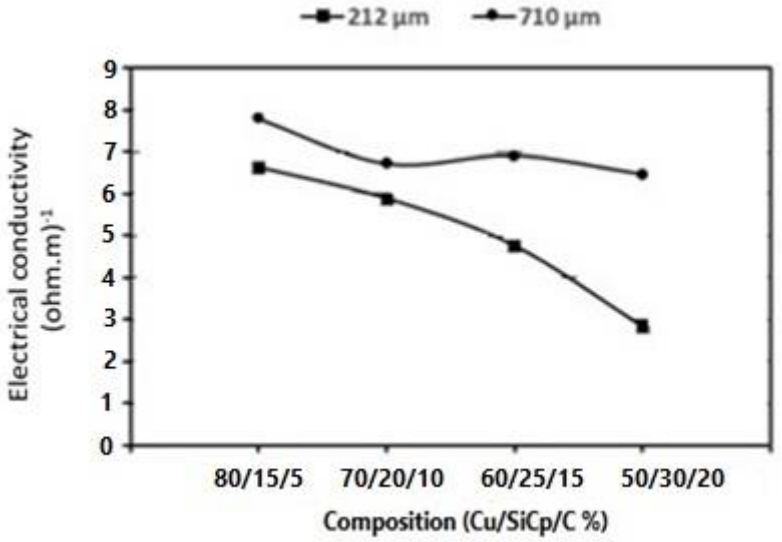

Figure 8. Electrical conductivity response of Cu alloy reinforced with SiC_p and graphite powder at higher Cu alloy content.

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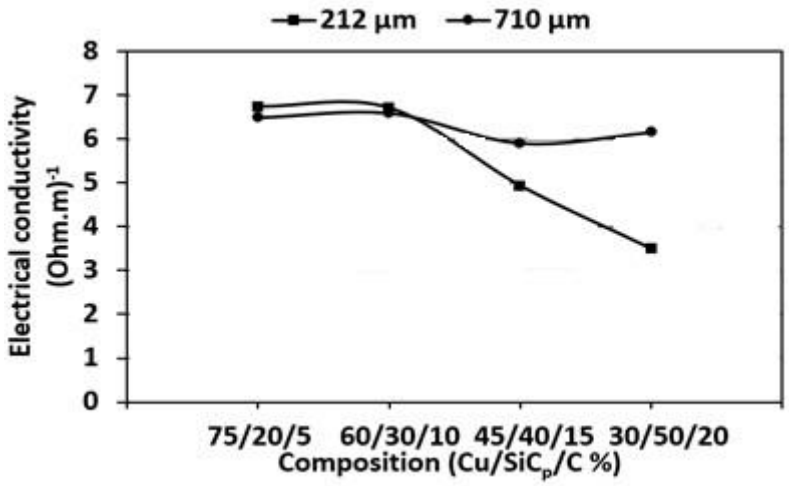

Figure 9. Electrical conductivity response of Cu alloy reinforced with SiC_p and graphite powder at lower Cu. alloy content

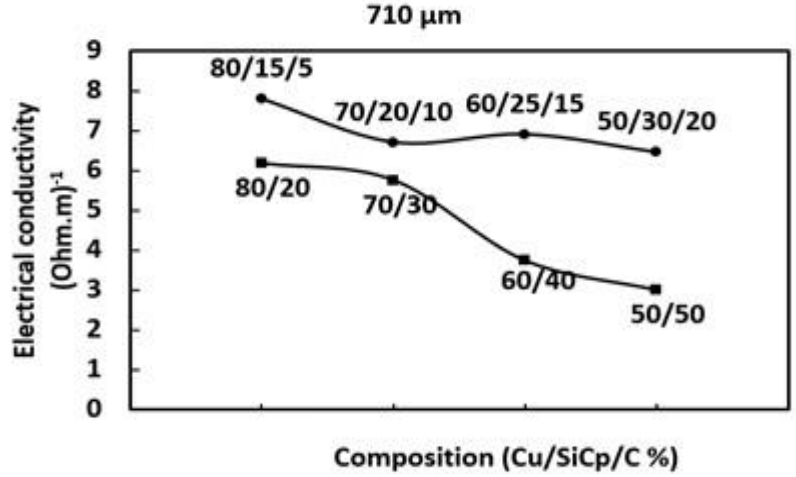

Figure 10a. Electrical conductivity response to SiC_p and C addition at constant Cu alloy for 710 µm.

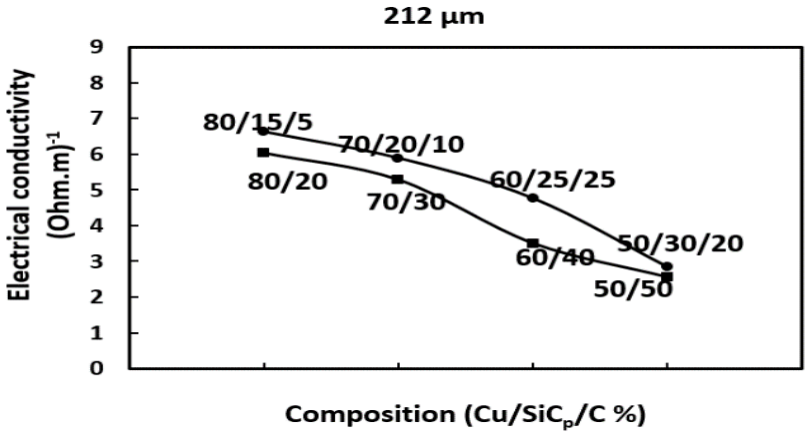

Figure 10b. Electrical conductivity response to SiC_p and C addition at constant Cu alloy for 212 µm.

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3.2. Mechanical Properties of the Composites

The stress-strain graphs of the composites developed with particle sizes of 212 and 710 µm respectively are illustrated (Figures 11 and 12). In Figure 11, there is no significant difference in the true stress of the samples at a true strain less than 0.01. However, at true strain above 0.01, the true stress differed significantly. Composites fabricated with mixing composition of 80/15/5 % (Cu/SiC_p/C) exhibited the highest true strain of 0.027 and lowest ultimate tensile strength of 50 MPa, which indicates its tendency of better ductility compared to other composites fabricated. This is due to the high percentage of copper alloy that is present in the composition. Increase in SiC_p and graphite contents enhanced the ultimate tensile strength significantly as illustrated (Figure 11). The peak ultimate tensile strength values of 80 and 90 MPa were obtained for 50/50 % Cu/SiC_p and 70/20/10 % Cu/SiC_p/C at a strain of 0.0211 and 0.0193 respectively. Similar trend was observed when the composites were produced with SiC_p of 212 µm as illustrated (Figure 12).

Addition of SiC_p and graphite a played significant role in enhancing the overall mechanical properties of the composites. The strength of the matrix phase (copper alloy) was enhanced by the addition of SiC_p/C based on principle of solid solution strengthening (Figures 11 and 12). This is in line with the mechanisms by which dislocations cause change in mechanical properties, which are strengthening by grain size reduction, solid solution alloying and strain hardening [15, 16]. The presence of solute SiC_p/C in the copper alloy generated dislocation across the length of composites. The parameters that control motion of dislocations are dislocation interaction and direct dislocation particulate interaction with the matrix structure. The dislocation interaction at the grain boundaries caused heavy dislocation pile-up, in other words, high dislocation density in which grain boundaries serve as barriers to dislocation motion. The particle-matrix interface also contributed significantly to the high dislocation density.

Increase in tensile strength seen in Figures 11 and 12 was due to the SiC_p/C addition that acted as barriers to the dislocation motion. Activities of the dislocations at the grain boundary increased the dislocation density, which positively contributed to the strengthening of Cu/SiC_p/C composites. Increase in the dislocation density will lead to an increase in yield strength. The material becomes harder. Ductility and tensile strength also increases [15, 16]. Another important factor that may have contributed to the increase in tensile strength is the inter-particulate distance between the reinforcement phases. Two reinforcement phases were used: SiC_p and C. The inter-particulate distances between the mixtures of the composites were reduced by the volume fractions and particle sizes of the reinforcement phases. The addition of SiC_p and C reduced the inter-particulate distance, which enhanced resistance of the composites to deformation thereby making them stronger and harder. The composite with 30/50/20 % formulation of Cu/SiC_p/C exhibited the highest ultimate tensile strength of 90 N/mm^2 and 92 N/mm^2 for the 212 µm and 710 µm particle sizes respectively. Other ultimate tensile strengths measured are presented (Figure 13).

Considering the hardness of the Cu-SiC_p/C composites (Figure 14), hardness increased with increasing SiC_p/C. In the hybrid composites, the hard SiC_p and graphite acted as load bearing materials, which enhanced the mechanical property of the composites. The composite with 30/50/20 % formulation of Cu/SiC_p/C exhibited hardness values of 28.7 HB and 30 HB for the 212 µm and 710 µm particle sizes respectively. The presence of hard silicon and graphite along the flow lines of the composites acted as barriers to the movement of dislocations within the copper alloy. It increased the volume fraction of hard particles, which increased the hardness of the composites. The strengthening mechanism of the hardness also depends on whether the dislocations are able to cut through the precipitation particles (Figure 15a) or form loops around the precipitation particles (Figure 15b). Actually, both strengthening mechanisms were largely responsible for increasing the strength and hardness of the composites. When a particle is introduced in a matrix, an additional barrier to the movement of dislocation is created and the dislocation will react by either cutting through the precipitating particles or by forming a loop around the obstacles which agrees with the postulation by [17].

Small particle sizes experienced dislocation movement by cutting while large particles experienced dislocation movement by looping. This is because finer particles indicate smaller inter-particle spacing while coarse particles growing at the expense of smaller ones indicate larger inter-particle spacing in the same volume. Small particles were coherent and deformable with dislocations cutting through them. In this case, strength and hardness weakly depended on

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the particles size. For large particles, dislocation bypass or looping over particles and strength or hardness were strongly dependent on particles size. Particles bypass resulted in high density of dislocation loops and thus leading to higher work hardening rate and hardness. This could be the reason for the higher hardness of the 710 µm than the 212 µm.

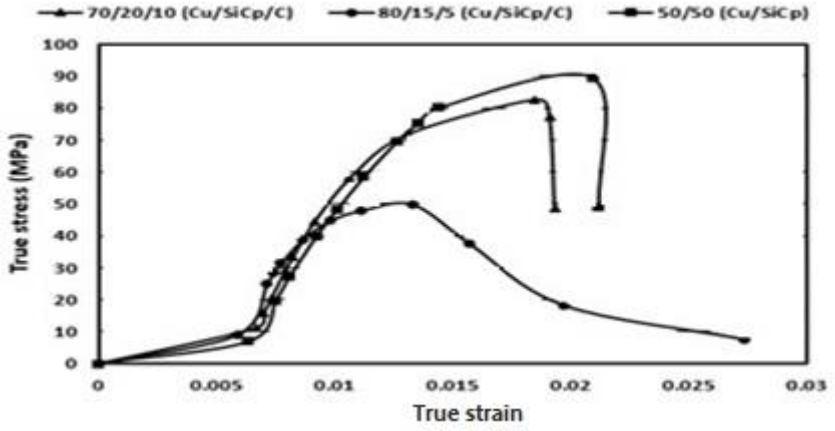

Figure 11. Variation of true sress with true strain of Cu alloy reinforced with SiC_p and C for 710 µm.

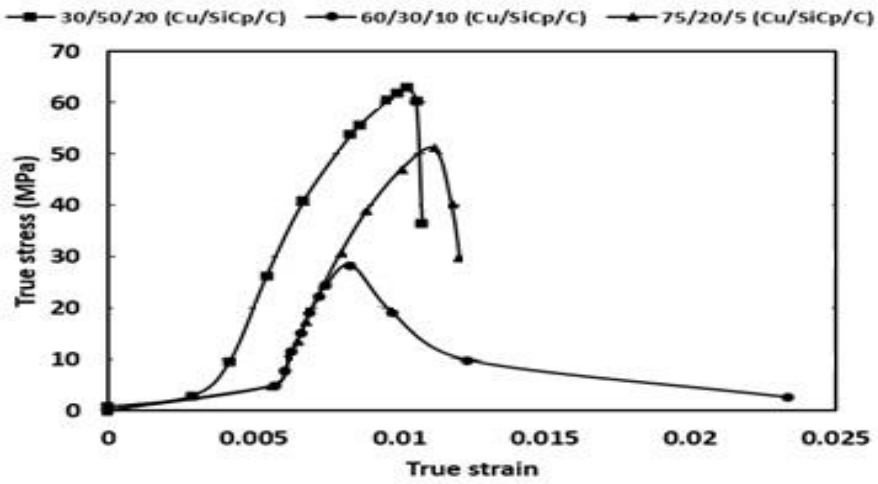

Figure 12. Variation of true sress with true strain of Cu matrix reinforced with SiC_p and C for 212 µm.

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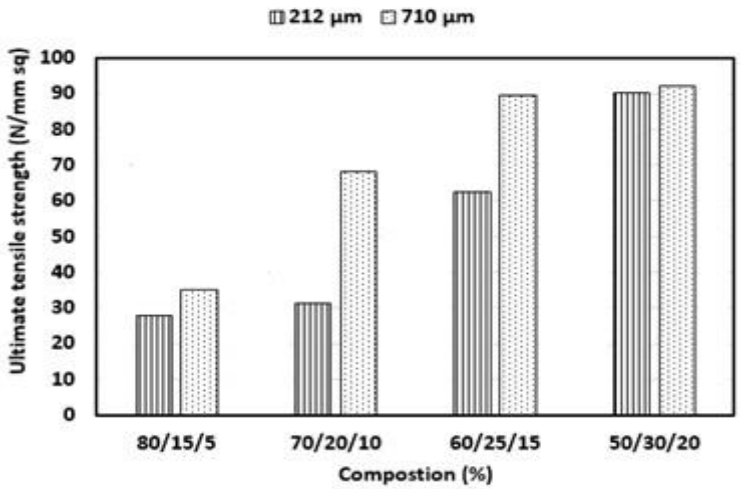

Figure 13. Ultimate tensile strength as a function Cu-SiC_p/C composite composition.

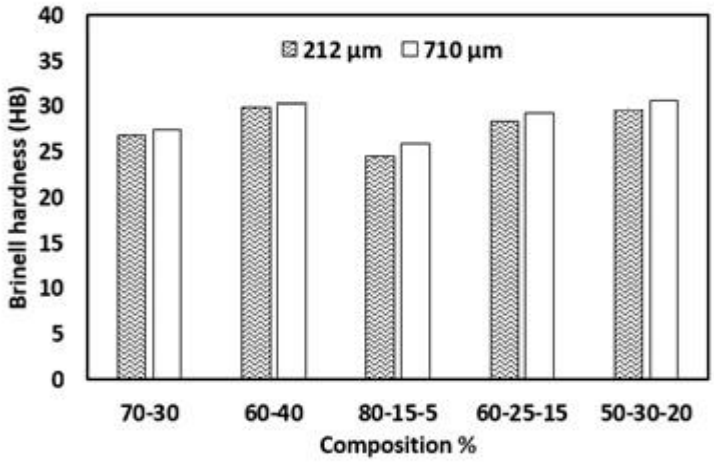

Figure 14. Brinell hardness as a function Cu-SiCp/C composite composition.

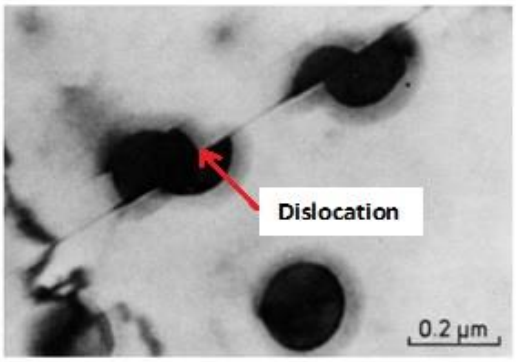

Figure 15a. Dislocations passing through the particles [17].

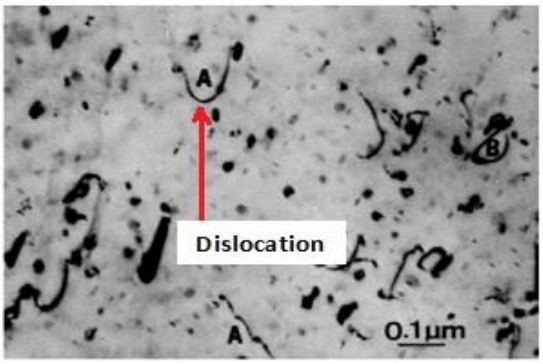

Figure 15b. Dislocations looping over the particles [17].

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3.3. Morphology of the Composites

The micrographs of Cu-SiC_p composites are illustrated (Figure 16). The dark and white phases are the reinforcement and matrix phases respectively. At 20 % addition of SiC_p (Figure 16a), the volume fraction of the white phase was significantly high with large grain sizes. This is an indication of high presence of copper. The volume fraction of the dark phases increased with increasing SiC_p (Figures 16b, 16c, and 16d). Figures 17a – 17d show the micrographs of the specimens containing silicon carbide and graphite particulates. The volume fraction of the dark phase further increased with addition of SiC_p and reduction in white Cu phase (Figures 17b, 17c, and 17d) as grain size reduced. Generally, there was a uniform distribution of SiC_p within the copper matrix. However, the introduction of low-density graphite particles caused clustering in some portions of the copper matrix phase as illustrated in Figure 17a to 17d. This is because graphite has a lower density compared to the copper alloy and SiC_p. This caused the graphite to float in the melt resulting in non-uniform distribution.

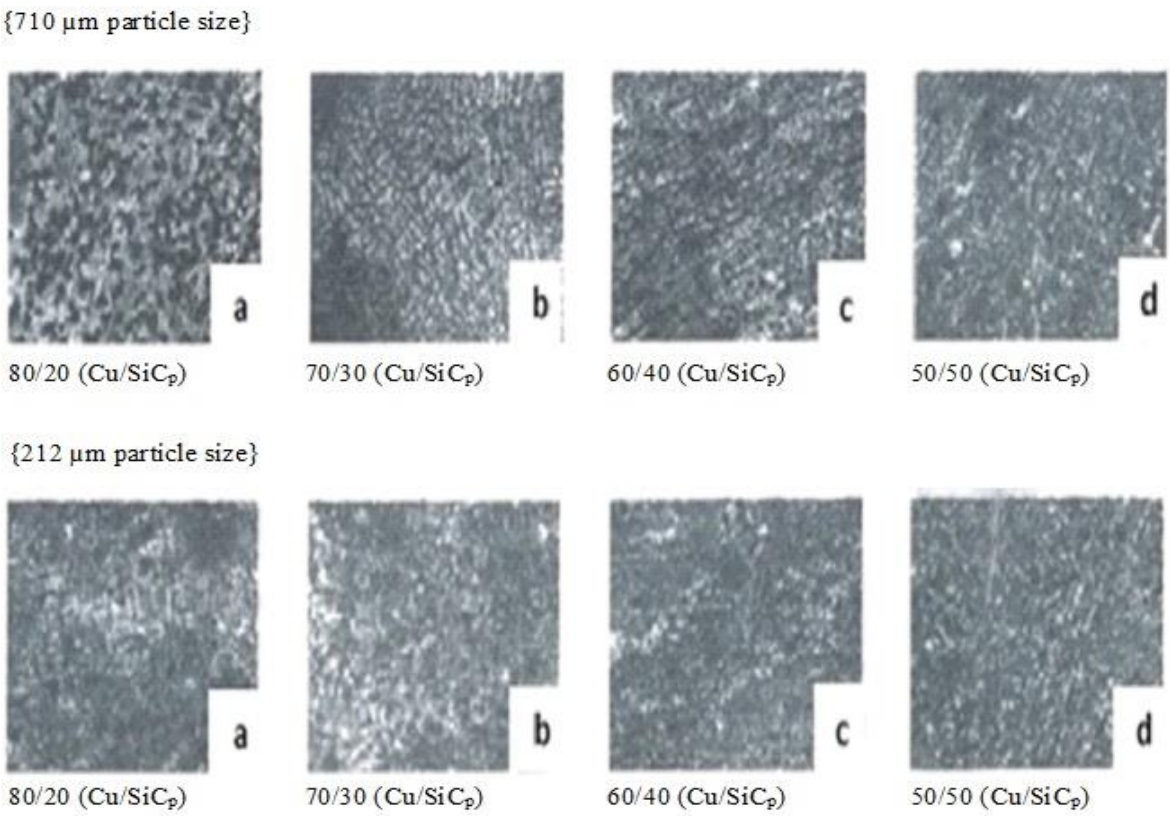

Figure 16. Optical micrographs of specimens produced with addition of silicon carbide particulate only x200.

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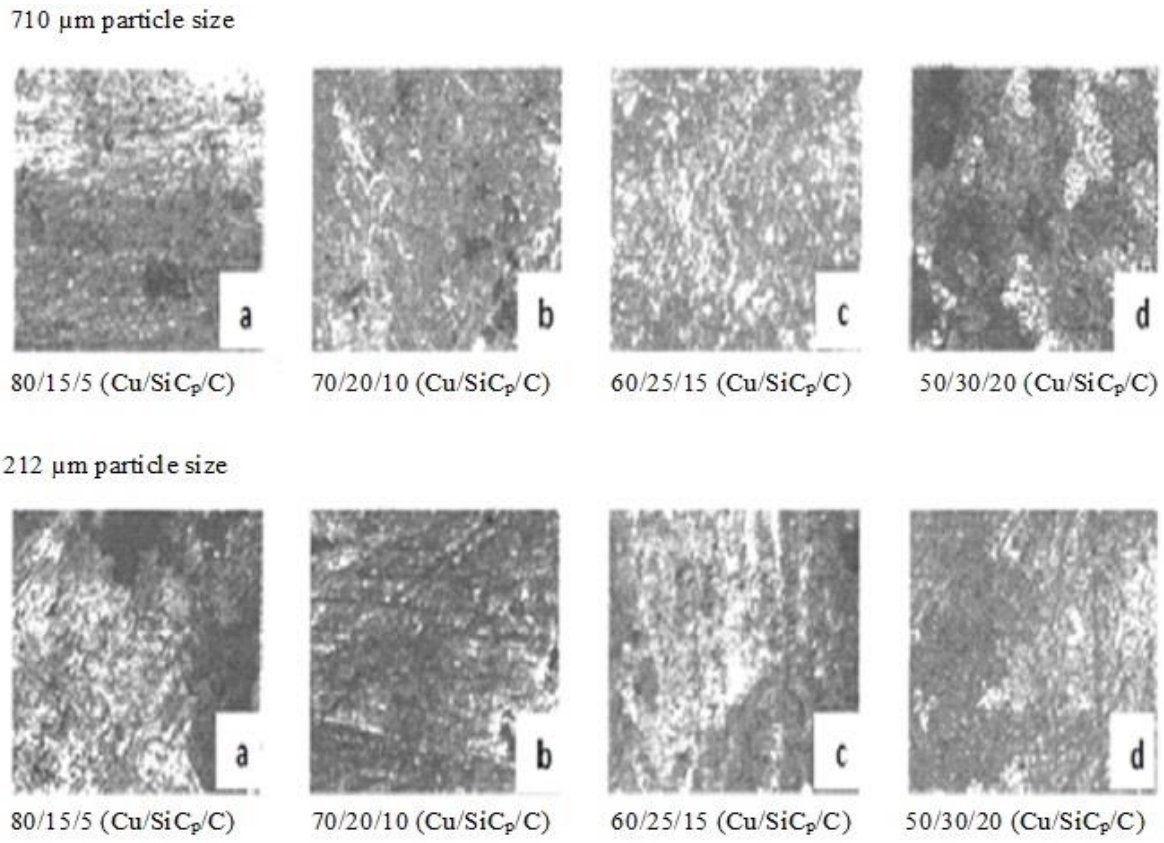

Figure 17. Optical micrographs of samples produced with addition of silicon carbide particulate and graphite x200.

4. CONCLUSION

The effects of graphite and silicon carbide particulates on copper alloy have been investigated. There was uniform dispersion of SiC_p within the copper matrix but the introduction of low-density graphite particles caused clustering in some portions of the copper matrix phase. The developed composites exhibited acceptable electrical conductivity despite the addition of SiC/graphite particulates. The electrical conductivity of the Cu-SiC_p/C composites is within the bound limits required in literature. The composite with 30/50/20 % formulation of Cu/SiC_p/C exhibited the highest ultimate tensile strength of 90 N/mm^2 and 92 N/mm^2 for the 212 µm and 710 µm particle sizes respectively. The composite with 30/50/20 % formulation of Cu/SiC_p/C exhibited hardness values of 28.7 HB and 30 HB for the 212 µm and 710 µm particle sizes respectively. Overall results confirmed that addition of SiC_p/C appreciably increased the tensile strength and hardness of the Cu-SiC_p/C composites. Silicon carbide and graphite are effective reinforcements in the development of hybrid copper metal matrix composites for wider engineering application.

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