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**MECHANICAL AND THERMO-ELECTRIC
PROPERTIES OF *Cu/SiC/Gr* MULTI-LAYER HYBRID
METAL MATRIX COMPOSITE**

Combined effect of both graphite and SiC content into Cu matrix has been studied during fabrication of Cu-graphite-SiC multilayer and single-layer composites by powder metallurgy. Mechanical properties of the composites are enhanced by simultaneous addition of equal weight fraction of SiC and Gr particulates of 3 %, 6 %, 9 %, and 12 % reinforcement into pure Cu, whereas electrical conductivity deteriorates. Multi-layer composites enhanced the wear resistance and improved the friction performance. The CTE could be decreased effectively through well bonding between reinforcement phase and metal matrix. Electrical conductivity decreases with increase in both graphite and SiC content. This single and multilayer hybrid metal composites will have promising future in electrical contacts, thermal and electronic packaging, friction and brakes materials applications.

Key words: *Cu-Gr/SiC*, HMMCs, multi-layer composites.

Introduction

Since the discovery of thermoelectric materials, great efforts have been put into the improvement of their efficiency and the development of devices for real life applications. This progress has been accelerated in the last few decades by the increasing demand for renewable energy sources. The discovery and design of compounds with intrinsically low thermal conductivity, especially compounds with a special bonding nature and stable crystal structure, is a new direction to broaden the scope of potential thermoelectric (TE) materials [1].

Copper is used extensively in thermoelectric instrument making as a material for connection of thermoelements. Therefore, research on the properties of *Cu* based materials, in particular, copper nanocomposites, is vital for thermoelectricity.

Copper is a widely used industrial and functional metal for various thermal and electronic applications, i.e. electronic packaging, electrical contacts and resistance welding electrodes. The scope of the devices could be significantly expanded from temperature sensors to mass produced, flexible thermoelectric generators [2]. This is because of good thermal and electrical conductivity, high plasticity and excellent resistance to corrosion and oxidation. Nevertheless, low mechanical strength and undesirable wear resistance limit its applications [3 – 7].

Suitable thermal and mechanical properties can be achieved by blending appropriate metallic and ceramic phases to form a composite. The incorporation of *SiC* hard reinforcing particles into the *CuMCs* improving their mechanical and tribological behavior, *Cu/SiC* metal matrix composites, because of their excellent electrical and thermal conductivity, enhanced hardness values, wear and frictional properties, has been the subject of extensive research [8 – 10].

Characteristics of *CuMCs* reinforced with soft reinforcement particles of *Gr*, being a solid lubricant and possessing good conductivity and anti-seizure properties are covered in [11 – 12].

Use of single reinforcement in copper may sometimes lead to the deterioration in the values of its physical properties. To overcome this, the concept of use of two different types of reinforcement is being tried out in copper matrix. To offset these effects, graphite being a solid lubricant and possessing good conductivity can be dispersed in copper along with *SiC* [13 – 15].

However, insufficient information is available about the processing and characterization of these novel laminated hybrid copper composites. The present study has been carried out to investigate combined effect of graphite and *SiC* into *Cu* matrix during fabrication of *Cu*-graphite-*SiC* multilayer hybrid metal matrix composite by layer compaction and pressure sintering at varying equal weight fraction of *SiC* and *Gr* particulates of 3 %, 6 %, 9 %, and 12 % reinforcement. This experimental analysis and test results on the thermo-physical properties of multilayer and single layer composites *Cu/SiC/Gr*-HMMCs will provide essential guidelines to the manufacturers.

Experimental procedure

Materials and preparation of composites

For the preparation of the composites, the raw material powders used were commercially pure powders of copper, tin, *SiC*, barium sulphate, graphite and zinc stearate. Silicon carbide particles of 45 μm average size and graphite particles of 90 μm average size were used in this study.

In order to manufacture *Cu*-graphite-*SiC* composite, powders were blended in a shaker mixer for 30 min to ensure the uniform distribution and homogeneous mixing of copper, graphite and *SiC* powders. The powder mixtures were cold compacted by uniaxial die pressing by applying pressure of 500 MPa for 15 min and then sintered in a tubular furnace at 750 °C for 90 minutes in argon atmosphere, while keeping the same compaction and sintering parameter. For now, we have produced composites *D* and samples test №2.

This samples were subject to the manufacturer’s recommended cure cycle. The purpose of addition of tin is to facilitate the liquid phase sintering for better densification. At this stage, we have processed multilayer hybrid composites (*A*, *B* and *C*) for samples test №1: The specimens have the same structure, i.e., *Cu* matrix, physical dimensions, and differ only in *SiC/Gr* reinforcement and laminate lay up as per table.

Table

The content of reinforcing and matrix materials

Composite	Layers	Silicon Carbide (<i>SiC</i>), %	Graphite (<i>Gr</i>), %	Matrix
<i>A</i>	3	9	9	1 % <i>Sn</i> , 10 % <i>BaSO4</i> and 83 % <i>Cu</i>
<i>B</i>	3	6	6	1 % <i>Sn</i> , 10 % <i>BaSO4</i> and 77 % <i>Cu</i>
<i>C</i>	3	3	3	1 % <i>Sn</i> , 10 % <i>BaSO4</i> and 81 % <i>Cu</i>
<i>D</i>	1	9	9	1 % <i>Sn</i> , 10 % <i>BaSO4</i> and 81 % <i>Cu</i>

Testing Procedures

Samples Test №1: Single to multi-layered *Cu*-HMMCs

Composites were cast in required diameter and length. The silicon carbide particles exhibit the form of solid crystal, whereas the graphite particles appear to be flakes.

The different samples were precision weighed on electronic balance to an accuracy of 0.1 mg. In general, the experimental data obtained from the three specimens cut from the same composite were taken as the average value.

The wear and friction behavior of the composites was studied using a computer controlled pad-on-disk laboratory scale inertial brake dynamometer in air at ambient temperature (25 – 30 °C).

The compression tests were carried out in a universal testing machine at a strain rate of 0.001 m/s. The size of the specimen for compression and thermal expansion was 5 × 5 × 20 mm. The coefficient of thermal expansion of specimens was tested from 20 °C to 200 °C, with the heating rate of 5 °C/min, using Linesis 75 Platinum Horizontal Dilatometer.

Samplpes test №2: Single Layer, 10 % BaSO₄ and 78 % – 87 % Cu, varying SiC\Gr contents of 3 %, 6 %, 9 %, and 12 %:

The density measurements were carried out to determine the porosity levels of the samples. The density of the samples was measured according to ISO 2738 standard. The measured density was compared to the value obtained using rule-of-mixtures so as to determine the volume fraction of porosity.

Four probe electrical resistivity measuring instrument was used to measure the electrical resistance of the sample, in which two probes were connected with the ammeter and another two with the voltmeter.

Results and Discussion

Density and Porosity

Fig. 1 shows the variation of relative density for single-layer Cu-graphite-SiC hybrid composites containing 3 %, 6 %, 9 %, and 12 % SiC and 3 %, 6 %, 9 %, and 12 % graphite. The percentage porosity present in the composites rises with the increase of the reinforcement content, as shown later in Fig. 4. Graphite is a solid lubricant material and facilitates movement and rearrangement of matrix and reinforcement particles resulting in higher densification. It is also observed that the value of relative density increases with increase in SiC content. The density of composite depends on the volume fraction of soft and hard phase. At low fraction of SiC, hard particle is well dispersed and soft graphite and Cu particles can deform to fill the gaps and the density value of around 85 % is achieved depending on graphite content. The volume fraction of porosity, and its size and distribution in a cast metal matrix composite play an important role in controlling the material mechanical properties. It is necessary that porosity levels be kept to a minimum.

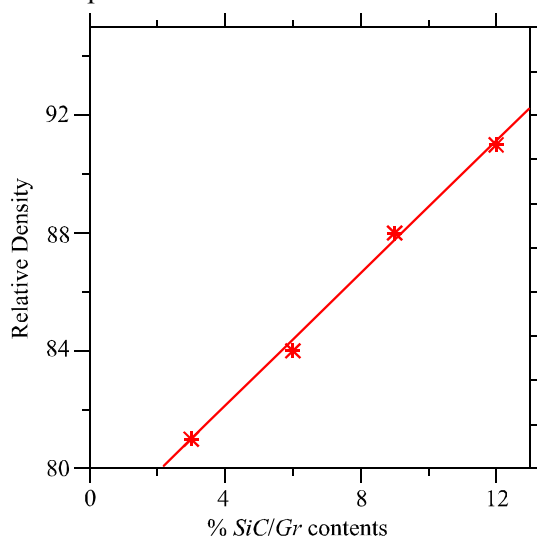


Fig. 1. The density variations with increasing SiC/Gr contents.

Wear Resistance

As shown in Fig. 2, with the increase in the SiC/Gr contents and layers, the wear rate of Cu/SiC/Gr HMMCs reduces. It was found that the amount of graphite released on the wear surface forms a tribofilm on the contact surfaces. This reduces the wear rate. The presence of graphite tribolayer (or mechanically mixed layer) also increases the seizure resistance and enables to run under boundary lubrication without galling. Increasing protrusion of SiC particles results in formation of a more stable lubricating film on the tribosurface of hybrid composites.

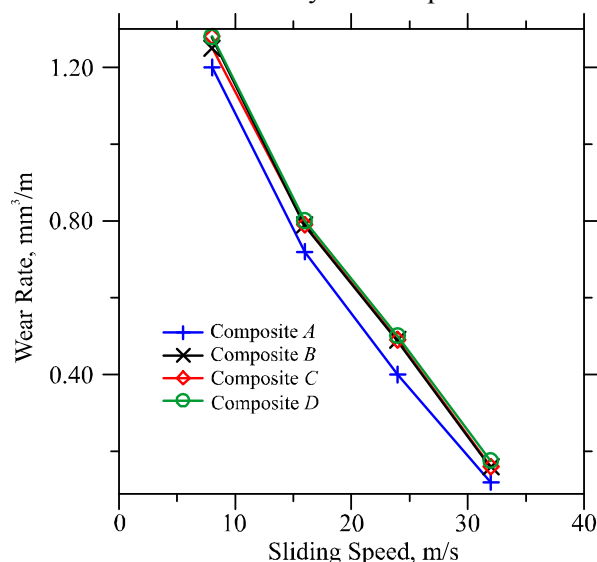


Fig. 2. The effect of SiC/Gr addition on the matrix wear rate of the investigated composites.

Braking Performance

As shown in Fig. 3, the amount of graphite released on the wear surface forms a tribofilm on the contact surfaces. This reduces the overall friction coefficient. Also, composites D show a very high friction coefficient of 0.565 at 4 m/s. This high friction does not translate into improving braking performance or wear resistance. The development of an oxide scale, greater participation of graphite in the sliding, weakening adhesive and abrasive frictional contacts diminish the frictional forces at the interface with growing sliding speeds.

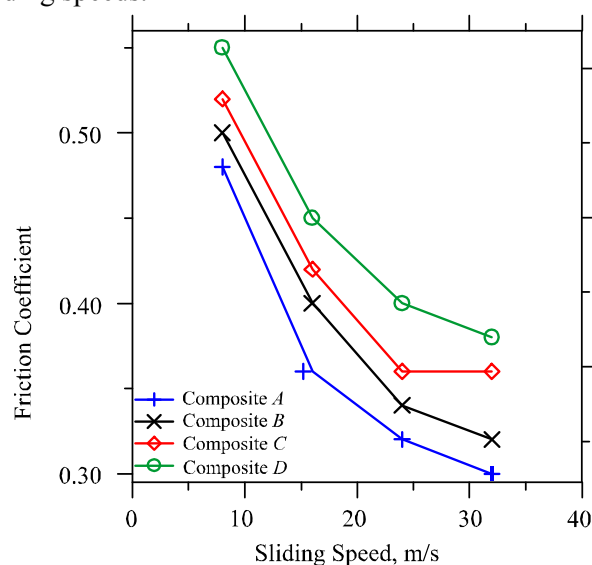


Fig. 3. Effect of sliding speed on mean friction coefficient of the composites.

Thermal Expansion and Porosity

It is evident from Fig. 4 that coefficient of thermal expansion (CTE) of a laminated composite is lower than that of a single-layer one. As SiC/Gr volume fraction increases, the CTE tends to decrease linearly with increase of porosity. The packaging materials in microelectronics should have high thermal conductivity to dissipate the heat, and low CTE to decrease the thermal expansion mismatch among the devices. It is evident from the plot in Fig. 4 that the CTE of Cu-SiC-Gr hybrid composites decreases with increase in % reinforcement. Introducing a high amount of graphite to the Al-Si matrix alloy was found to be beneficial for the dimensional stability. Results revealed that graphite particles absorb the thermal expansion because of their layered structure.

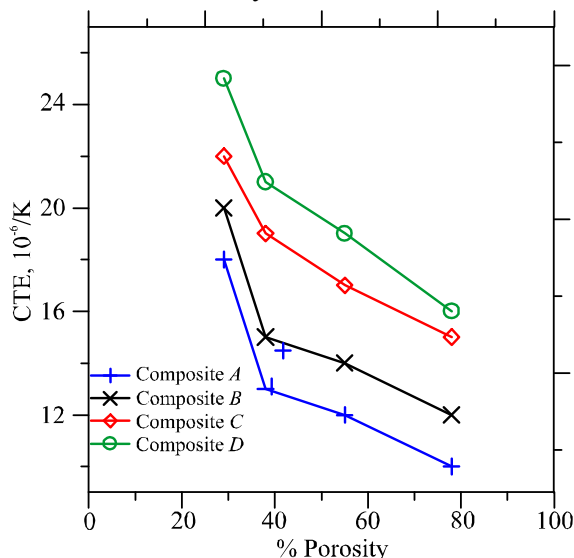


Fig. 4. Variations in CTE for different particulate composites (temperature: 100 °C).

Compressive Strength

Fig. 5 shows the compressive stress–strain curve for Cu-graphite-SiC composites. It is observed that compressive strength of a laminated composite is greater than that of a single layer one. This is due to higher density and hardness of the composites. It has been observed that the bending strength of three-layer Cu based composites is much greater than that of a single layer, due to residual compressive stresses in the outer layer.

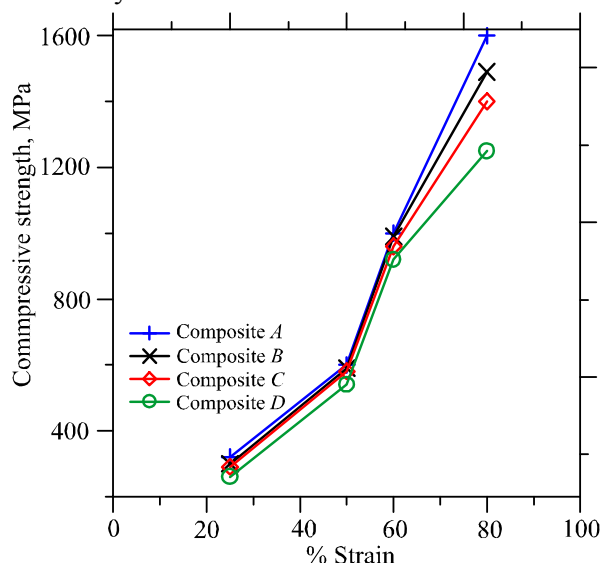


Fig. 5. Effect of reinforcements volume fraction on compressive strength.

Electrical Conductivity

From Fig. 6 it can be observed that the electrical conductivity of layered Cu hybrid composites containing 3 %, 6 %, 9 %, and 12 % SiC and 3 %, 6 %, 9 % and 12 % graphite tends to decrease linearly with increase of SiC/Gr. The ceramic based SiC forms a barrier to motion of copper electrons, providing electrical conductivity. SiC particles added into pure copper redouble the electrical resistivity via distorting the structure, and so the electrical conductivity of composites decreases with increasing the volume ratio of SiC [13].

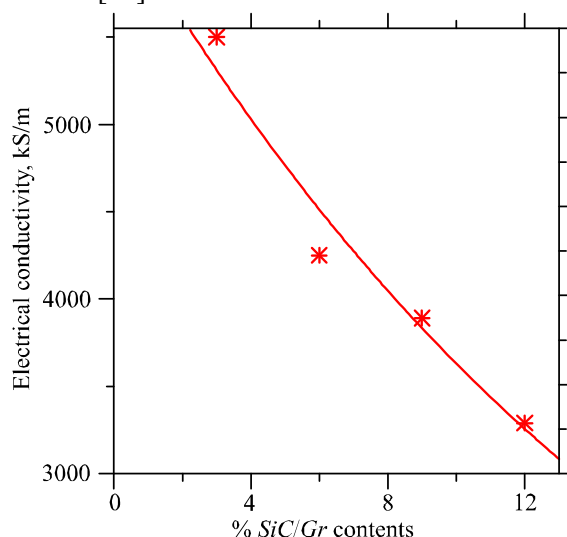


Fig. 6. Effect of reinforcements volume fraction on electrical conductivity.

Cu based metal matrix composite with very low SiC/Gr (0 – 4 %) is highly recommended to be used in many electrical contacts, thermal and electronic packaging applications, as it possesses high electrical conductivity.

Conclusions

The following points were concluded from the conducted tests of the composites:

1. Cu hybrid composites have been successfully fabricated by layer compaction and pressure sintering with varying dispersion of SiC and Gr particles. We have studied thermophysical properties of Cu/SiC/Gr hybrid composites in three-layer and single-layer configurations containing 3 %, 6 %, 9 % and 12 % SiC and 3 %, 6 %, 9 % and 12 % graphite.
2. The processing employed in this paper would enable realization of electrical contacts, friction materials, brakes and electronic packages made of Cu-Gr/SiC HMMCs.

References

1. Baoli Du, Ruizhi Zhang, Kan Chen, Amit Mahajan, and Mike J. Reece, The Impact of Lone-Pair Electrons on the Lattice Thermal Conductivity of the Thermoelectric Compound $CuSbS_2$, *J. Mater. Chem. A* **5**(7), 3249 – 3259 (2017).
2. Virgil Andrei, Kevin Bethke, and Klaus Rademann, Adjusting the Thermoelectric Properties of Copper(I) Oxide-Graphite-Polymer Pastes and the Applications of Such Flexible Composites, *Phys. Chem. Chem. Phys.* **18**, 10700 – 10707 (2016).
3. M. Lekka, D. Koumoulis, N. Kouloumbi, and P.L. Bonora, Mechanical and Anticorrosive Properties of Copper Matrix Micro- and Nano-Composite Coatings, *Electrochim Acta* **54**, 2540 –

- 2546 (2009).
4. Y. Zhan, G. Zhang, The Effect of Interfacial Modifying on the Mechanical and Wear Properties of *SiCp/Cu* Composites, *Mater Lett* 57, 4583 – 4591 (2003).
 5. K.M. Shu, G.C. Tu, The Microstructure and the Thermal Expansion Characteristics of *Cu/SiCp* Composites, *Mater Sci Eng A* 349, 236 – 247 (2003).
 6. J. Zhu, L. Liu, H. Zhao, B. Shen, and W. Hu, Microstructure and Performance of Electroformed *Cu/nano-SiC* Composite, *Mater Des* 28, 1958 – 1962 (2007).
 7. Th. Schubert, B. Trindade, T. Weibgarber, and B. Kieback, Interfacial Design of *Cu*-based Composites Prepared by Powder Metallurgy for Heat Sink Applications, *Mater.Sci.Eng.A* 475, 39 – 44 (2008).
 8. Mohsen Barmouz, Mohammad Kazem Besharati Givi, and Javad Seyfi, On the Role of Processing Parameters in Producing *Cu/SiC* Metal Matrix Composites via Friction Stir Processing: Investigating Microstructure, Microhardness, Wear and Tensile Behavior, *Materials Characterization* 62, 108 – 117 (2011).
 9. K.M. Shu, G.C. Tu, Fabrication and Characterization of *Cu-SiCp* Composites for Electrical Discharge Machining Applications, *Mater Manuf Processes* 16(4), 483 – 502 (2001).
 10. S.G. Sapate, A. Uttarwar, R.C. Rathod, and R.K. Paretkar, Analyzing Dry Sliding Wear Behaviour of Copper Matrix Composites Reinforced with Pre-Coated *SiCp* Particles, *Mater Des* 30, 376 – 386 (2009).
 11. Y. Zhan, G. Zhang, The Role of Graphite Particles in the High-Temperature Wear of Copper Hybrid Composites against Steel, *Mater.Des.* 27, 79 – 84 (2006).
 12. Y. Qin, Y. Wu, D. Wang, P. Li, X. Huang, and Y. Zheng, Influence of *SiC* Particle Size on the Wear Properties of *SiC/Cu* Composites, *Adv.Mater.Res.* 311 – 313, 635 – 639 (2011).
 13. T. Ram Prabhu, V.K. Varma, and Srikanth Vedantam, Tribological and Mechanical Behavior of Multilayer *Cu/SiC/Gr* Hybrid Composites for Brake Friction Material Applications, *Wear* 317, 201 – 212 (2014).
 14. C.S. Ramesh, R.N. Ahmed, M.A. Mujeebub, and M.Z. Abdullah, Development and Performance Analysis of Novel Cast Copper-*SiC-Gr* Hybrid Composites, *Mater. Des.* 30, 1957 – 1965 (2009).
 15. A. Meher & D. Chaira, Effect of Graphite and *SiC* Addition into *Cu* and *SiC* Particle Size Effect on Fabrication of *Cu-Graphite-SiC* MMC by Powder Metallurgy, Published online by Trans Indian Inst Met (2017).

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