IOP Conference Series: Earth and Environmental Science

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The Effect of Graphene Addition on the Microstructure and Properties of Graphene/Copper Composites for Sustainable Energy Materials

The Effect of Graphene Addition on the Microstructure and Properties of Graphene/Copper Composites for Sustainable Energy Materials

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Abstract. Graphene is a single thin layer (mono layer) of a hexagon-bound carbon atom and is an allotropic carbon in the form of a hybrid atomic plane, with a molecular bond length of 0.142 nm. Graphene is the thinnest and lightest material with 0.77 mg square meters, which exhibited excellent electricity and heat conductor. However, the perfect uniform microstructure, strength and optimum thermal properties of copper-graphene composites cannot be achieved because the amount of graphene does not reach the optimum level. In order to solve this problem, copper-graphene composites were produced by metal injection molding method (MIM) with various percentage of graphene, specifically 0.5%, 1.0% and 1.5% in the composite, to compare the physical and mechanical properties of these samples. MIM process involves the preparation of feed materials, pre-mixing process, mixing process, mold injection process, binding process and sintering processes. Feeding materials were used are copper and graphene, which have the powder loading of 62% with a mix of binder comprising 73% polyethylene glycol (PEG), 25% polymethyl methacrylate (PMMA), and 2% stearic acid (SA). Densification and tensile test were conducted to determine the mechanical properties. Scanning electron microstructure (SEM) was performed to obtain the microstructure of the composites. From the research, the result revealed that the 0.5% graphene content had the optimum parameter, which the hardness and tensile stress values were at 94.2 HRL and 205.22 MPa.

IOP Conf. Series: Earth and Environmental Science 1216 (2023) 012028 doi:10.1088/1755-1315/1216/1/012028

1. Introduction

Copper (Cu) is a ductile metal which has the properties of high thermal and electrical conductivity. It is commercially produced through the process of decomposition or smelting, followed by electrode solution which consists of sulphate solution. It is extensively used in electrical industry. Various studies and research were conducted to improve the physical and mechanical properties of Cu mainly on the nature of its thermal and electrical conductivity, in the area of Cu production processes or the preparation of alloyed Cu [1–3]. The graphene-copper alloys have become a hotpot where it has attracted many researchers globally to study on the mechanical and physical properties of the compositions [4,5]. Therefore, graphene was added to improve the physical and mechanical properties of this metal.

Graphene is a single thin layer (mono layer) of a hexagon- bound carbon atom like honeycomb, and also allotropic carbon in the form of a hybrid plane sp2 with a molecular bond length of 0.142 nanometers. Graphene is the thinnest compound and the lightest material with 0.77 mg square meters, which made it become the most powerful material being studied with the behavior as good electrical and heat conductor [6,7]. Based on researchers in Korea Advanced Institute of Science and Technology (KAIST), composite-reinforced graphene can achieve the tensile strength up to 500 times compared with the tensile strength exhibited by raw materials or monolithic materials [8,9].

Nowadays, the graphene continues to be aggressively studied for alloys as well as composites to improve the physical and mechanical properties of the metal [10–12]. Graphene was widely used for to form various type of composites due to its excellent mechanical properties. Powder injection molding (PIM) is a manufacturing process for producing near-net-shape metals and ceramics [13]. This technology produced excellent results in the production of complex or complex geometries, with significant cost savings and a large production capability. Conventional processes such as powder metallurgy (die compaction, isostatic pressing), deformation processes (forging, stamping), and casting processes (sand, die and investment casting) could not achieve these extraordinary results. PIM applications, however, are limited to monolithic materials such as stainless steel, titanium, alumina, cemented carbides, tungsten, and other alloys [14]. In this case, assembly of PIM product with other components were only performed after the PIM fabrication.

Metal injection molding (MIM) is one of the methods for the preparation of various composites. MIM is one of the processes in PIM. Powder metallurgy is a metal processing technique to integrate particles, where the metal powders were pressed into molds in stages and followed by sintering processes. Powder metallurgy portraited advantages in producing complex components and parts through special compositions by mixing elements and structural formulation methods in three-dimensional. Besides, the high performance and economically aspects had made researchers to consider applying powder metallurgy for this formulation [15].

In the metal powder process, metal powders and binders were mixed or kneaded to produce feed materials, which later injected into moulds in a green body at high pressure. Thermoplastic materials and candles were used widely as binder in this process. The binder was then removed from the green body via the binding process using solvent and heating, resulting in hollow and brittle brown bodies. The brown bodies are then sintered to produce the desired physical and mechanical properties of the composite material, such as density, strain, and microstructure.

Although the metal injection molding process has a good performance in physical, mechanical and microstructure properties of graphite copper composite, there were also other factors affected the composite microstructure. This includes the number of mixtures of graphite powder mixed for the manufacture of graphene copper composite during the metal injection molding process [16]. Perfect copper and graphene dispersion is hard to attain and the performance of physical and mechanical properties of copper graphite composites are still being investigated to optimize the physical and mechanical properties. Therefore, this study was conducted to determine desired amount of graphene to achieve optimum performance of its physical and mechanical properties.

2. Methods

The copper-graphene composite was prepared via powder injection molding (PIM) by using copper powder and graphene powder as a feedstock. The graphene composition was varied with 0, 0.5, 1.0 and 1.5%. The steps involved in PIM process were mixing, injection molding and debinding. After the PIM process, synthetized sample were tested for its hardness and tensile strength. Meanwhile, the grinding and polishing process is final approach for sample preparation, and it has several other steps before completion. For the grinding process, smooth sandpaper with the grades of 600, 800 and 1200 were used, where the direction of grinding was changed frequently for better grinding performance without any rough scratches. Water was used as lubricant for the grinding process, meanwhile oil and diamond-based lubricants were used for polishing to obtain the grain boundary during the analysis. In this method, ammonia and hydrogen peroxide were used as substance solution, with the constant ratio of 1:1. The ready-to-mix and polished samples were dipped into a mixture of ammonia and hydrogen peroxide solutions for 30 minutes before being observed under a microscope.

3. Results and Discussion

3.1. Microstructure of Copper-Graphene Composite

By using optical microscope, the copper-graphene composites with various graphene compositions (0.5, 1.0 and 1.5%) had revealed a microstructure, Figure 1(a)-(c). Through this analysis, it can be seen that the microstructure formed still has pores between high particles. The size of the area was increasing with the addition of the graphene composition in the copper matrix. The large area will cause dislocation of the copper matrix easily. This proves that the large area size can affect the strength of the metal composite. Furthermore, the viscosity of the copper graphene composite increased as the graphene composite increased. Because of the high viscosity, the materials flow thickly and slowly. As a result, the copper-graphene composite was more resistant, making it more difficult to fill the mold with pores between the particles and reduce the composite densification. As a result, during the grinding process, ammonia and hydrogen peroxide solutions eroded the metal between the metal's boundaries.

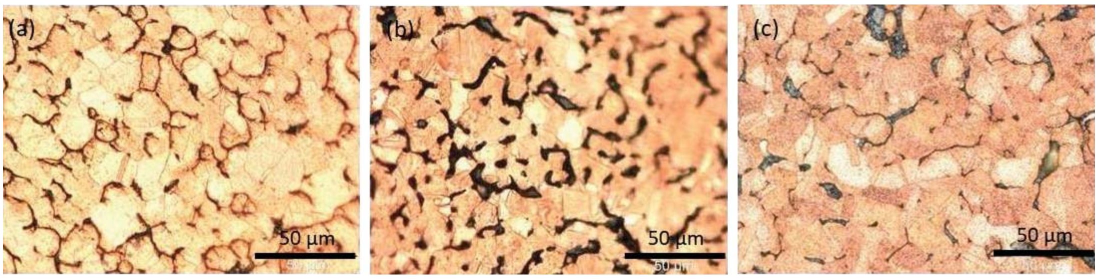

Figure 1. Microstructure of Copper-Graphene Composite by Optical Microscope; (a) 0.5%, (b) 1.0%, and (c) 1.5% of graphene.

SEM was used to observe the fracture surface of the copper-graphene composites after mechanical tensile test as shown in Figure 2(a)-(d). In addition to the temperature and binder type used, the composition of graphene content used in the formation of copper-graphene composites are also an important factor in determining the plastic properties of the composites. According to the fracture surface, graphite dispersion caused some changes in broken surfaces with the addition of graphene Figure2(a) illustrates the fracture surface of the reference pure copper specimen, which has huge voids and shallow dimples, as well as a higher percentage of the ruptured area covered by teardrop areas, all of which point to the ductile fracture in this specimen. Figure 2(b) shows the copper with 0.5% of graphene composite. It can be seen that the number of microscopic shallow dimples increased as a result of the plastic deformation that took place in this specimen during the tensile test, which contributed to attain the higher tensile strength. The mechanical strength decreases as the graphene composite increased. Themore cracks in the surface's fractured structure proved this Figure 2(c)-(d). In addition, the plasticity property of the copper-graphene composites was influenced by temperature and types of binder were used in this composite synthesis.

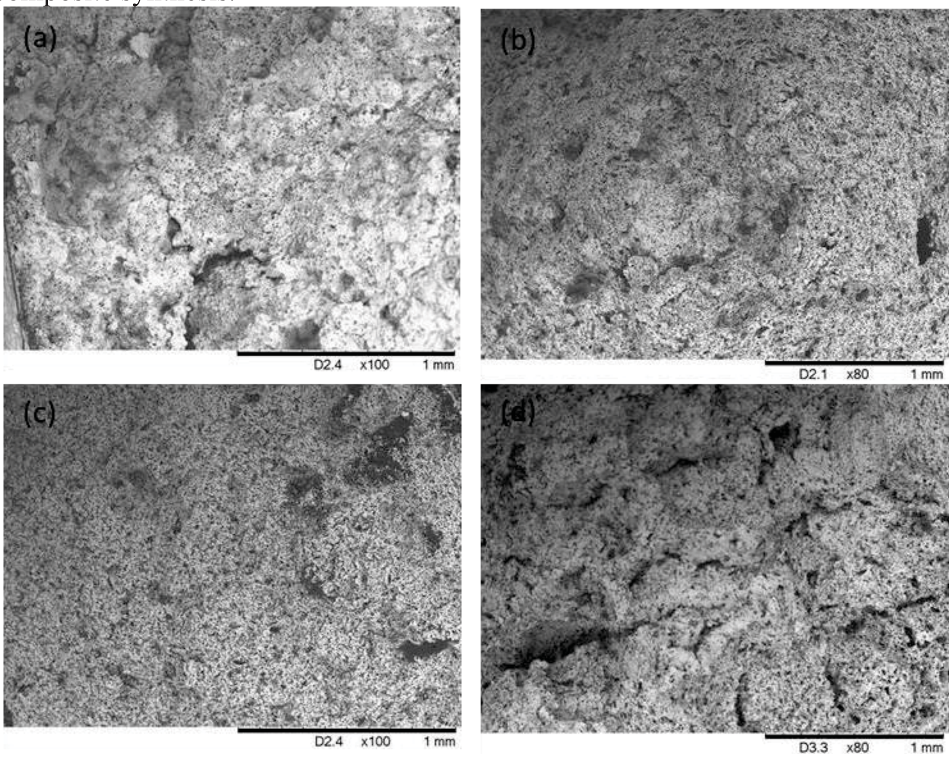

Figure 2. Fracture surfaces of the (a) reference pure copper, (b) Cu/0.5% graphene composite, (c) Cu/1.0% graphene composite and, (d) Cu/1.5% graphene composite

3.2. *Mechanical testing*

The Figure 3 shows the tensile strength and hardness values of copper-graphene composites with various graphene composition of 0%, 0.5%, 1.0% and 1.5%. Based on the above study, it can be observed that the compressive strength increases decently with the graphene content from 0 to 0.5%, but the strength decreases when the graphene content increased from 1.0% to 1.5%. For the tensile strength, the optimum strength was observed for graphene at 0.5%. This is due to graphene's effective dispersion in the copper matrix being best at 0.5% graphene. As the graphene composition increased, the graphene would agglomerate and disperse non-uniformly within the copper, decreasing the tensile strength value. Meanwhile, the Rockwell hardness graph for copper-graphene composites with various graphene composition of 0%, 0.5%, 1.0% and 1.5%. According to the studies, the composite hardness increases with graphene content of 0.5%, but gradually decreases with graphene content of 1.0% and 1.5%. This is due to the presence of graphene agglomerates in the copper matrix, which create obstacles in composite consolidation, increasing the distance between Cu powder particles and reducing the hardness value.

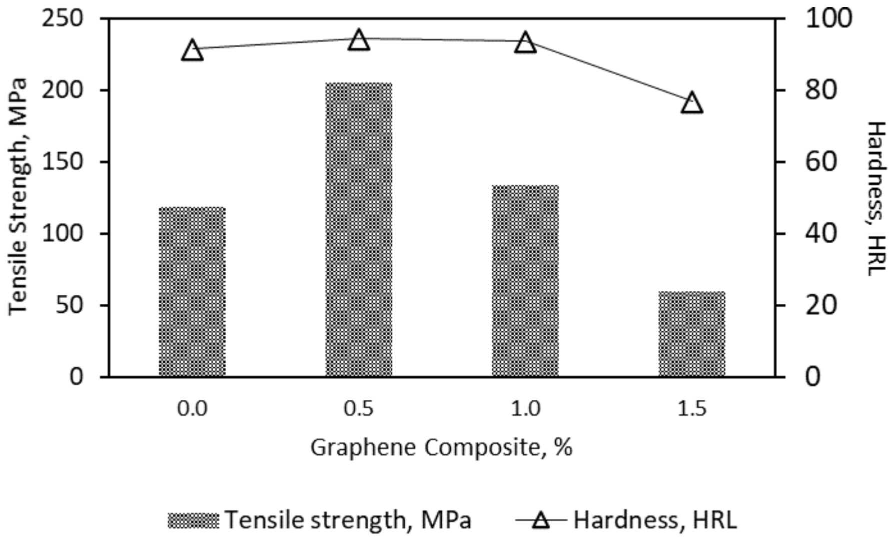

Figure 3. Tensile strength and hardness value for copper-graphene composite

4. Conclusion

Through this study, the optimum parameters were determined to improve the mechanical and physical properties of copper-graphene composites. The copper matrix is improved by the graphene through several mechanisms. The addition of the graphene causes microstructural changes in the matrix of the metal. Zone plastic changes also can be seen when the dislocation between the copper matrix occurs. This is due to the uneven thermal expansion between the graphene and the copper matrix. Another visible change is the reduction in particle size, wherein the addition of graphite can reduce the size of the particles and increase the resistance to the dislocation of the copper matrix. Copper was strengthened by graphene through several mechanisms. Graphene addition caused microstructural changes of the metal matrix. The plasticity behavior changes were observed due to thermal expansion mismatch between graphene and copper matrix. The first comparison, the results showed that copper-graphene composite hardness increased from 0% to 0.5% denoted as 91.9 to 94.2 MPa, respectively. Then the value of the copper-graphene composite decreased when the graphene content increased from 1.0% and 1.5%. For tensile strength, graphene patterns show the same trend as hardness graphs, where the tensile strength increased when the graphite composition is at 0.5%. Further increment of graphite shows the decreasing trend, respectively. The optimum value of strength was observed for graphite with 0.5% and 205.22 MPa.

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