Effect of mechanical ball grinding time on the properties and microstructure of graphite-copper composite

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Abstract. In this paper, the graphite-copper composite was prepared by powder metallurgy sintering method which used copper powder and graphene powder as raw materials. The powders were mixed by ultrasonic dispersion and mechanical ball grinding method. The effect of mechanical ball grinding time on the properties and microstructure of graphite-copper composite was investigated using Optical microscope, scanning electron microscope (SEM) and Vickers hardness meter. The result showed that the distribution of graphite in the copper matrix was uniformly and formed a good interface by using the ultrasonic dispersion and mechanical ball grinding method. When the mechanical ball grinding time was raised from 2 hours to 16 hours, the relative density and Vickers hardness of the graphite-copper composite reached their maximum value, the relative density was 92.1%, the Vickers hardness (HV0.5) was 61.9HV. However, the properties of graphite-copper composite showed a downward or stabilize trend the mechanical ball grinding time increasing from 16 hours to 48 hours.

1. Introduction

The components in engineering applications such as sliding parts of pantographs for trains and trams, brushes for Auto starters or welding machines are required to have good material properties: high specific electrical conductivity, satisfactory thermal conductivity and low friction coefficient [1]. Copper is known to has excellent electrical-thermal conductivity, high mechanical strength, good corrosion resistance, easy to obtain and low cost which has been selected as the matrix to prepare high performance electronic packaging materials. Graphite is known to its outstanding thermal properties, mechanical properties, corrosion resistant and low density. Graphite-copper composite materials as a new type of composite material gradually become a hot research topic. Cheng have successfully prepared copper-nickel-iron composites with 80% graphite content by using discharge plasma sintering technology(SPS) [2]. Ling have prepared graphite-reinforced copper-based composite coatings with good self-lubrication and wear resistance through a low-pressure cold spraying process [3]. Liu have prepared graphite sheet-copper composites using vacuum thermal pressure [4]. Zhu used powder metallurgy to prepare natural graphite/copper (NG/Cu) composites and carbon-coated graphite/copper (CCG/Cu) composites [5]. Kavaliauskas used plasma generators to mix two different powders with different concentrations of powder preparation coating, and then screen the raw powder thus a composite coating of graphite copper is prepared using stainless steel (304L) and quartz glass as substations [6]. Zhu L use hot-press sintering to prepare copper-graphite composites [7]. However, the preparation of high-performance copper-graphite composites is not easy with existing technologies. Because the solubility of graphite in the copper matrix is relatively low, copper cannot react with graphite to form the corresponding carbides. Therefore, the wetting between the copper matrix and
graphite powder is very poor, so the interface between the two materials are not good. In this paper, the copper powder and graphite powder were used as raw materials to fabricate the graphite-copper composite by low temperature cold pressure powder metallurgy sintering method. The powders were mixed by ultrasonic dispersion and mechanical ball grinding process. The effects of mechanical ball grinding time on the organization and performance of graphite-copper composite were investigated.

2. Experimental procedure
The Graphite-Copper composites with 1 wt.% content of graphite phase were prepared from the mixture of copper and graphene powder by the powder metallurgy sintering method. The raw powder materials used in this experiment are granular 3.4-8nm, with a surface area of 100-300m$^2$/g, the selected powder purity is greater than 99%, and the graphene powder has the number of layers between 6 and 10 layers which is mechanically stripped.

Firstly 0.5g graphene powder and 0.5g titanium powder were added to a beaker filled with 200 mL of waterless ethanol for ultrasonic shock of 45 min to disperse; then the ball grinding tank was placed in the planetary ball mill (GQM-5-2) for mechanical ball grinding mixing, ball grinding speed was 400 r/min, ball grinding ratio was 10:1, mechanical ball grinding time was 2 hours, 6 hours, 12 hours, 16 hours, 24 hours, 36 hours and 48 hours. After the ball grinding, the slurry is filtered through a metal mesh and placed into an electric heating and drying tank at 60 ℃. Secondly an electronic balance was used to take 5 g dry compound powder, loaded into a high-strength graphite mold with 20 MPa pressure to maintain 15min pre-pressurization forming. It was then sintered in a vacuum carbon tube sintering furnace (CXZT-20-20) at temperatures of 700 ℃. After sintering process, the oven was cooled to room temperature for de-molding sampling.

Finally, the Leica microscope (Germany, DM1500M E) was used to observe the morphologies of graphite-copper composites and the field emission scanning electron microscope (HITACHI, SU8010) was used to observe the combination between the copper matrix tissue and the graphite tissue of the composite materials. Archimedes drainage method was used to measure relative density of composites (electronic balance accuracy 0.001 g, measured 5 times, averaged); Vickers hardness meter (HVS-1000S) was used to measure the hardness of the composite (test load is 1.961 N, the pressure is 10 s, 7 measurements are taken, averaged).

3. Results and discussion

3.1. Effect of mechanical ball grinding time on relative density of the graphite-copper composite

![Figure 1. The relative density of composites with varies mechanical ball grinding time](image)

The relative density of the graphite-copper composites with mechanical ball grinding time varying from 2 hours to 48 hours were listed in figure 1, the result showed that the relative density of the composites increased gradually with the mechanical ball grinding time increasing at the beginning. The relative density of the composites reached its maximum value 92.1% at the mechanical ball
grinding time of 16 hours. And then the relative density of the composites decreased as the mechanical ball grinding time continued to increase. With the mechanical ball grinding time increasing from 2 hours to 16 hours, the diffusion of graphite in the copper matrix becomes more uniform and with the increase of the deformation of the material, it was more conducive to welding pores thus increasing the tightness and binding of the composite material. As the mechanical ball grinding time increased over 16 hours, there were more defects or disordered tissues in graphene as ball grinding time increases, and the energy accumulated during ball grinding was greater than the chemical energy of the C-C bonding. So the mechanical ball grinding does facilitate the dispersion of graphene in copper matrix, but long-term ball grinding could cause serious damage to graphene.

3.2. Effect of mechanical ball grinding time on Vickers hardness of the graphite-copper composite

![Graph showing Vickers hardness vs. mechanical ball grinding time](image)

Figure 2. The Vickers hardness of composites with varies mechanical ball grinding time

From the Vickers hardness measurement experiments of graphite-copper composite after sintering (figure 2), it could be seen that the strength performance of the composite was very sensitive to the mechanical ball grinding time, after the short period of ball grinding time (2 hours), the Vickers hardness of composite was 52.9. with the increase of ball grinding time, the Vickers hardness of the composite increases a lot, but when the ball grinding time was more than 12 hours, the Vickers hardness stabilized and at the mechanical ball grinding time of 16 hours, the Vickers hardness of composite achieved the maximum value 61.9. The reason why the composite's Vickers hardness was improved mainly because while in short ball grinding time, the two material powders did not occur more complete mixing uniform, after a long time of mechanical ball grinding, the two materials powders mixed more fully, the mechanical performance could be more uniform and could jointly bear the load imposed by deformation, complementary advantages which showed a very good performance of the force.

3.3. Effect of mechanical ball grinding time on microstructure of the graphite-copper composite

Figure 3 shows the microstructures of composite with various mechanical ball grinding time. It could be observed that there were two main tissues of composite materials: white copper matrix tissue and gray enhanced graphite tissue. The combination between the copper matrix tissue and the graphite tissue was well and the enhanced graphite tissue was more uniformly dispersed, but there was still a reunion. Through scanning electron microscope (SEM) observation, it could be seen that the enhanced graphite tissue body was flaky and the reunion part had more pores. The reunion phenomenon happened because graphene and copper lubricity was poor. However, with the proper extension of mechanical ball grinding time, the tissue of the composite become denser and there were no obvious pores, and the graphite phase could be evenly distributed in the copper matrix, increasing its density and hardness.
4. Conclusions
In this paper, powder metallurgy sintering method was used to prepare graphite-copper composite materials while graphite and copper powders were used as raw materials. The effects of mechanical ball grinding time on the properties and microstructure of graphite-copper composite were investigated using Optical microscope and scanning electron microscope (SEM) and hardness meter. The results showed that the distribution of graphite on the copper matrix was uniformly and formed a good interface by using the ultrasonic dispersion and mechanical ball grinding method. When the mechanical ball grinding time was raised from 2 hours to 16 hours, the relative density and Vickers hardness of the graphite-copper composite reached their maximum value, the relative density was 91.1%, the Vickers hardness (HV0.5) was 61.9HV. However, the properties of graphite-copper composite showed a downward or stabilize trend with the mechanical ball grinding time increasing from 16 hours to 48 hours.

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