Effect of sintering insulation time on the properties and microstructure of graphite-copper composite

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Abstract. 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 sintering insulation time on the properties and microstructure of graphite-copper composite were investigated. The result 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 process. When the sintering insulation time was raised from 3 hours to 18 hours, the friction coefficient and wear rate were decreased in the initial stage (from 3 hours to 12 hours) meanwhile the Vickers hardness (HV0.5) reached its maximum value 61.9HV when the sintering insulation time was 12 hours. The SEM (scanning electron microscope) images indicated that the increased sintering insulation time was helpful to distribute the graphite phased in composite relatively homogeneous with the sintering insulation time within 12 hours. However, the properties of graphite-copper composite also showed a downward or stable trend with the sintering Insulation time increasing from 12 hours to 18 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 becomes a hot research topic. Cheng successfully prepared copper-nickel-iron composites with 80% graphite content by using discharge plasma sintering technology (SPS) [2]. Ling prepared graphite-reinforced copper-based composite coatings with good self-lubrication and wear resistance through a low-pressure cold spraying process [3]. Liu 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 screened the raw powder thus a composite coating of graphite copper was prepared using stainless steel (304L) and quartz glass as substations [6]. Zhu L used 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 is not good. In order to further explore the effect of sintering insulation time on the organization and performance of graphite-copper composite, this study took natural graphite and atomized copper powder as raw materials, used ultrasonic dispersion and mechanical ball grinding to prepare graphite-copper composite powder, then used low temperature cold pressure sintering technology to prepare graphite-copper composite and finally investigated the microstructure and properties of graphite-copper composite with various sintering insulation time.

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 low temperature cold pressure powder metallurgy sintering method. The raw powder materials used in this experiment was granular 3.4-8nm, with a surface area of 100-300m²/g, the selected powder purity was greater than 99%, and the graphene powder had 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 16 hours. After the ball grinding, the slurry was filtered through a metal mesh and placed into an electric heating and drying tank at 60 °C.

Secondly an electronic balance was used to take 5g 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 °C. The sintering insulation time was set to 3 hours, 6 hours, 9 hours, 12 hours, 15 hours and 18 hours. The oven was cooled to room temperature for de-molding sampling.

Finally, the Leica microscope (Germany, DMI5000ME) 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. 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). The friction wear test machine (MMUD-10B) was used to test the friction coefficient and wear rate of the graphite-copper composites.

3. Results and discussion

3.1. Effect of sintering insulation time on Vickers hardness of the graphite-copper composite

![Figure 1](image1.png)

Figure 1: The Vickers hardness of composites with various sintering insulation time

From the Vickers hardness measurement experiments of graphite-copper composite after fabricating (figure 1), it could be seen that the strength performance of the composite was very sensitive to the
After the short period of sintering insulation time (3 hours), the Vickers hardness of composite was 42.8. With the increase of sintering insulation time, the Vickers hardness of the composite increased a lot, but when sintering insulation time was more than 9 hours, the Vickers hardness almost stabilized and at the sintering insulation time of 12 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 the sintering insulation time increased in the beginning, the diffusion of graphite in the copper matrix became more uniform. With the increase of the deformation of the material, it was more conducive to compact pores thus increasing the tightness and binding of the composite material. As the sintering insulation time increased over 12 hours, the diffusion of graphite in the copper matrix became stable and the Vickers hardness of graphite-copper composite almost had no increase. Meanwhile with the energy accumulated during sintering insulation period was greater than the chemical energy of the C-C bonding, the mechanical properties of composites had a slowly decrease trend. So the increasing sintering insulation time does facilitate the dispersion of graphene in copper matrix, but long-term sintering insulation could cause the hardness to decrease.

3.2. Effect of sintering insulation time on friction coefficient and wear rate of the graphite-copper composite

The friction and wear tests were conduct by using MMUD-10B test machine. Figure 2 showed the revolution of friction coefficient and wear rate of the graphite-copper composite. It revealed that the trend of wear rate followed the trend of friction coefficient that with sintering insulation time increased from 3 hours to 12 hours, the wear rate and friction coefficient decreased gradually and reached the minimum volume which were 0.013g and 0.21. The reason of obtaining decreased wear rate and friction coefficient was that with the increase of sintering insulation time, graphene was more evenly distributed in copper powder matrix, metal atoms in the composite are more diffused, and the bonding strength between graphite powder and copper matrix was enhanced thus making the composite getting more excellent frictional wear performance. But when the sintering insulation time increased from 12 hours to 18 hours, the wear rate increased gradually from 0.013g to 0.019g and the friction coefficient increase gradually from 0.21 to 0.23. The reason was that the distribution of graphene in copper powder tended to stabilize and with the energy accumulated during sintering insulation period greater than the chemical energy of the C-C bonding, the structure of graphene could be damaged, which increased the friction coefficient and wear rate.

3.3. Effect of sintering insulation time on microstructure of the graphite-copper composite

Figure 3 shows the microstructure images and SEM image of graphite-copper composite with various sintering insulation time. It could be observed that the graphite-copper composite was free from obvious pores due to Cu/Cu contact during sintering. And there were two main tissues of composite materials:
white copper matrix tissue and gray enhanced graphite tissue. The graphite phased in composite distributed relatively homogeneous. The combination between the copper matrix tissue and the graphite tissue was well and the enhanced graphite tissue was more uniformly dispersed. But through scanning electron microscope (SEM) observation there was still the existence of some gaps in the interface between copper and graphite. These gaps as crack sources would be easy to extend when the composites were subjected to an applied normal load, thus greatly reducing the mechanical properties.

![Image](image_url)

Figure 3 The microstructure of composites with various sintering insulation time

4. Conclusions

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 sintering insulation time on the properties and microstructure of graphite-copper composite were investigated. 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 process. When the sintering insulation time was raised from 3 hours to 18 hours, the friction coefficient and wear rate are decreased in the initial stage (from 3 hours to 12 hours) meanwhile the Vickers hardness (HV0.5) reached its maximum value 61.9HV when the sintering insulation time was 12 hours. The SEM (scanning electron microscope) images indicated that the increased sintering insulation time was helpful to distribute the graphite phased in composite relatively homogeneous with the sintering insulation time within 12 hours which caused the combination between the copper matrix tissue and the graphite tissue was well and the enhanced graphite tissue was more uniformly dispersed. However, the properties of graphite-copper composite showed a downward or stable trend with the sintering insulation time increasing from 12 hours to 18 hours.
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