リン酸マグネシウム処理したグラファイト粒子を分散した銅系複合材料の開発

Development of copper-based composite material dispersed with magnesium phosphate treated graphite particles.

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ABSTRACT

Graphite as solid lubricant often added into the copper matrix friction materials to stabilize the friction coefficient and reduce wear rate during sliding by forming a graphite-rich transfer layer on the counterpart surface. The use of magnesium phosphate to treat graphite powder has been proven in prior report as a method to improve the oxidation resistance of graphite, and the results showed that after magnesium-phosphate treatment, the treated graphite still maintaining the original structure of graphite.

The present study using this magnesium phosphate treated graphite and copper powder together to make a more superior Cu-G composite material. In this study, copper/magnesium phosphate-treated graphite (Cu-MgPG) composites were prepared by powder metallurgy technology, and the structure and friction properties of the prepared samples were analyzed.

1. Introduction

Copper-graphite composites have the properties of copper with excellent thermal and electrical conductivity, and the properties of graphite with solid lubrication and low coefficient of thermal expansion. Therefore, it is often used as a brush and bearing material as a composite material with self-lubricating properties [1].

The self-lubricating mechanism of the copper-graphite composite is thought to be that the graphite exposed on the frictional contact surface acts as a solid lubricant in the process of friction. As a result, the properties of the graphite phase in copper-graphite composites play the most important role in the self-lubrication of the material.[2]

In the case of liquid lubrication, it is not a difficult technique to continuously and smoothly supply lubricating oil to the sliding surface. On the other hand, in the case of solid lubrication, it is important how to supply it to the sliding surface or how to continue to exist on the sliding surface for a long time.

In the conventional copper-graphite composite material, graphite, which is a dispersion material, is supplied to the surface as it wears. However, when sliding in the atmosphere, the sliding surface becomes hot, and it is expected that the graphite particles will oxidize and volatilize. As a result, friction and wear characteristics are reduced.

Therefore, we performed magnesium phosphate treatment in order to improve the oxidation resistance without impairing the characteristics of graphite particles, which are layered compounds, as a solid lubricating material. This treatment not only improves the oxidation resistance, but also easily adheres to the sliding surface, and by forming a film, it may stay on the sliding surface for a longer period of time, and as a result, it takes a longer time than before. We thought that it might be possible to maintain low friction and low wear.

The aim of this research is using powder metallurgy to produce more superior Cu-G composite material. While, in the past work [3] we proposed a method to improve the oxidation resistance of graphite while maintaining the lubricating properties of graphite. In this study, we used magnesium phosphate-treated graphite as a solid lubricating phase to mix with copper powder, and used powder metallurgy technology to make copper/magnesium phosphate-treated graphite (Cu-MgPG) composites.

Past study[4] have shown that when the volume content of graphite is less than 50%, sintering temperature lies in 700 °C~950 °C to improve the metallurgy bonding between copper and copper in the material. When the graphite volume content is more than 70%, sintering temperature is 120 °C~550 °C, and the bond strength of brush materials is obtained by resin cross-linking curing. In addition, considering that the sintering temperature for the preparation of magnesium phosphate treated graphite is 800 °C [3], this study will prepare copper matrix composites with low graphite content (10 vol%) at a sintering temperature of 800 °C. At the same time, parallel compacts made from pure copper powders and parallel compacts made from copper powders and natural graphite powders were consolidated under the same conditions. The structure and friction properties of the prepared samples were analyzed.

2. Experiment

2.1. Material and procedure

The magnesium phosphate-treated graphite (MgP-graphite) raw material used in this study was prepared according to the method of previous study[3]. The arithmetic mean particle size of the magnesium phosphate-treated graphite (natural graphite powder, D50 93.16 μ m; Mg(H2PO4)2 • 4H2O, MW:290.34, JUNSEIKAGAKU) used in this work was 131.70 μ m.

The composites were fabricated by powder metallurgy technique. 10vol% MgP-graphite powder and copper powder (Nilaco) were dry mixed with a bench kneader machine (Irie Shokai Co., Ltd, Tokyo Japan) for 3h. after mixing, the powder mixture was put into a 20×20mm size cylindrical mold and hot-pressing in a nitrogen environment at 800°C with a pressure of 30Mpa for 3h. After the specimens cooled to room temperature, the preparation of composite materials was completed. For comparison, parallel compacts made from pure copper powders and parallel compacts made from copper powders with natural graphite powders were consolidated under the same conditions applied for Cu-MgPG composites. All specimens were polished and degreased with acetone before every experiment.

2.2. Friction coefficient and wear test

The friction coefficient and wear sliding tests was carried out in a pin-on-disc tribometer (T-18-0162, NANOVEA, US) in ambient conditions (20 ± 0.5 °C and $45\% \pm 5\%$ RH). Tribological performance of the prepared composite materials was studied in dry sliding where counterbodies were polished balls with 8 mm diameter made out of commercially available balls - SUJ2 bearing steel (Sato Tekkou, Japan). Before test, both the composite materials and the counterbody were ultrasonically cleaned in an acetone bath for 10 min. Sliding friction and wear tests were done at 10N loads with a circular sliding and the sliding ve locity was 0.1 m/s, the wear track radius was 5 mm, and the total sliding distance was 2000 m. The friction coefficients and wear volume were recorded continuously.

2.3. Measurements

The densities of the sintered MgP-graphite/copper composite and copper block were measured by Archimedes' principle. The hardness was investigated using a Micro Vickers hardness tester (HMV-G, SHIMADZU) under a load of 0.5kg with a dwell time of 10 s. The measurements for each sample were carried out 5 times, and the data obtained were the mean values. The surface morphology and elemental distribution of the samples were characterized using a scanning electron microscope (SEM, JSM-7500F, JEOL) equipped with energy dispersive X-ray detector (EDS, JEOL).

3. Results and discussion

The physical properties of the sintered composite are shown in Table 1. The theoretical density was calculated by dividing the density of the sintered composite by the rule of mixtures using 8.95 g/cm3, and 2.25 g/cm3 as densities for copper and graphite, respectively.

Table 1.1 hysical properties of the sintered composite block			
Sample types	Cu block	Cu-G block	Cu-MgPG block
Relative density (g/cm ³)	8.89	8.35	8.27
Theoretical density (g/cm ³)	8.95	8.36	8.36
Vickers hardness(Hv)	39.73	50.95	54.83

Table 1. Physical properties of the sintered composite block

From the comparison of the relative density and theoretical density of the tested samples, the samples prepared by the powder metallurgy method used in this study all have dense structures. Among the samples prepared under the same conditions, the Cu-MGPG composite has higher hardness.

Friction coefficients of these composites were determined from their adjacent areas of center using pin-on-disk tribometer. Figure 1 shows result of friction coefficient test for Cu, Cu-G, Cu-MgPG composite. Reported friction coefficient in this study is average of these data in steady state stage.

Fig. 1 (a) shows the changes friction coefficients of Cu with distance. And the final friction coefficient in steady state stage is 0.81, while Fig.1 (b), (c) shows the changes friction coefficients of Cu-G and Cu-MgPG with distance, the final friction coefficient of Cu-G and Cu-MgPG are 0.19 and 0.15 respectively. which proves that the Cu-MgPG composite has better friction properties.

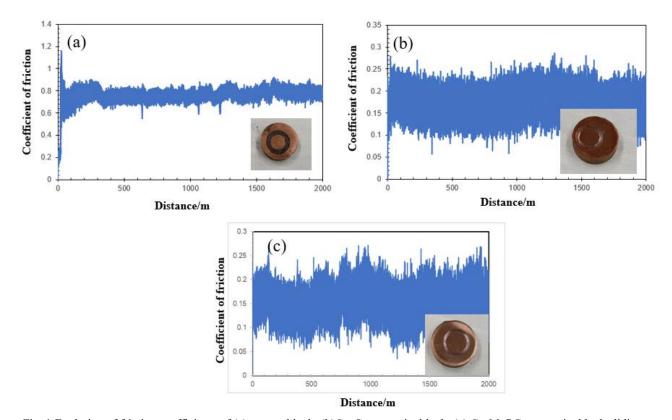


Fig. 1 Evolution of friction coefficients of (a) copper block; (b)Cu-G composite block; (c) Cu-MgPG composite block sliding against SUJ₂ counterbodies at 10N.

Figure 2 displays SEM images of all samples after sliding test, it can be seen from Fig.2 that under the same experimental conditions, after the friction test, the surface of the copper block will leave wider scratches, and the surface of the scratch is distributed with a large area of debris. The surface morphology of the Cu-G sample and the Cu-MgPG sample is similar. It is clear that the graphite particles dispersed in the copper, and the scratches after sliding are narrower than that appeared in pure copper block.

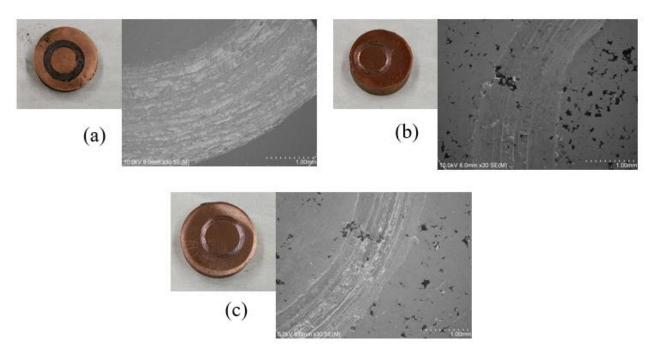


Fig. 2 SEM image of (a) copper block; (b)Cu-G composite block; (c) Cu-MgPG composite block after sliding test

Figure 3 shows elemental distribution of the samples on the worn surface with EDS. Fig.3 (a) shows that the presence of Fe

element was detected in the wear scratches left on the copper surface after sliding against SUJ2 ball. This shows that there is a lot of loss in the counterbody during the friction process, and it remains on the copper surface. According to Fig. 3(b), after the sliding experiment, Fe element was not detected in the scratches left on the surface of the Cu-G sample. The detected C element proved that graphite was dispersed in the sample. The O element proves that during the sliding friction process, a large number of oxides are produced on the contacting surfaces. Figure 3(c) shows that the surface element distribution of the Cu-MgPG sample is similar to that of the Cu-G sample, and the P element detected at the same time as the C element proves the existence of magnesium phosphate treated graphite.

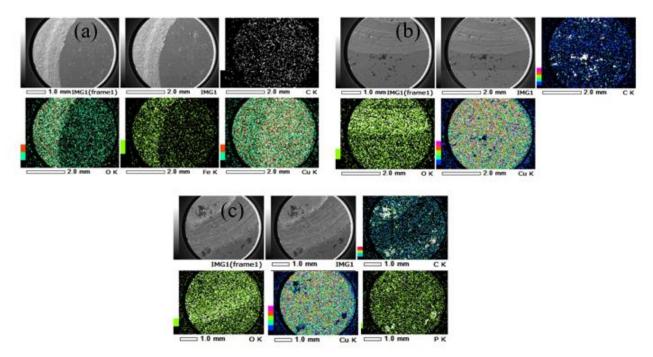


Fig. 3 EDS mapping results of (a) copper block; (b)Cu-G composite block; (c) Cu-MgPG composite block after sliding test

4. Conclusion

Copper / magnesium phosphate treated graphite (Cu-MgPG) composites were prepared by powder metallurgy technology, the structure and friction properties of the prepared samples were analyzed.

The present study demonstrates the feasibility of preparing composites with self-lubricating properties using magnesium phosphate-treated graphite and copper powders. At the same time, it was found that the Cu-MgPG composites prepared by using magnesium phosphate-treated graphite as raw materials have better friction properties than ordinary Cu-G composites. Which means that Cu-MgPG composites have the opportunity to be applied in a wider temperature range than normal Cu-G composites material, while also possessing better friction properties

In the future research, we will conduct a more in-depth analysis of the wear surface composition of the three samples, such as using Raman, XRD, XPS, etc. As a result, we want to study and analyze the mechanism of the friction performance improvement of Cu-MgPG composites.

References

- S.F. Moustafa, S.A. El-Badry, A.M. Sanad, B. Kieback, Friction and wear of copper–graphite composites made with Cucoated and uncoated graphite powders, Wear 253 (2002) 699–710.
- 2) H. Sarmadi n, A.H. Kokabi, S.M. Seyed Reihani, Friction and wear performance of copper–graphite surface composites fabricated by friction stir processing (FSP), Wear 304(2013) 1-12
- 3) R.Li et al, Investigating the chemical bonding state of graphite powder treated with magnesium (II) phosphate through EELS, TEM, and XPS analysis, Diamond & Related Materials, S0925-9635(21)00186-2
- 4) Xia JT, Zhong-Liang HU, Chen ZH. Preparation of carbon brushes with thermosetting resin binder. Trans Nonferrous Met Soc China 2007;17(6):1380–4.