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Surface modification of h-BN and its influence on the mechanical properties of $CuSn_{10}$/h-BN composites

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ABSTRACT

$CuSn_{10}$ matrix composites with hexagonal boron nitride (h-BN) in concentrations of 4, 7, 10, and 14 vol.% were prepared using a powder metallurgy process. The effect of modifying the h-BN with anionic polyacrylamide (APAM) by high-energy ball milling was investigated. The bonding mechanism between h-BN and APAM was characterized by Fourier transform infrared spectroscopy (FT-IR), X-ray photoelectron spectroscopy (XPS), and X-ray diffraction (XRD). The microstructure, hardness, tensile strength, and bending strength of sintered composites were investigated. The composite $CuSn_{10}$-4 vol%h-BN showed the lowest porosity of 20.1%. The long APAM chains could be introduced on the h-BN surface without altering its crystal structure. The surface modified h-BN exhibited less agglomeration in the matrix than pristine h-BN, due to the physical entanglement and chemical bonding between h-BN and APAM. Comparing the composites with 4 vol.% h-BN, without and with APAM modification, a reduction in the porosity from 20.1% to 5.5%, and increase in the tensile strength from 134 to 147 MPa, and a significantly increased in the bending strength from 238 to 351 MPa, respectively, were observed.

1. Introduction

Many tribological system working in high-temperature aggressive environments demand combinations of a low friction coefficient, good high-temperature oxidation resistance, and good wear resistance [1]. Tin bronze matrix powder metallurgy (PM) composites with excellent tribological properties and good heat conductivity and corrosion resistance have been widely investigated [2–4].

Traditional solid lubricants, such as graphite and MoS_2, are good choices for tin bronze friction materials, where their lamellar structures are beneficial for decreasing friction, but most such lubricants can only be used under ambient temperature [3,5–7]. Hexagonal boron nitride (h-BN), is an inorganic analogue of graphene and has a lamellar structure consisting of a stack of hexagonal sheets with a strong covalent bond between boron and nitrogen atoms. The sheets are held together by weak van der Waals forces that allow shearing when force is applied parallel to the sheets. Thus, it provides the expected friction reduction and results in very efficient lubrication over a wide temperature range [1,8–13]. However, due to the surface incompatibility between h-BN and the matrix alloy, the physical mixing of h-BN particles leads to phase segregation between the h-BN and matrix, giving rise to agglomeration of h-BN particles, which may negatively affect the physical and mechanical properties of the resulting composites [1,14–16]. Thus, reducing the agglomeration of h-BN particles and improving the interfacial adhesion between h-BN and the matrix surface have been the most important issues for the development of a tin bronze/h-BN composite.

To break down the particle agglomerates and improve the dispersibility of the particles in the matrix, surface modification is an effective and commonly used method [17]. Silicon carbide has previously been modified by a silane coupling agent and hexadecyl iodiele [18]. Silane coupling agents with different carbon chains (C3 and C16) were introduced on the BN surface to improve the affinity of BN for the epoxy resin [19]. In the work of Li et al., TiO_2 was modified by KH550 through a silanization reaction [20].

Anionic polyacrylamide (APAM) is produced during the polymerization reaction of acrylamide and has both amide and carboxyl functional groups [21]. APAM gel is a typical soft material that has a unique, three-dimensionally cross-linked network structure swollen with a large amount of water. The unique structure of these

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systems means they can be safely applied in materials processing and hence the study of these materials is of high utility. Currently, APAM is mainly applied in cosmetics, pharmaceutics and paint production, minerals flotation, oil recovery, agriculture, and environmental protection applications [22–24].

In order to realize homogeneous dispersion of h-BN in a tin bronze matrix, here we propose to prepare $CuSn_{10}$/h-BN composites using APAM as a surface modifier. The modification mechanism of APAM on the surface of h-BN was studied. In addition, the influence of the APAM on the mechanical properties of the composite made with the modified h-BN was investigated.

2. Experimental

2.1. Materials and processing

$CuSn_{10}$ powder with an average particle size ~35 μm with a purity of 99.99% was used as the metal matrix material. Commercial h-BN (99.9% purity) was used as a lubrication material while the other powders (Ni, Fe, Mn, and WC) were used as reinforcement materials. All these powders were passed through a 200 mesh sieve. APAM has the chemical formula $-CH_2-CH(CONH_2)-CH_2-CH(COONa)-$ and an average molecular weight of 14000000.

Fig. 1 shows a flowchart of the experimental procedure used in this study.

Table 1 shows the five different $CuSn_{10}$ composite materials prepared here with different volume fractions of h-BN. The $CuSn_{10}$ composites with 4, 7, 10, and 14 vol.% h-BN are designed as samples 1A–4A, while sample 1B is the $CuSn_{10}$ composite with 4 vol.% modified h-BN.

Firstly, for samples 1A–4A, mixtures of $CuSn_{10}$, h-BN, and the strengthening elements were prepared by planetary ball milling at about 80 rpm for 48 h with a mass ratio of balls to powder of 6:1. Then the mixtures were bidirectionally pressed at room temperature under 500 MPa. The obtained pellets were sintered under a hydrogen atmosphere at 600 °C for 60 min and then at 1100 °C for 90 min with a heating rate of 10 °C/min.

For sample 1B, the $CuSn_{10}$, 4 vol%h-BN, strengthening elements, and 1.5 wt.% APAM and 15 wt.% water were mixed for 48 h in a planetary ball mill with a mass ratio of balls to powder of 6:1. After drying at 80 °C for 8 h, the obtained powders underwent the same compaction and sintering process as samples 1A–4A.

2.2. Characterization and testing

The relative bulk density was calculated from the ratio of the bulk density to the theoretical density. The porosity was calculated from the density differences measured using Archimedes principle.

Table 1
Chemical compositions of $CuSn_{10}$/h-BN composites (vol.%).

Sample designation	h-BN	Ni + Fe + Mn	WC	$CuSn_{10}$
1A	4	18	1	Balance
2A	7	18	1	Balance
3A	10	18	1	Balance
4A	14	18	1	Balance
1B	4	18	1	Balance

The morphology and composition of the samples were characterized using field-emission scanning electron microscopy (SEM) using an instrument equipped with an energy-dispersive X-ray spectrometer (EDS). The X-ray diffraction (XRD) analysis was used to identify the crystal structure of the pristine and modified h-BN powders. Electron probe microanalysis (EPMA) was used to investigate the dispersion of h-BN.

Fourier transform infrared (FT-IR) spectrometry was performed using a Thermo Scientific Nicolet 6700 instrument and the KBr pellet method in the range of 500–4400 cm^{-1} with a resolution of 4 cm^{-1}. The chemical composition and functional groups were characterized by X-ray photoelectron spectroscopy (XPS; Thermo Scientific K-Alpha) at 300 W (Al K radiation).

The zeta potential of untreated and modified h-BN powders in an alcoholic solution was measured with a Zetazier 3000HSA instrument. Wet grinding of h-BN modified by APAM was carried out by high-energy ball milling. Pristine h-BN with APAM in concentrations of 0, 1, 1.5, 2, and 2.5 wt.% and 15 wt.% deionized water were mixed for 24 h. After drying the wet mixture, the samples were homogeneously dispersed in alcohol by sonication for 1 h.

Vickers microhardness measurements of the obtained specimens were determined on the polished surface considering an average of five indentations for each specimen using Vickers indentation with an indentation load of 4.9 N for 10 s. Tensile tests were carried out according to ASTM D638 with a crosshead speed of 2 mm/min and a gauge length of 50 mm. Flexural testing of the composites was conducted using a universal testing machine in three-point bending mode at a constant speed of 2.0 mm/min and a span length of 32 mm.

3. Results and discussion

3.1. Powder characterization

SEM micrographs of CuSn10 and h-BN powders are presented in Fig. 2. The morphology of the CuSn10 powders showed fine spherical particles, while the h-BN powders had a fine flake-like structure with a particle size ranging between 10 and 15 μm.

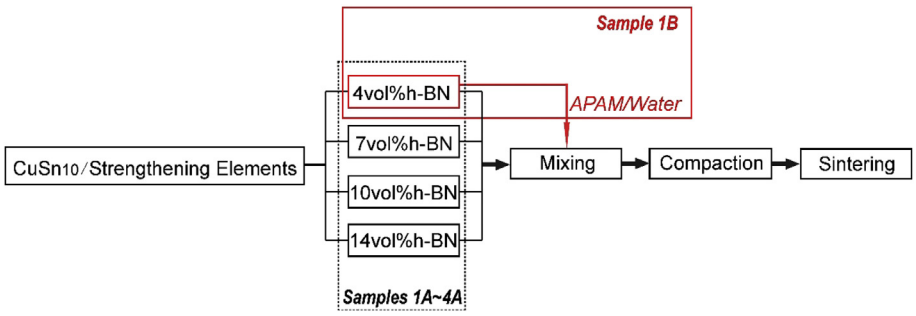

Fig. 1. Flowchart of the experimental procedure used in this study.