

Effect of ball size on nanostructured copper-tungsten carbide composite prepared by mechanical alloying

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Abstract

In this study, nanostructured copper tungsten carbide composite was produced by mechanical alloying. Mechanical alloying was carried out by milling of copper (Cu), tungsten (W) and graphite mixture at 40 h with milling speed of 400 rpm in planetary ball mill using two different ball sizes. The crystallite size of the powder milled with 10 mm ball is smaller than 20 mm ball due to higher collision frequency. Lower expansion of Cu lattice was found for the powder milled with 20 mm ball than that of 10 mm ball. The composite obtained by milling with 20 mm ball had produced greater impact energy that facilitated the formation of tungsten carbide (WC).

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1. Introduction

Copper-based composite benefits from its excellent electrical properties, thermal conductivity and good resistance to oxidation are mostly utilized in electrical application (Lee and Kim, 2004). One of the most significant electrical applications of copper-tungsten carbide composite is as a material for electrical contact.

Mechanical alloying is a rapidly developing physical technique that has been widely used to produce nanostructured copper-tungsten carbide composite. This technique involves of fracturing and rewelding of composite powder mixture in a high ball milling. Thus, the nature of milled composite powder is greatly

influenced by milling parameters such as milling time, milling speed, size of balls, the addition of process control agent and ball to powder ratio. The use of different ball diameter has generally produced different impact energy (Lü and Lai, 1998) to the material. Bigger balls which essentially have higher weight produce a great deal of impact force onto powder. According to Maurice and Courtney (1996) large balls may cause an increase in plastic work per unit collision energy. Increasing plastic work usually results in a rise of the ignition temperature, leading to combustion. In the existing literatures (Stobrawa and Rdzawski, 2007, 2009; Zhao *et al.*, 2004), little discussion is available

regarding the microstructural and structural evolution of copper tungsten carbide composite prepared by mechanical alloying. Therefore, the purpose of this study was to investigate the effect of different ball sizes on the phase formation and microstructure of nanostructured copper tungsten carbide composite.

2. Materials and method

Elemental copper (Cu; 99.8% purity; average particle size 22.3 μm), tungsten (W; 99.9% purity; average particle size 11.4 μm) and graphite powders (C; 99.8% purity; average particle size 17.0 μm) were used in this study. The powder mixtures were prepared without n-heptane with two different ball sizes (10 mm and 20 mm ball). The powder mixtures were milled for 40 h at 400 rpm speed in a planetary ball mill (Fritsch Pulverisette 5) in an argon environment with a ball-powder ratio of 10:1. The powders were then compacted at 300 MPa and sintered under argon atmosphere at 900°C for 1 hour. The characterization for milled powder and sintered composite involve of X-ray diffraction analysis and scanning electron microscopy. Crystallite size and internal strain were calculated by the Williamson-Hall method (Eq. 1) (Hesabi *et al.*, 2009):

$$B_r \cos \theta = \frac{0.89\lambda}{\langle D \rangle} + 2\eta \sin \theta \quad (1)$$

where B_r is line broadening, θ is Bragg's angle, λ is wavelength, D is crystallite size and η is internal strain. The instrumental broadening B_i was removed using Gaussian profile (Eq. 2) (Hesabi *et al.*, 2009):

$$B_r^2 = B^2 - B_i^2 \quad (2)$$

where B is the full width at half maximum. Particle size of the milled powder was determined by laser diffraction using Malvern Mastersizer 2000S. In this work, equivalent volume mean diameter, $d(4,3)$ is much preferable to incorporate particle size than the mean particle value, d_{50} since d_{50} calculation is only limited to a single size distribution (Palaniandy *et al.*, 2008). $d(4,3)$ was presented by plotting it against different ball diameter. The lattice parameters of the milled powders were obtained by Cohen's method using the method of least square.

3. Results and discussion

XRD patterns of as-milled Cu-W-C mixture

X-ray diffraction of Cu-W-C powder milled with different ball size is shown in Fig. 1. Broadening of Cu and W peaks was pronounced for the both powder milled. However, the powder milled with 20 mm ball has more broadened peaks than that of 10 mm ball indicating that 20 mm ball produce greater impact energy to the powders. WO_3 is obtained as a result of the interaction of W and available oxygen in a jar. Instead of react with W, oxygen might react with Cu. The reason is oxygen has more affinity towards W than Cu hence, more preferable to form tungsten oxide than copper-based oxide. Generally, too much cold welding may have less fracturing. In this case, the powder has been concealed by the formation of oxide which drives the diffusion distance to increase, hence impeded the atomic diffusion (Lu and Zhang, 1999). Therefore, the fracturing stage becomes difficult to be achieved for both ball sizes.

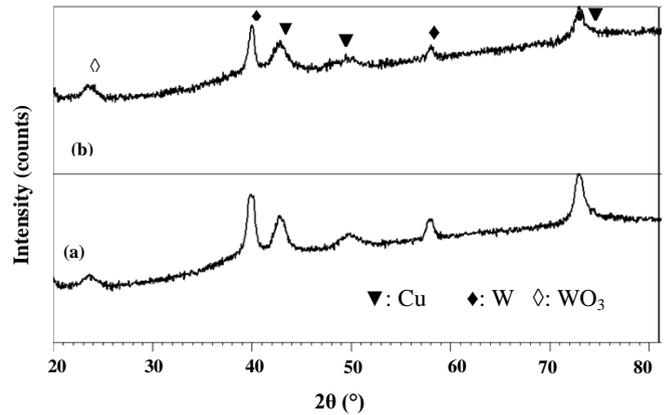


Fig. 1: XRD patterns of as-milled powder milled without n-heptane for the powder milled with (a) 10 mm and (b) 20 mm ball size

Cu crystallite size, internal strain and lattice parameter of as-milled powder

Table 1 shows the crystallite size, internal strain and lattice parameter for the powder milled with different ball sizes. The value of crystallite size for the powders milled with 10 mm ball is seen lower to that of the powder milled with 20 mm ball. The rate of plastic deformation is not critically count on the ball-to-ball or ball-to-wall collisions, which mainly determined by

subsequent kinetic energy, but also affected by severity of the powder particles (Shaw *et al.*, 2003). Without any lubricant, the particles are in the lowest distance and tend to accumulate together (Lu and Zhang, 1999) that can lead to cold welding. In mechanical alloying, cold welding may facilitate for interdiffusion between two adhered particles (Hadeif *et al.*, 2011). However, a great deal cold welding during MA may reflect to large particle size. Ball size 10 mm ball can produce enlargement of Cu lattice greater than in 20 mm ball, indicating the amount of Cu solid solution was greater for 10 mm ball.

Table 1: Cu crystallite size, internal strain and lattice parameter for as-milled powder milled with different ball sizes

Ball size (mm)	$d(4,3)$ (μm)
10	16.18
20	11.63

SEM images of as-milled powder

Fig. 2 shows SEM images of the powder milled with different ball size. The microstructure of the powder milled with 10 mm and 20 mm balls have porous and homogenous distribution with several regions consist of large unreacted W particles. The powder milled 20 mm ball presented finer distributed W particles than that of 10 mm ball since it could not be resolved under the same magnification.

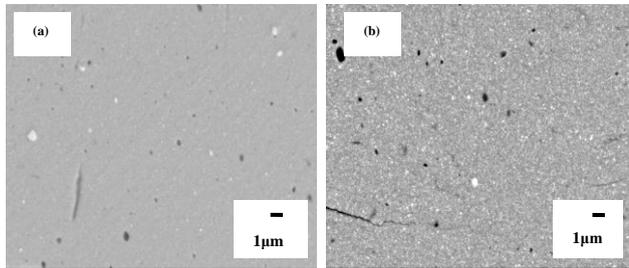


Fig. 2: SEM images of the powder milled (a) 10 mm ball and (b) 20 mm ball

Particle size

Table 2 shows volume mean diameter ($d(4,3)$) for the powder milled with different ball size. 20 mm ball has higher energy impact than 10 mm ball. It would be possible for 20 mm ball to have greater particle fracturing than 10 mm ball, hence lead to smaller

distribution of particle size. These particles were not prevented from fully touching with each other during milling and decreased the powder productivity (Liu *et al.*, 2007). At this condition, cold welding is more dominant instead of fracturing which leads to coarse particles. Therefore, lower particle size in 20 mm ball is as a result of consisted more deformed and fractured particles than that of 10 mm ball.

Table 2: $d(4,3)$ value for the powder milled with different ball size

Ball size (mm)	Crystallite size (nm)	Internal strain (%)	Lattice parameter (nm)
10	3.67	0.225	0.4076
20	4.86	0.16	0.3823

XRD patterns of sintered compact

Fig. 3 shows the sintered Cu-W-C composite with different ball sizes. There are W_2C was detected in the patterns while only WC was determined. It is suggested that without the addition of process control agent, the formation of WC becomes easier since the amount of welding was increased. This would be contributed by the atomic substitution during milling. Moreover, the formation of $\text{Fe}_3\text{W}_3\text{C}$ is derived by iron contamination from steel balls because of no addition of n-heptane to the jar. Sharper XRD peaks of tungsten carbide are found in 20 mm ball than in 10 mm ball. It is suggested that 20 mm ball has higher impact energy to result in higher grain boundary generation which promotes faster diffusion than in 10 mm ball. Smaller particle size from the powder milled with 20 mm ball than that of 10 mm ball had also increased the availability of C in Cu matrix, hence forming solid solution to be diffused into W to form WC during sintering.

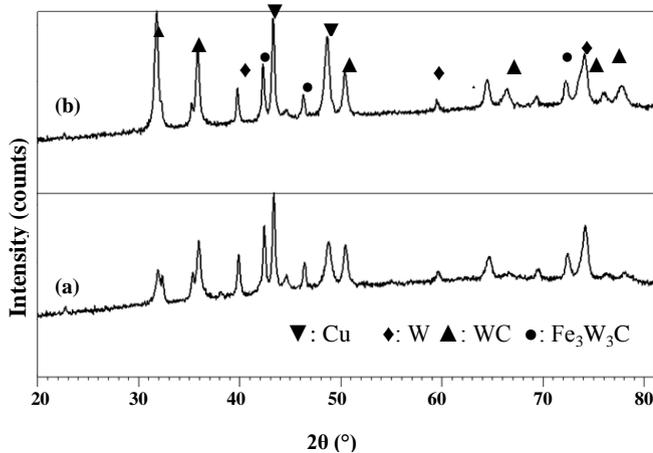


Fig. 3: XRD patterns of sintered Cu-W-C composite for the powder milled with (a) 10 mm ball and (b) 20 mm ball

4. Conclusion

Nanostructured copper tungsten carbide was successfully synthesized by mechanical alloying of an elemental Cu-W-C powder mixture. This work found that using different ball sizes broadened the peaks of Cu and W in the powder milled. 20 mm ball shows lower Cu crystallite size than 10 mm ball as a result of higher impact energy. After sintering, tungsten carbide formed greater in the composite milled with 20 mm ball than 10 mm ball.

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