# **Influence of the P/M process on the microstructure and properties of WC reinforced copper matrix composite**

NAIQIN ZHAO∗, JIAJUN LI, XIANJIN YANG

*School of Materials Science and Engineering, Tianjin University, Tianjin 300072, People's Republic of China E-mail: nqzhao@tju.edu.cn*

The influence of powder metallurgy pressing procedure on 9%WC reinforced copper matrix composite was investigated, and the fracture surfaces of the composite in different states of process were observed by scanning electron microscope. The mechanism of fracture of the composite was analyzed. The results show that the pressing process apparently influences the properties and microstructure of the composite. The properties improved with processing sequence progression and the microstructure in terms of the distribution of WC particles in the composite became more uniform. The pressing process can improve the bonding at the interface between copper and WC particles and increases the critical stress for crack extension. -<sup>C</sup> *2004 Kluwer Academic Publishers*

## **1. Introduction**

Copper alloys are widely used where high electrical or thermal conductivity is required. Resistance welding electrodes, high voltage switches, motor commutators, continuous casting molds and heat exchangers are applications of interest [1]. These applications will require unusual performance, e.g., high conductivity and ability to withstand very high brazing or joining temperatures, and excellent mechanical properties in the range of temperatures from 20 to 400◦C [2].

Traditional high strength copper alloys rely on precipitation hardening for raising their strength. But they do not offer satisfactory strengthening at high temperature although they can provide exceptional strength levels at low and intermediate temperatures. Moreover, alloy additions reduce the conductivity of copper by various amounts [3]. In recent years, with the advances in particle reinforced metal matrix composites (PMMCs), searching for new materials with high conductivity and strength has become possible. Unlike aluminium alloys, copper and its alloys have not been used popularly as matrix materials in MMCs despite the desirable properties of copper [4]. The limited work on copper based MMCs has invariably been through powder metallurgy [5–7], and other techniques, such as internal oxidation [8, 9] mechanical alloying [10], compocasting [11], internal reactive method [12], infiltration [13] etc.

The Powder metallurgy (P/M) process plays an important role in novel materials production. P/M route is ideal to prepare PMMCs because it is enabling to produce PMMCs for a wide range of demanding commercial applications. The use of P/M allows the selection of various alloys as the matrix and to adjust the reinforcement level for different applications.

Tungsten carbide, which has high hardness and stability at high temperature, can be used as a raw material for hard alloys. In the case of carbide particle reinforcements, little information is available in the literature concerning the fabrication and properties of WC particle reinforced copper based composites. In the present work, a WC reinforced copper matrix composite was produced by press-sintering of P/M for high electrical conductivity and strength application. The pressing process of the material is discussed in this work.

# **2. Experimental procedure**

Pure copper supplied as −300 mesh powder made by electrolytic process was used as the composite matrix. Tungsten carbide powder with an average particle size of 2  $\mu$ m was used as reinforcement. Fig. 1a and b shows the morphologies of the as-received pure copper powder and WC particles. It can be seen that the pure copper powder exhibits a dendritic shape. WC particles have an equiaxed shape. The dendritic shape of copper is beneficial since the 2  $\mu$ m WC particles enter between the interdendritic spaces of the copper for a uniform distribution of the reinforcement. The physical and mechanical properties of the raw materials are listed in Table I.

Pure copper powder and 9 vol% WC were blended in a milling machine for 48 h. The milling media was ceramic balls. The weight ratio of milling media to powder

TABLE I The physical and mechanical properties of the raw materials

	Density $(g/cm^3)$	Melting temperature $(^{\circ}C)$	Linear expansion coefficient $(x10^{-6}$ 1/K)	Thermal conductivity coefficient $(KJ/m \cdot h \cdot K)$	Electric conductivity $(\Omega \cdot m)^{-1} \times 10^8$	Vickers hardness (HV)	Strength (MPa)	
Powder							Tensile	Compression
Cu WC	8.96 15.8	1083 2600-2800	16.6 $5.1 - 7.2$	1384	58 5.21	$80 - 90$ 2400	216 350	$\qquad \qquad -$ 540



(a) pure copper

(b) tungsten carbide

3.0um

*Figure 1* The morphology of as-received powders.

was about 5:1. Rotation speed was 200 rpm. Ethanol was used as a process control agent. The milled powder was sieved with −200 mesh sieves to obtain −200 mesh composite powder with an average particle size of 75  $\mu$ m. The milled powders were annealed in H<sub>2</sub> at 700◦C for 1 h for the reduction of copper oxide. The powders were compacted into samples under a pressure of 700 MPa using a single-floating die in a hydraulic press. The green compacts were sintered in a vacuum furnace at  $1 \times 10^{-4}$  Torr at 950°C for 3 h (State 1). The samples were repressed under 1300 MPa (State 2) and resintered at 950◦C for 3 h. After a second repressing (State 3), the samples were rolled 64% in length at room temperature (State 4). The processing procedure is shown in Fig. 2.

The density of the green and sintered compacts was measured by the Archimedes technique, as specified in MPIF standard 42. Fracture surfaces of the samples were characterized using scanning electron microscopy (SEM). The Vickers microhardness was measured with 200 g loads.

## **3. Results and discussion**

### 3.1. The microstructures of the composite

The microstructure of WC particle reinforced copper composite after sintering, repressing and rolling are showed in Fig. 3. It can been seen that the particles of the copper matrix were sintered together after heating at 950◦C for 3 h (Fig. 3a), but some cracks and pores between the copper particles can be seen clearly and several WC reinforcements were desquamated during



*Figure 2* Processing diagram for production of WC/Cu composites.

the sample preparing, which indicates that the combination between the matrix and WC reinforcement was weak. After repressing and subsequent sintering, the interface of the copper and WC particles became closer. It can be also noticed that the porosity and grain size of the composites decreased with the processing comparing with that of State 1, as shown in Fig. 3b and c.



(a) sintered at  $950^{\circ}$ C for 3 h

(b) repressed and sintered



(c) second repressed

*Figure 3* The microstructures of the samples in different process.

After rolling for 64% (Fig. 3d), the contribution of WC particles was improved.

## 3.2. Properties of the composite

The properties of WC particle reinforced copper composite after cold press-sintering, repressing and rolling are summarized in Table II. The data indicates that the physical and mechanical properties of the composite increase with the sequence of the working processes from State 1 to State 4. After rolling, the density of the composite reached theoretical level; the electric conductivity was up to 86.2% of pure copper; the hardness was HV150, which is enhanced about 41.5% compared with

201

(d) rolled  $64%$ 

that after sintering (State 1). Significantly, the strength of the composite in State 4 almost increases 3 times from that of State 1.

# 3.3. The structure of fracture

Fig. 4 shows the variation of the tensile fracture surface of the composite at different process stages. The large pits on the fracture surface for State 1 can be seen clearly. The fracture surface characteristic shows no obvious plastic deformation occurred before the fracture, and WC particles were not distributed homogeneously. Being observed by naked eye, the fracture surface of the sample has no metallic luster. All of these

TABLE II The properties of the composite in different working stages

	Density	Electric	Hardness	Tensile	
State of process	Measured value $(g/cm3)$	Measured/theoretical $(\%)$	conductivity %IACS	(HV)	strength (MPa)
1. Sintered	8.6628	91.5	75.9	106	103
2. Repressed	9.0317	94.5	81.0	131	148
3. Second repressed	9.3310	98.1	86.2	140	214
4. Rolled $(64%)$	9.5160	$\sim$ 100	86.2	150	396



x2.00K 15.0um

(c) second repressed

*Figure 4* Tensile fracture micrographs in different process.

 $(d)$  rolled  $64%$ 

X2.00K 1

w

are attributed to brittle fracture at this stage. Then, after repressing and rolling, the characteristics of plastic deformation of the fracture surface became apparent; the pits became finer, and the dispersion of WC particles was clearly improved, as shown in Fig. 4b, c and d. Moreover, Fig. 5, depicting a micrograph of the dispersion of WC for rolled composite, indicates that WC particles can be distributed uniformly after repressing and rolling.

# 3.4. Fracture mechanism of WC/Cu composite

According to the crack theory of Griffith, cracks existing in materials can result in stress concentration, which leads to their strength decreasing. A certain size of crack has a critical stress  $\sigma_c$ . When the external force exceeds  $\sigma_c$ , the crack will extend rapidly and lead to fracture. Based on the energy balance, when deduced energy due to the crack propagation equals the increase of surface *Figure 5* SEM micrograph of WC dispersion within Cu matrix.



energy, the critical tensile stress can be obtained:

$$
\sigma_{\rm c} \approx \left(\frac{E\gamma}{c}\right)^{\frac{1}{2}}
$$

where  $E$  is the elastic modulus,  $\gamma$  represents the surface energy per unit area, *c* is the length of the crack.

The equation denotes that the critical stress of crack extension is proportional to the reciprocal square root of the crack length. Our previous research results [14–16] have shown that the pores at the interface between WC and Cu lead to a weak combination of the boundary between the matrix and the reinforcements. Hence, cracks may result from the interface pores and the fracture may initially occur at the boundary layer. This suggests that the fracture of sintered particle reinforced composite starts from the interface. The crack extends along the boundary between the matrix and reinforcements, and the matrix near the boundary would be deformed. When the external stress exceeds the fracture strength of the material, fracture occurs. A scheme of the fracture process is shown in Fig. 6.

In the results of this research, it is noticed that no obvious shrinkage occurs during sintering. Therefore, the bonding of the WC/Cu interface has no strong combination after sintering (Fig. 3a), that is, there are preexisting cracks in the composite. This is confirmed from Fig. 4a, in which the fracture surface reveals a brittle characteristic. As a result, the maximum tensile strength of the sintered composite is poor. In order to obtain high strength, the combination between matrix and reinforced particles must be improved by further pressing. The data from Table II and the microstructures from Fig. 3 indicate that the strength of the composite increases after repressing and secondly repressing as well as rolling. The pit size decreases with the stage of the processing, while the matrix fracture happens only after a large amount of plastic deformation, as shown in Fig. 4. All of these revealed that the initial cracks were reduced after every deformation. Therefore, the critical stress for crack extension increased, which results in the strength increase of the composite.

From the above results and the analysis on the fracture mechanism of the composite, the reasons of strengthening are summarized as follows:

1. The cold working process obviously improves the bonding of the interface between WC particle and copper matrix. Therefore the size of pre-existing crack is



*Figure 6* A schematic drawing of fracture process.



*Figure 7* TEM micrographs of dislocations in the matrix at WC particles.

reduced and the critical stress for crack extension increases.

2. Since copper and WC have different thermal expansion coefficients, a large number of dislocations are expected around their interface resulting in increasing strength if a strong bond exists at the interface. But if the bonding of the interface is weak, the matrix would expand freely due to the stress effect. As a result, a high-density of dislocations would not exist at the interface, thus leading to a low strength of the composite.

3. A series of cold deformations improves the homogeneity of particle distribution. Since the aggregated WC particles will be dispersed by the external force, WC will re-disperse within the matrix and get more uniform during the deformation. A TEM micrograph of WC/Cu composite after deformation is shown in Fig. 7. The pile-up of dislocations at the interface shows that WC particles resist the dislocation motion. The result confirms that WC particles strengthen the composite.

#### **4. Conclusion**

1. The properties of WC reinforced copper composite were improved by the subsequent processing after sintering. The tensile strength increases to 396 MPa after rolling 64% at ambient temperature, and the electrical conductivity is 86.2%IACS; hardness is HV150.

2. The tungsten carbide particles in WC/Cu composites distribute non-uniformly in the sintered samples and the fracture surface of the tensile sample shows a brittle characteristic. Post-repressing process enhances the dispersion of WC particle in the composites.

3. The fracture of WC/Cu composite results from the porosity existing at the interface between WC particles and copper matrix. The repressing and rolling processes can improve the bonding at the interface between copper and WC particles and increases the critical stress of crack extension.

#### **Acknowledgment**

The authors wish to thank Professor Philip Nash for his kind help and suggestion in this work.

#### **References**

- 1. A. V. NADKARNI, "High Conductivity Copper and Aluminum Alloys" (American Institute of Mining, Metallurgical, and Petroleum Engineers, Inc., New York, 1984) p. 77.
- 2. N. J. GRANT, A. LEE and M. LOU, "High Conductivity Copper and Aluminum Alloys" (American Institute of Mining, Metallurgical, and Petroleum Engineers, Inc., New York, 1984) p. 103.
- 3. P. W. TAUBENBLAT, W. E. SMITH and A. R. GRAVIANO, "High Conductivity Copper and Aluminum Alloys" (American Institute of Mining, Metallurgical, and Petroleum Engineers, 1984) p. 20.
- 4. K. PRAKASAN, S . PALANIAPPAN and S . SESHAN,*Composites Part* A **28** (1997) 1019.
- 5. S. F. MOUSTAFA, Z. ABDEL-HAMID and A. M. ABD-ELHAY, *Mater. Lett.* **53** (2002) 244.
- 6. S. OCHIAI, M. MIZHUHARA and Y. MURAKAMI, *Trans. Jpn. Metals Inst.* **41** (1977) 625.
- 7. S . C. TJONG and K. C. LAU, *Mater. Lett.* **43** (2000) 274.
- 8. ZIYUAN SHI and MAOFANG YAN, *Appl. Surf. Sci.* **134** (1998) 103.
- 9. J. L. MEIJERING, *Adv. Mater. Res.* **5** (1971) 1.
- 10. I. SALINAS , L. NUNEZ, R. PALMA and T. LOBELL, in Proceedings of Copper 95-cobre 95 International Conference, Chile, 1995, Vol. 1, p. 503.
- 11. KIYOSHI ICHIKAWA and MASAKAZU ACHIKITA, *Mater. Trans. JIM* **35** (1994) 833.
- 12. A. CHRYSANTHOU and G. ERBACCIO, *J. Mater. Sci.* **30** (1995) 6339.
- 13. N. FRAGE *et al.*, *Metal Powder Report* **57**(11) (2002) 40.
- 14. M. F. CHEN, N. Q. ZHAO and G. J. LI, Ordn. Mater. Sci. *Engng.* **21**(6) (1998) 22.
- 15. M. F. CHEN, N. Q. ZHAO and C. YOU, Mater. Mechan. *Engng.* **22**(3) (1998) 28.
- 16. N. Q. ZHAO and G. J. L I, *Ordn. Mater. Sci. Engng.* **20**(3) (1997) 39.

*Received 23 September 2003 and accepted 29 April 2004*