

Densification of W-brass Composites by Infiltration

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Abstract: *This paper describes direct infiltration of W-brass with brass and copper at both top and bottom respectively. The infiltration was carried out without pre-sintering of tungsten to obtain a porous tungsten skeleton. The green compacts were sintered in a horizontal tube furnace at the temperature of 1150°C under pure hydrogen environment with the heating rate of 5°C/min. and cooling rate of 8°C/min. The sample infiltrated at the bottom with brass has a relative sintered density of 99.9%, while the one infiltrated at the bottom with copper has relative sintered density of 99.8%. Their relative sintered densities were respectively 99.8% and 99.6% with top infiltration. The brass infiltrated sample at the top has the highest hardness value (137Hv). XRD studies revealed the presence of elemental tungsten and copper without intermetallic compounds due to lack of mutual solubility between tungsten and copper. This work implies that direct infiltration of W-brass with brass or copper is a promising and time saving method to produce W-brass composites.*

Keywords: Infiltration, Densification, W-brass, Microstructure, Tungsten

1. Introduction

Tungsten-copper and Tungsten-copper alloys are composite materials. Composite materials as classified by Roosta et al [1] are the materials engineered or naturally occurring, made from two or more constituent materials with significantly different physical or chemical properties and remain separate and distinct at both macroscopic and microscopic level within the finished structure. Because of the exceptional properties of W-Cu composites, which are a combination of high thermal and electrical conductivity of copper and low thermal expansion of tungsten, it is an appropriate candidate in a wide range of application [2]. These areas of application include electrical contact materials, thermal management device, kinetic energy projectiles [3-5], plasma facing materials (PFM) and heat sink materials used in fusion reactors [6].

W-Cu composites can be produced by the conventional solid state, super solidus and liquid phase sintering. However, solid state sintering is not recommended because the composites produced by this technique did not yield adequate properties in many areas of application. In the same manner, liquid phase sintering after mechanical alloying of elemental powders does not produce high density samples due to lack of mutual solubility of W in liquid Cu [7]. As a result of mutual insolubility between W and Cu, alloying of W and Cu is very difficult. Their equilibrium diagram indicates that the two metals are completely immiscible in both solid and liquid phases. This set back makes the development of full density via conventional alloying method impossible [8-9]. Benzhe et al [10] discussed these problems of conventional sintering (poor sinterability) to be poor wettability of W by liquid Cu, low densification and microstructural inhomogeneity. Infiltration of a porous-sintered tungsten skeleton by liquid copper is normally the most common method for producing W-Cu composites [1, 11-12]. The mechanical property of W-Cu composites is dependent on the fabrication method. In

general, W-Cu composites produced by infiltration of the W skeleton with liquid Cu demonstrate superior properties than those produced by mixing and sintering the powders [13]. Many factors were recognized to contribute to composite sinterability, they are; heating rate, heating temperature, particle size, constituent elements, sintering time and sintering environment [14].

A lot of research has been done on the densification of W-Cu composites. To the best of our knowledge, no literature is available on the infiltration of W-brass composite. In this work, to reduce the effects of dezincification, a gilding brass containing 95% copper and 5% zinc was used since the resistance to dezincification increases with copper content and brasses containing less than 15% Zn rarely dezincify [15]. The type of infiltration used in this work is direct infiltration. This is because, Zn has a tendency to evaporate at the infiltration temperature, and creating pores almost like the pores in sintered skeletal W. The inherent problem of zinc evaporation becomes an advantage here, since it excludes pre-sintering of W to form the W skeleton before liquid infiltration. The conventional sintering temperature of W to prepare the W skeleton is greater than 2000°C, to produce fully open and interconnected porosities that have the required density for infiltration [16]. Ghaderi et al [16] also noted that W-Cu composites with high W content higher than 60 wt. % are producible only by infiltration method. The direct infiltration of the green compacts saves time and high energy cost.

2. Experimental Procedure

Elemental W (99.9% purity), Cu (99.5% purity) and Zn (99.7% purity) with particle size of 12µm, 45µm and 63µm respectively were used in the experiment. The compositions of the powder mix used were 60wt. %W and 40wt. % of gilding brass. The powders were manually mixed in a glass container for about 30min. to reduce the effects of density variations. A permanent mould produced from tool steel and

a Caver mechanical press were used to produce the samples (green compacts) with diameter of 12.5mm and height of 3-4mm. 350Mpa pressure was applied and the green density was determined by weight per volume ratio. Four samples were produced, two for brass infiltration at the top and bottom while the other two was used for copper infiltration at the top and bottom also. The green samples with the brass plate at the top and bottom and another two samples with the copper plate at the top and bottom respectively were charged into a Lenton horizontal sintering furnace. The samples are heated to the temperature of 1150⁰C under a high purity hydrogen atmosphere with the flow rate of 10L/h. The heating rate and cooling rates were 5⁰C/min. and 8⁰C/min. respectively. The sintered density was determined using Archimedes' method as well as weight per volume ratio. In preparation for metallography, the samples were mounted using equal weight of hardener and epoxy. The mounted samples were allowed to set for 24hrs. Grinding of the samples was done with the aid of SiC paper from 340-2000 grit finish. Polishing was followed using alumina suspension of 1µm, 0.5µm and 0.3µm on a woven synthetic pad. The samples were rinsed with distilled water and dried with the aid of compressed air. The microstructural features were observed using a metallurgical microscope and SEM while the phases present and their compositions were determined by XRD and EDS respectively. Vicker micro hardness tester was used to determine the hardness of the infiltrated samples.

3. Results and Discussions

3.1 Relative Sintered Density

From table 1, it is observed that the difference in density between bottom and top infiltration of insignificant for both brass and copper infiltrated samples. In the process of infiltration, brass and copper were respectively melted and infiltrated into the pores created by the evaporation of zinc by capillary force until the pores were completely filled up. At the temperature of 1150⁰C, the wetting ability of liquid copper and brass respectively on the W skeleton is improved which leads to decrease in porosity and increase of relative density [12]. From a theoretical point of view, gravity difference, gradient porosity profile and capillary force contribute to the non-homogeneous distribution of the microstructure [17]. Another factor that contributes to the inhomogeneity is the manual mixing process [16]. The purpose of infiltration of composites is to obtain maximum density by filling of the pores and more densification of the parts. This means more elimination of porosity, increased hardness, electrical conductivity and density [18]. In both brass infiltrated and copper infiltrated composites, the

highest densities (99.9-99.6 T.D) was obtained. The brass infiltrated sample at the bottom gave the highest density (99.9 T.D) while the brass infiltrated at the top had almost the same density (99.8 T.D). The copper infiltrated at the top has relatively the lowest sintered density of 99.6 T.D. The brass infiltrated samples; both top and bottom have superior infiltration characteristics. This is because they have smooth surfaces and no distortion. The copper infiltrated samples both at the top and bottom are characterized with distortions and the surfaces are not smooth. The densities of the copper infiltrated samples at the top and bottom were relatively lower (99.6 and 99.8 T.D) than those infiltrated with brass. At the infiltration temperature of 1150⁰C, both liquid phase sintering and infiltration are taking place at the same time. At this temperature, the brass is already in the liquid state and surrounds the W particle forming a liquid bridge among W particles which initiate capillary forces. This capillary force reduces the diameter of the residual porosities thereby aiding infiltration [19].

Table 1: properties of W-brass composites

Composition	Relative green density (%)	Relative sintered density (%)	Vicker micro hardness (Hv)
60W-brass (brass infiltrated-bottom)	80	99.9	117
60W-brass (brass infiltrated-top)	83	99.8	137
60W-brass (copper infiltrated-bottom)	79	99.8	136
60W-brass (copper infiltrated-top)	80	99.6	114

3.2 Microstructural Characteristics

The microstructure of W-brass infiltrated (top and bottom) and copper infiltrated (top and bottom) are shown in figure 1. Fig.1 (a) shows the SEM of brass infiltration at the bottom. This figure shows the presence of minor pores in the microstructure as well as W agglomerates. The white grey portions in the SEM represent W while the dark grey portions represent brass matrix. The agglomeration in the microstructure is lesser in Fig. 1 (b). In Fig. 1 (c) and (d), there is no noticeable change in the microstructure due to top and bottom infiltration. In general, the inhomogeneity in the microstructure is as a result of density variation and immiscibility of W and Cu.

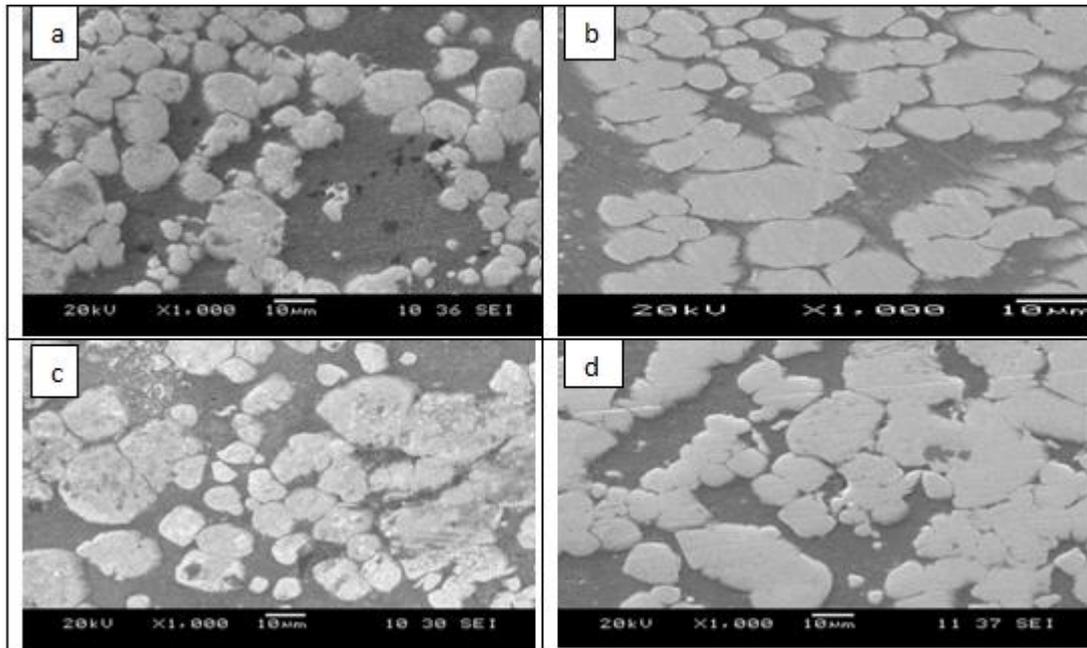


Figure 1: SEM image of infiltrated samples, (a) bottom infiltrated with brass, (b) top infiltrated with brass, (c) bottom infiltrated with copper and (d) top infiltrated with copper.

3.3 XRD Results

Fig. 2 compares the XRD patterns of the composition in both brass infiltrated and copper infiltrated (top and bottom) and at the same temperature. In brass infiltrated samples, both top and bottom, only tungsten peaks appear. This might be as a result of formation of solid solution of Cu in W. In Fig. 2 (b)- the sample that was copper infiltrated, there were both W and Cu peaks. The appearance of the Cu peak is from Cu that was used to infiltrate the samples. This shows that the intensity of Cu phase is totally dependent on its content and method of fabrication [19]. The result shows no intermetallic compound formed between the constituent elements. This is as a result of immiscibility between W and Cu, and the bond formation in this composite might be due to capillary forces and wetting of W by the liquid Cu.

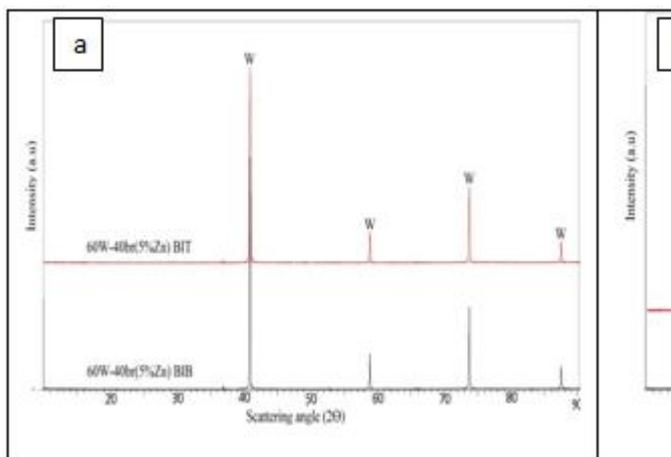


Figure 2: XRD of (a) brass infiltration-top and bottom, (b) copper infiltration-top and bottom.

3.4 Microhardness Results

The result of Vicker microhardness test is shown in Table 1. Both brass and copper infiltrated samples at the top and at

the bottom show higher value of microhardness. The high hardness is directly related to the high rate of densification. The hardness in the brass infiltrated sample (at the top) is higher (137Hv) than the copper infiltrated sample at the top (114Hv). This is equally related to the densification as the former has a relative sintered density of 98.8% while the latter has a relative sintered density of 98.6%. The hardness of the copper infiltrated sample (bottom) is higher than that of brass infiltrated (bottom). The variation in hardness and densification are so minimal. Of all the samples, brass infiltrated (top) has the highest value of microhardness (137Hv).

4. Conclusions

Nearly full density brass and copper infiltrated W-brass have been produced in this study. It was observed that after infiltration;

- Smooth surface finish was observed in both top and bottom brass infiltrated samples. On the other hand, distortions were observed on top and bottom of the copper infiltrated samples.
- In general, the brass infiltrated W-brass has the highest densification (99.9%) and hardness (137Hv).
- Brass and copper direct infiltration of W-brass composites is economical and time saving and is a promising means of W-brass production.

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References

- Roosta, M, Baharvandi, H, and Abdizade, H. An Experimental Investigation on the Fabrication of W-Cu

- Composite through Hot-press. *Int J Ind Chem* (2012) 1-6.
- [2] Abbaszadeh, A, Masoudi, A, Safabinesh, H and Takestani, M. Investigation on the Characteristics of Micro- and nano-structured W-15 wt. % Cu Composites Prepared by Powder Metallurgy Route. *Int J Refract Metals and Hard Mater* 30 (2012) 145-151.
- [3] Avijit, M, Upadhyaya, A and Agrawal, D. Effects of Heating Mode and Copper Content on the Densification of W-Cu Alloys. *Indian J Mater Sci* (2013) 1-8.
- [4] Zhang, Q, Shi, X, Yang, H and Duan, X. Microstructure and Properties of W-15Cu Alloys Prepared by Mechanical Alloying and Spark Plasma Sintering process. *J Wuhan Uni Tech Mater Sci Ed* (2008) 399-402.
- [5] Lai-Ma, L, Xiao-Yue, T, Ze-Long, L, Xiao-Yong, Z, Xiang, Z, Guang-Nan, L and Yu-Cheng, W. Sintering Behavior of W-30Cu Composite Powder, Prepared by Electroless Plating. *Int J Refract Metals and Hard Mater* 42 (2014) 51-56.
- [6] Xiaoqiao, T, Haibin, Z, Daming, D, Dong, Q, Chunfeng, H, Rongjun, X and Yi, F. Fabrication of W-Cu Functionally Graded Material by Spark Plasma Sintering Method. *Int J Refract Metals and Hard Mater* 42 (2014) 193-199.
- [7] Ozkal, B, Upadhyaya, A, Ovecoglu, M.L and German, R.M. Realtime Sintering Observation in W-Cu system: Accelerated Rearrangement Densification Via Copper Coated Tungsten Powders Approach. *Euro PM* (2004) 1-7.
- [8] Laszlo, J.K, Matthew, D.T, Bradley, R.K, Kyu, C.C and Robert, J.D. *ARL* 2465 (2001) 1-62.
- [9] Shu-dong, L, Jian-hong, Y, Ying-li, G, Yuan-dong, P, Li-ya, L and Jun-ming, R. Microwave Sintering of W-Cu Composites: Analysis of Densification and Microstructural Homogenization. *J. Alloys and Comp* 473 (2009) 15-19.
- [10] Benzhe, S, Jiupeng, S, Yang, Y, Zhigang, Z, Mingjie, N, Yan, L, Tinghui, Z and Yang, Q. Microstructural Studies of W-10wt. %Cu Composites Prepared by Using Ultrafine Composite Powder. *Int J Refract Metals and Hard mater* 45 (2014) 76-79.
- [11] Ahangarkani, M, Borgi, S, Abbaszadeh, H, Rahmani, A.A and Zangeneh-Madar, K. The Effects of Additive and Sintering Mechanism on the Microstructural Characteristics of W-40Cu Composites. *Int J Refract Metals and Hard Mater* 32 (2012) 39-44.
- [12] Lihui, D, Wensong, L, Jieli, W and Guoliang, Y. Thermal Properties of W-Cu Composites Manufactured by Copper Infiltration into Tungsten Fiber Matrix. *Int J Refract Metals and Hard Mater* 46 (2014) 96-100.
- [13] Yutaka, H, Takeshi, I, Hideaki, H and Naoyoshi, A. Ductile-to-Brittle Transition Characteristics in W-Cu Composites with Increase of Cu Content. *Mater Trans* vol 46 No.7 (2007) 1663-1670.
- [14] Mahani, Y and Zuhailawati, H. Effects of Sintering Parameters on Microstructure and Properties of Mechanically Alloyed Copper-Tungsten Carbide Composite. *Int J Mater Mec and Man* Vol 1 No.3 (2013) 283-286.
- [15] Davies, D. D, 1993. A Note on the Dezincification of Brass and the Inhibiting effects of Elemental Additions, Copper Development Association Inc, Madison Avenue, New York, pp. 1-9.
- [16] Ghaderi, A.H, Arabi, H and Rastergari, S. Tungsten-Copper Composite Production by Activated Sintering and Infiltration. *Int J Refract Metals and Hard Mater* 29 (2011) 538-541.
- [17] Yingli, G, Jianhong, Y, Shudong, L, Chengshang, Z, Lifang, C and Yuandong, P. Fabrication of W-Cu Composites by Microwave Infiltration. *J Alloys and Comp* 492 (2010) 75-78.
- [18] Daneshjou, K and Ahmadi, M. Optimizing the Effective Parameters of Tungsten – Copper Composites. *Trans. CSME/de la SCGM* 30 No3 (2006) 321-327.
- [19] Hafed, I, Azizan, A and Azmi, R. Enhanced Liquid-phase Sintering of W-Cu Composites by Liquid Infiltration. *Int. J. Refract Metals and Hard Mater* 43 (2014) 222-226.