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INFILTRATION BEHAVIOR OF MECHANICAL ALLOYED 75 wt% Cu-25 wt% WC POWDERS INTO POROUS WC COMPACTS

PROCES INFILTRACJI POROWATYCH KSZTAŁTEK WC Z WYKORZYSTANIEM PROSZKU 75% Cu-25 WC WYTWORZONEGO METODĄ MECHANICZNEGO STOPOWANIA

In this work infiltration behavior of mechanical alloyed 75 wt% Cu – 25 wt% WC powders into porous WC compacts were studied. Owing to their ductile nature, initial Cu powders were directly added to mechanical alloying batch. On the other hand initial WC powders were high energy milled prior to mechanical alloying. Contact infiltration method was selected for densification and compacts prepared from processed powders were infiltrated into porous WC bodies. After infiltration, samples were characterized via X-Ray diffraction studies and microstructural evaluation of the samples was carried out via scanning electron microscopy observations. Based on the lack of solubility between WC and Cu it was possible to keep fine WC particles in Cu melt since solution reprecipitation controlled densification is hindered. Also microstructural characterizations via scanning electron microscopy confirmed that the transport of fine WC fraction from infiltrant to porous WC skeleton can be carried out via Cu melt flow during infiltration.

Keywords: electrical contacts, WC-Cu system, mechanical alloying, contact infiltration

1. Introduction

Refractory metals and their carbides offer unique advantages based on their high temperature properties [1-3]. On the other hand they have poor electrical and thermal conductivities and they do not show resistance against oxidation. In order to compensate these drawbacks usually transition metals are added to refractory metals and their carbides leading a composite matrix [4-6]. Electrical contacts are one of the well-known application fields of these composites and it is expected to withstand high currents and more arcing than other usual contact materials [6-11]. In most applications high conductivity materials like Cu and Ag are used as filler matrix phase. Depending on the desired composition, final products can be fabricated via powder metallurgy taking advantage of penetration of filler matrix phase in liquid state.

WC-Cu system is a typical system having negligible mutual solubility. Based on the lack of solubility between WC and Cu, solution recipitation controlled densification is hindered and contribution of solid state sintering to densification is limited until Cu melts [12]. Infiltration of liquid Cu into porous WC skeleton[13-14] or liquid phase sintering of the compact pressed from the mixture of WC-Cu powders [5, 15-16] are the possible approaches and both involve liquid Cu penetration. Mechanical alloying (MA) can be applied to obtain composite powders in WC-Cu system and simultaneously enhance the solid state diffusion compared to sintering performance of initial as-blended WC-Cu powder mixtures. Al-

though mechanical alloying was applied for WC-Cu mixtures or their precursors [17-18], use of mechanical alloyed powder as infiltration material has not been studied in literature.

The aim of this work is to observe the transport of the fine WC particles through liquid Cu infiltration into porous WC powder compacts. Fine WC particles used in this study were prepared by high energy milling (HEM) of sole WC powders and these powders were added to mechanical alloying batch and their fragmentation continued inside of Cu matrix. During mechanical alloying 75 wt% Cu – 25 wt% WC (Cu25WC) powder mixture was selected as starting composition and mechanical alloying was performed against time. Then, the compacts prepared from mechanical alloyed powders were infiltrated into porous WC skeleton targeting 70 wt% WC – 30 wt% Cu (WC30Cu) final composite composition.

2. Experimental procedure

Tungsten carbide (WC) (Alfa Aesar[TM], 99.9% purity, 3.3 μm average particle size) and copper (Cu) (Alfa Aesar[TM], 99.9 % purity, 45 μm average particle size) powders were used in the present investigation. WC powders were subjected to HEM to achieve fine WC powders. For this purpose WC powders were HEM'ed using a Spex[TM]Duo Mixer/Mill 8000D at 1200 rpm up to 64 h. WC vial with WC balls having a diameter of 6.35 mm used as milling media. WC powders obtained under optimum conditions were mixed to initial Cu powders to constitute the composition Cu25WC. This mixture then MA'ed

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up to 8h. Spex^{TM}Duo Mixer/Mill 8000D and same parameters used in HEM were used and selected for MA studies. Vials were sealed inside a Plaslabs^{TM}glove box under purified Ar gas (99.995% purity) to prevent oxidation during MA. The ball-to-powder weight ratio (BPR) was selected as 10:1. Phase analyses were performed using Bruker^{TM}D8 Advance X-Ray Diffractometer (XRD) with Cu K$_{\alpha}$ radiation and crystallite sizes were characterized basis of peak shifting and Vegard's law [19]. Crystallite sizes and the retained strain of the MA'ed powders were measured by using TOPAS 3 (Bruker^{TM}AXS) software [20]. Powder particle size distributions were carried out in a Malvern^{TM}Mastersizer Laser particle size analyzer. Single action press and mold assemblies having cylindrical cross section were used to obtain compressed bodies for contact infiltration. Same compaction pressure of 150 MPa was applied to both infiltrant and to be infiltrated WC porous body. While to be infiltrated WC porous bodies pressed in the cylindrical mold having 12 mm diameter, infiltrants were pressed in the mold having 6 mm diameter to assure safe infiltration. All calculations have been made leading the target WC30Cu composition in overall composite after infiltration step. Contact infiltration of these pressed samples was performed at 1150°C for 1 hour under H$_2$ atmosphere and Ar gas was used during cooling stage. Experimental details are summarized in Fig. 1.
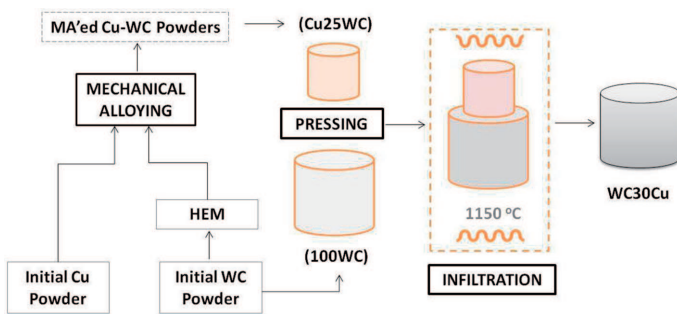

Fig. 1. Brief summary of experimental studies

3. Results and discussion

During HEM of initial WC powders a rapid decrease in mean particle sizes was observed at early milling times which is associated with the brittle nature of WC and formation of less than 1 μm sized WC particles as average was obtained even after 4 h milling. Since agglomeration tendency of the WC powders were increasing at these smaller particle sizes, no further particle size reduction was detected after 8 h milling during particle size measurement with the used instrument (Fig. 2 (a)). During MA of Cu25WC mixture Fig. 2 (b) slight and continuous reduction in mean particle sizes was observed.

The X-ray diffraction patterns belong to Cu25WC are given in Fig. 3. While simultaneous fragmentation of WC particles continues in the ductile Cu surrounding matrix of a composite powder, crystallite size of Cu matrix was also decreased and degree of deformation increased during mechanical alloying (Table 1). Because of repeated cold welding and fragmentation occurs during MA, certain decreases observed in Cu peak's intensities and peak belong to (200) plane almost merged to the peak in its neighborhood belong to WC (Fig. 3).
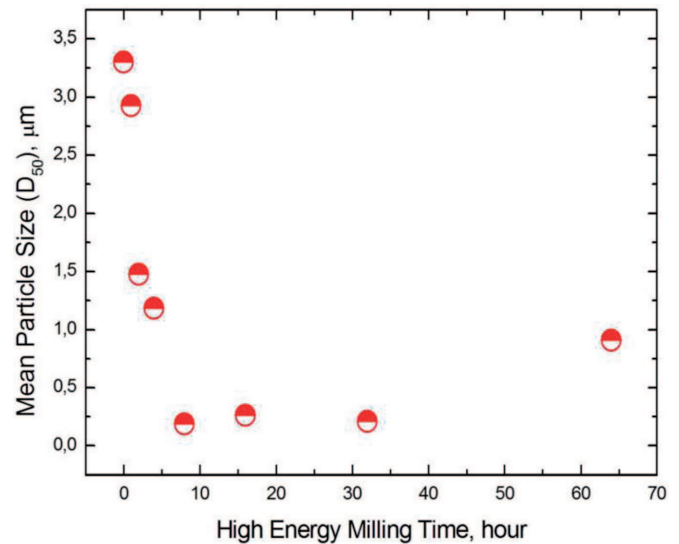
(a)
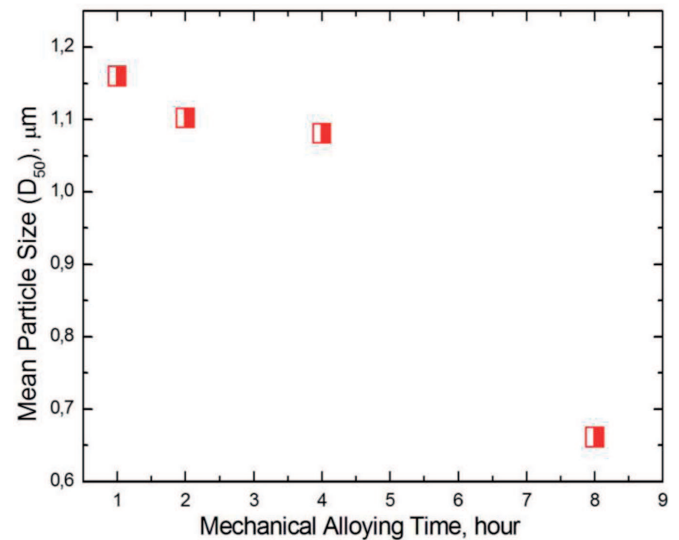
(b)

Fig. 2. Particle size of the high energy milled WC (a) and mechanical alloyed Cu – WC (b) powders
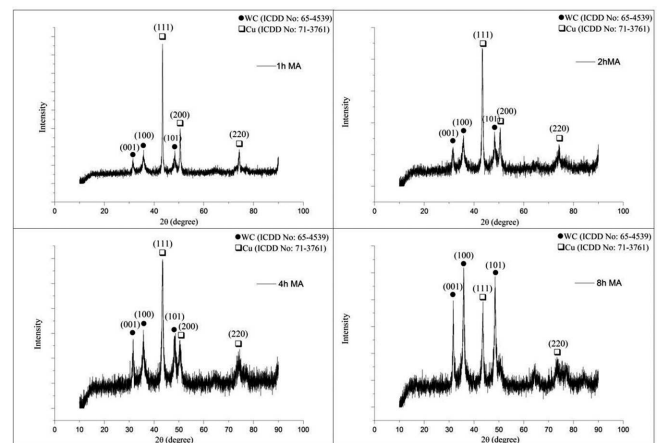

Fig. 3. X-Ray diffraction patterns of mechanical alloyed Cu and WC powders

TABLE 1

Calculated crystallite size and retained strain on MA'ed Cu25WC powders against milling time

Mechanical Alloying Time (hour)	Crystallite size (nm)		Retained Strain (%)	
Crystal plane	(200)	(111)	(200)	(111)
1	53.6	85.1	0.394	0.286
2	52.8	46	0.391	0.517
4	10.3	22.3	0.606	0.735
8	16.9	10.4	1.266	2.750

Microstructural investigations after contact infiltration of mechanical alloyed Cu25WC preform into porous WC skeleton were carried from the cross sections of metallographically prepared samples through the infiltration direction. Characteristic SEM micrographs are given in Fig. 4. As it is seen in general view Fig. 4 (a) a two phase microstructure was observed throughout the infiltration direction and clearly all voids in the WC skeleton were filled by liquid flow during contact infiltration. Fig. 4 (b) shows a detailed view and in this figure brighter WC phase (indicated as X) and darker Cu rich matrix (indicated as Y) regions can be clearly recognized. Bigger and relatively rounded particles seen in brighter tone belong to initial WC powders pressed directly to prepare porous WC skeleton. On the other hand, smaller and relatively irregular shaped particles belong to the milled WC particles which were fragmented during HEM and further MA in presence of Cu and transfered into the voids of porous skeleton by Cu melt flow during infiltration. When we focus into the matrix phase, we can see the very fine WC particles in less than 100 nm inside of the darker Cu matrix (Fig. 4 (c) and (d)). Also incomplete fragmentation of some of the WC particles which were marked by arrows in Fig. 4 (d) was noticed.

With increasing MA time, relative peak intensity ratios of Cu to WC were decreased. It is clear that ductile behavior of Cu is the important factor leading this phenomenon. As a result of agglomeration tendency of the WC powders increases at these smaller particle sizes; no certain changes were detected for further milling.

It was clearly seen that all voids in the WC skeleton throughout the infiltration direction were filled by liquid flow during contact infiltration. Relatively smaller and finer WC particles which are mechanical alloyed in presence of ductile Cu matrix and were transfered into the voids of porous WC sketeton during Cu rich melt flow. Although the density of WC is higher than the Cu melt it is remarkable that these WC nanoparticles which were produced during HEM and subsequent MA process were successfully transfered into WC skeleton without leading any aggregation. Relatively homogeneous distribution of these nanoparticles was seen throughout the microstructure. On the other hand some of the coarser WC particles were also transfered together with these nanoparticles.

Since their fragmentations were not completed during HEM and following MA proceses, they are easily recognized during microstructural investigations from their shapes showing incomplete fragmentation features.
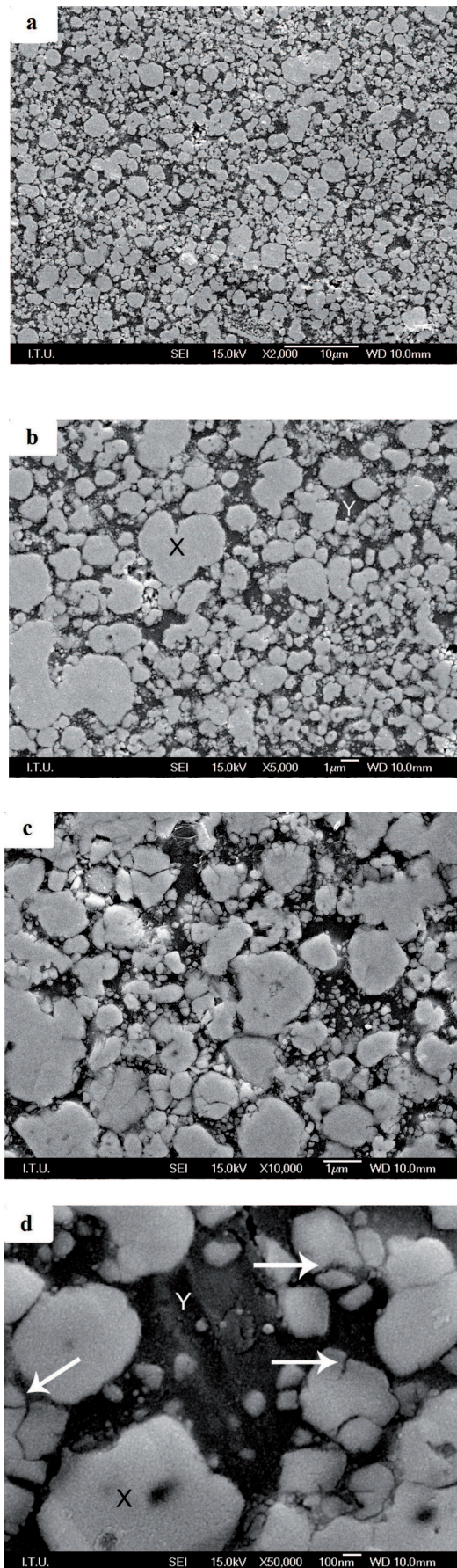
Fig. 4. SEM micrographs after infiltration microstructure (a) General view (1.000X), (b) Detailed view (10.000X) (c) Higher magnification view of the white frame given in (b) and (d) Incomplete fragmentation of WC particles (50.000X)

4. Conclusions

During infiltration of MA'ed Cu25WC mixture, fine WC particles easily transfered into WC skeleton via by liquid Cu flow. The distribution of fine WC particles in the skeleton was found much more homogeneous for mechanical alloyed samples compared to mixed state. Nanoparticle reinforcement of matrix phase plays important role for obtaining better strength materials. Although density of WC higher than the Cu, during contact infiltration process fine WC powders having nanosized fraction have been successfully transferred into voids of porous body without leading aggregation. This idea can be applied to many infiltration or impregnation related processes in where the interactions with carrier liquid phase and fine solid particles can be hindered. This can be either done by selection of immiscible systems like WC-Cu worked in this study or by selection of low melting point liquid phase compared to nature of fine solid particles.

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