Proceedings of ICXRI 2010
International Conference on X-Rays & Related Techniques in Research & Industry
June 8 – 9, 2010, Aseania Resort Langkawi, Malaysia

ICXRI 2010

Microstructure Investigations of In-Situ Copper Tungsten Carbide Composite by Mechanical Alloying

*Mahani Yusoff, Zuhailawati Hussain, Radzali Othman
School of Materials and Mineral Resources Engineering,
Engineering Campus, Universiti Sains Malaysia,
14300 Nibong Tebal, Penang, Malaysia
*email:mahaniyu@gmail.com, phone: 604-5995258, fax: 604-5941011

Abstract

An in-situ composite of copper and tungsten carbide powder was prepared by mechanical alloying of elemental powder. The sample has been milled in a high-energy ball mill for 20 h at different milling speed i.e. 100, 200, 300 and 400 rpm in an argon atmosphere. Investigations in terms of microstructural features and phase constitution of in-situ composites powder were performed by X-ray diffraction (XRD) and scanning electron microscopy (SEM). At higher milling speed, W_2C is found to be precipitated with a small amount of WC was formed. Crystallite size of copper is reducing while internal strain is increasing with increasing milling speed.

Keywords: Copper matrix composite, Mechanical alloying, In-situ processing, Carbide formation, Microstructure

I. INTRODUCTION

Particulate copper matrix composite offered high electrical conductivity due to copper itself as it showed excellent performance in electrical application but low in strength. The addition of reinforcement such as hard ceramic particles somehow increased the mechanical properties of the composite. According to Tjong et al. (2000) [1], the mechanical properties of discontinuously reinforced titanium matrix composite are mainly dependent upon the composition or microstructure of matrix, shape and volume content of reinforcement, and matrix–reinforcement interface. In this study, tungsten carbide was chosen as a reinforced material that known high in hardness, high in elastic modulus and also did very well in wear resistance. It is interest to study the copper matrix composite reinforced tungsten carbide in respect of its microstructure and phases.

This research was supported by the Science Fund Grant provided by the Ministry of Science, Technology and Innovation, Malaysia (03-01-05-SF0374) and Research University Postgraduate Research Student Grant Scheme of University Sains Malaysia (1001/PBAHAN/8041003).

Mahani Yusoff is with Universiti Sains Malaysia School of Materials and Mineral Resources Engineering, Engineering Campus, Universiti Sains Malaysia, 14300 Nibong Tebal, Penang, Malaysia (email:mahaniyu@gmail.com, phone: 604-5995258, fax: 604-5941011).

Zuhailawati Hussain is with Universiti Sains Malaysia School of Materials and Mineral Resources Engineering, Engineering Campus, Universiti Sains Malaysia, 14300 Nibong Tebal, Penang, Malaysia (email: zuhaila@eng.usm.my).

Radzali Othman is with Universiti Sains Malaysia School of Materials and Mineral Resources Engineering, Engineering Campus, Universiti Sains Malaysia, 14300 Nibong Tebal, Penang, Malaysia. He is professor in this university.

Mechanical alloying (MA) is a powder metallurgy route that has been used for a long time in synthesized a broad of materials. The impact energy produced from ball to ball and ball to wall collision makes the powder particle in non equilibrium condition as a consequence being heavily and repeatedly deformed. MA introduces a variety of defects which raise the free energy of the powder system making it possible to homogenously dispersed carbide particle in a copper matrix. The mechanically alloyed powder usually pressed and sintered to produce high quality of end product with desired

phases. Sintering of pressed composite could be conducted in an inert atmosphere at a suitable temperature to form bonding with possibly accompanied shrinkage. As reviewed so far in literatures, copper tungsten carbide composite was fabricated by chemical method by ex-situ processing. Solid state

processing by in-situ method only involve single stage of processing thus reinforced phase could be homogenously dispersed in a copper. Early work by Baikalova et al. (2000) [2] on synthesizing copper reinforced tungsten carbide only discussed on alteration of copper, tungsten, and graphite composition. The effect of milling parameters on composite microstructure has not yet been discussed in open literatures.

In this investigation, the aim was to assess the effect of milling speed on the phase constituent and microstructure of in-situ Cu-W-C composite powder synthesized by mechanical alloying. The microstructure of sintered composite is discussed.

II. EXPERIMENTAL PROCEDURE

In this study copper (99.8% purity), tungsten (99.9% purity) and graphite (99.8% purity) powder were mechanically alloyed in "Fritsch Pulverisette P-5" planetary ball mill machine. All of elemental powders were charge in a stainless steel jar with 20 mm ball in an inert atmosphere. 1% of n-heptane was added in order to minimize severe cold welding. The powders were milled for 20 h with different milling speed (100 - 400 rpm). The as-milled composites were consolidated by cold pressing in a stainless steel die with 10 mm in diameter. Sintering was conducted in an argon environment at 900°C for an hour. All composite powder and sintered samples were characterized by X-ray diffraction (XRD) and scanning electron microscope (SEM) in backscattered mode for microstructure evolution as a function of milling speed. All XRD analysis was examined using EVA software program. The crystallite size and internal strain were determined by Williamson-Hall method as shown by (1) [3]:

$$B_r \cos\theta = \frac{0.89\lambda}{< D >} + 2\eta Sin\theta \qquad (1)$$

where B_r is line broadening, θ is Bragg's angle, λ is wavelength, D is crystallite size and η is internal strain.

and the instrumental broadening, B_i was removed by Gaussian' peak shape as shown by (2) [3]:

$$B_r^{\,2} = B^2 - B_i^{\,2} \qquad (2)$$

here B is the full width at half maximum (FWHM).

III. RESULTS & DISCUSSION

Fig. 1 shows the XRD peaks profile of in-situ Cu-W-C composite powder milled with different milling speed. At lower milling speed, all charge powders are in crystalline structure and peak of Cu_2O also could be detected. Peak of Cu and W are well pronounced at low energy milling, but somehow become broaden when reaching 400 rpm of milling speed. Disappearance of graphite peak in XRD profile reveals that it has a very fine particle to be diffracted by X-rays owing enough energy is supplied with increasing of milling speed. Another possibility is graphite having a very low scattering factor between those two dominant phases and making it easily to be diffuse upon alteration of kinetic energy. One unanticipated finding was that strongest peak of W (110) is higher than Cu (111) when increased the milling speed. Possible reason is that W is difficult to deform during mechanical alloying since its atomic weight is about two times of atomic weight of Cu [4]. The oxide phase slightly decomposed with increasing of input energy as a result milling speed that has been increased. However, no carbide phase is formed during mechanical alloying process which indicates that the impact energy is insufficient to form WC.

When dealing with mechanically alloyed powder, peak broadening should be taking into account that they are highly dependable on crystallite size, internal strain and instrumental broadening. As shown in Fig. 2, crystallite size of Cu of in situ Cu-W-C composite is decreased along with increased of milling speed. At lower milling intensity, cold welding may become a dominant event since the kinetic energy is low and contribute to large crystallite size. Inversely, increasing milling speed would supply very high kinetic energy to be transferred into Cu-W-C powders that promotes the collision event in a container. Hence, lots of strain were generated when a large amount of dislocations were induced during severe deformation by high impact energy.

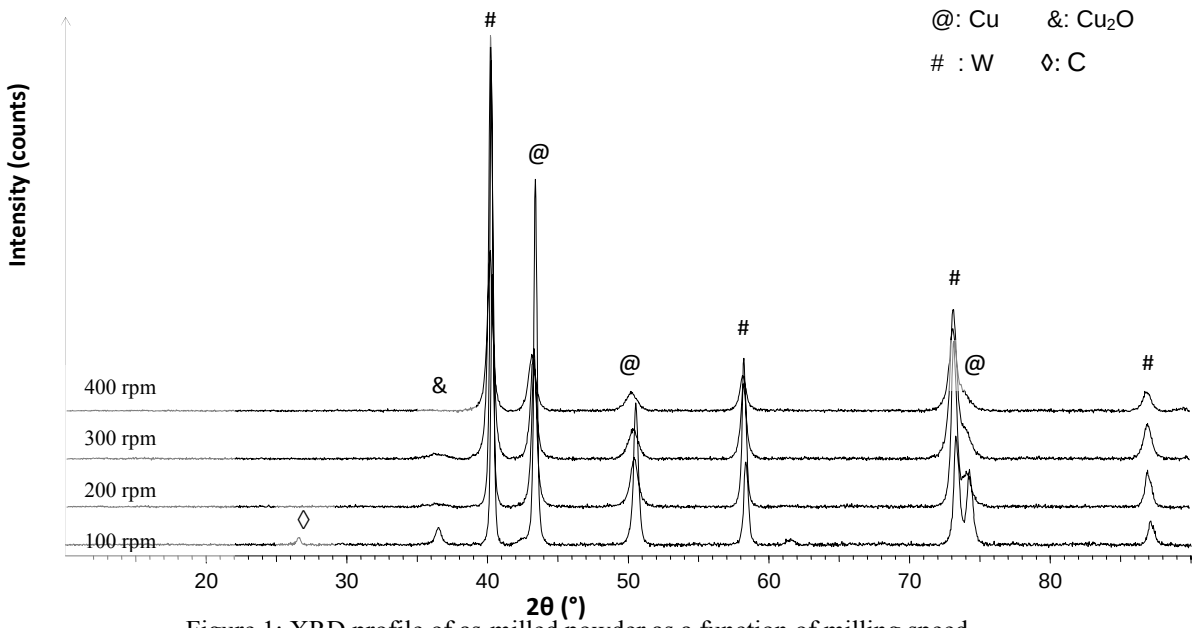

Figure 1: XRD profile of as-milled powder as a function of milling speed

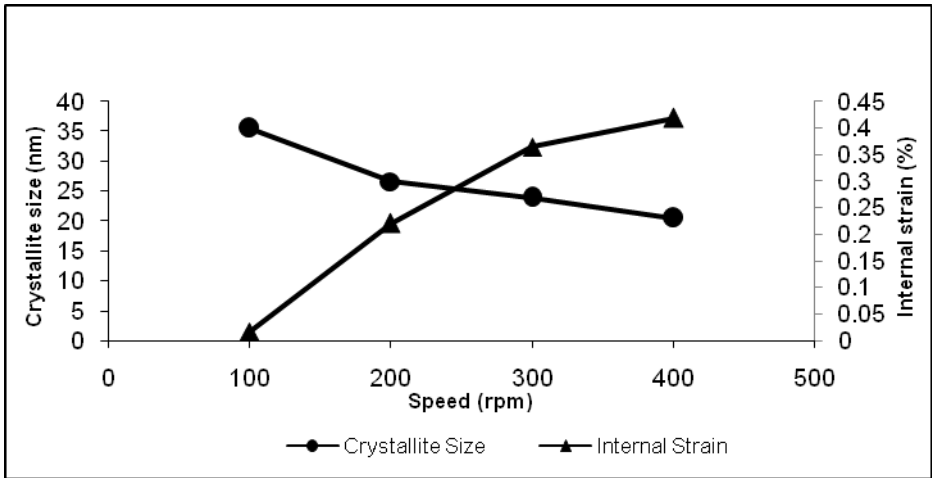

Figure 2: Cu crystallite size and internal strain as a function of milling speed of as-milled powder

Fig. 3 presents the XRD profile of sintered Cu-W-C composite as a function of milling speed. Milling at lower speed did not change the phase constituent compared to that mechanically alloyed powder. It is apparent from this figure that carbide formation only induced by heat energy associated with the presence of residual tungsten and graphite. It could be noted that W_2C first to be formed before WC formation. The reason for this is not clear but it may have something with temperature inside the vial. At low temperature, W_2C is found to be unstable whereas WC is in equilibrium state [5]. High milling speed is possible to increase the temperature due to collision between balls to ball and/or ball to wall that alter the entrapped powder. Therefore, only peaks of W_2C are found at lower milling speed. Small peak of WC only lingers at 400 rpm of milling speed. According to Balakong et al., (2010) [6] at this time, the reaction of W_2C + C to form WC is more likely preferable than that of W_2C amophization

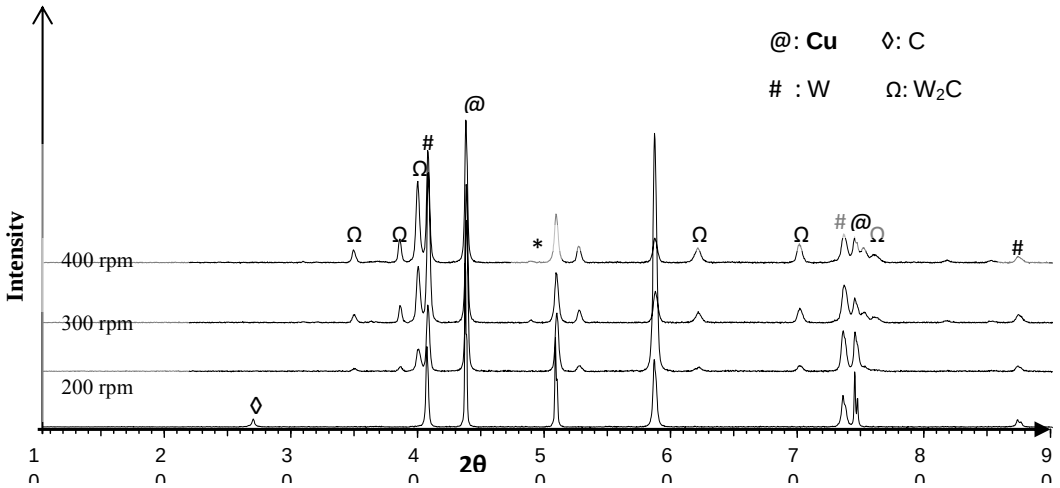

Figure 3: XRD profile of sintered pellet as a function of milling speed

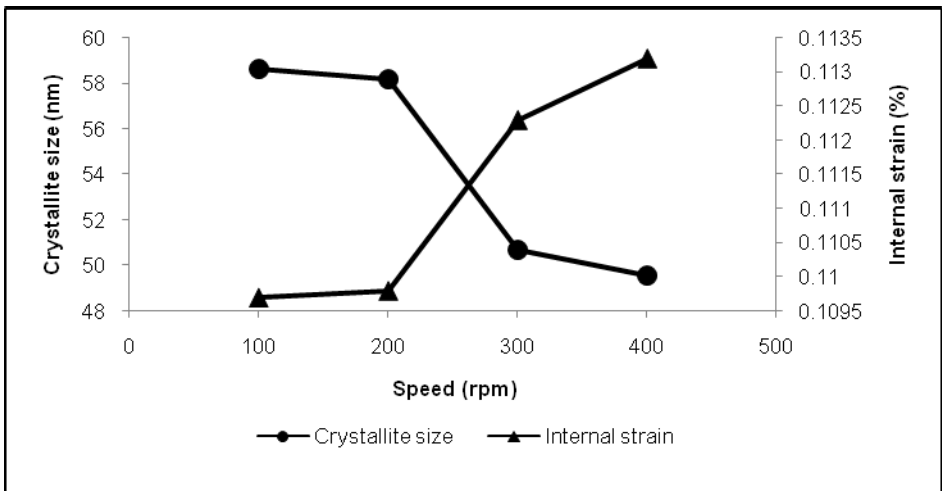

Figure 4: Cu crystallite size and internal strain as a function of milling speed of sintered pellet

The Cu crystallite size of sintered Cu-W-C composite is shown in Fig. 4. The crystallite size is found to decrease with increasing the milling speed. The internal strain of Cu in sintered pellet is slightly lower than as-milled powder as a result of stress relieving accompanied with multiple of dislocations during sintering. Fig. 5 presents the morphology of sintered Cu-W-C composite at 400 rpm of milling speed. It is difficult to obtain composite with low porosity due to serious agglomeration of mechanically alloyed powders. Breaking up the agglomerates powder is impossible during cold compaction thus generates residual porosity and voids after sintering. The dark grey region representing the copper matrix (A area) whereas white region (B area) represents rich side of tungsten and tungsten carbide. Tungsten and tungsten carbide phase are seems to be surrounded by the copper matrix which explained that tungsten particle is hardly deformed during mechanical alloying hence, influenced its diffusivity in the copper matrix even though higher impact energy is applied. However, increase the milling speed would pick up iron contamination from excessive wear of milling media and container.

.

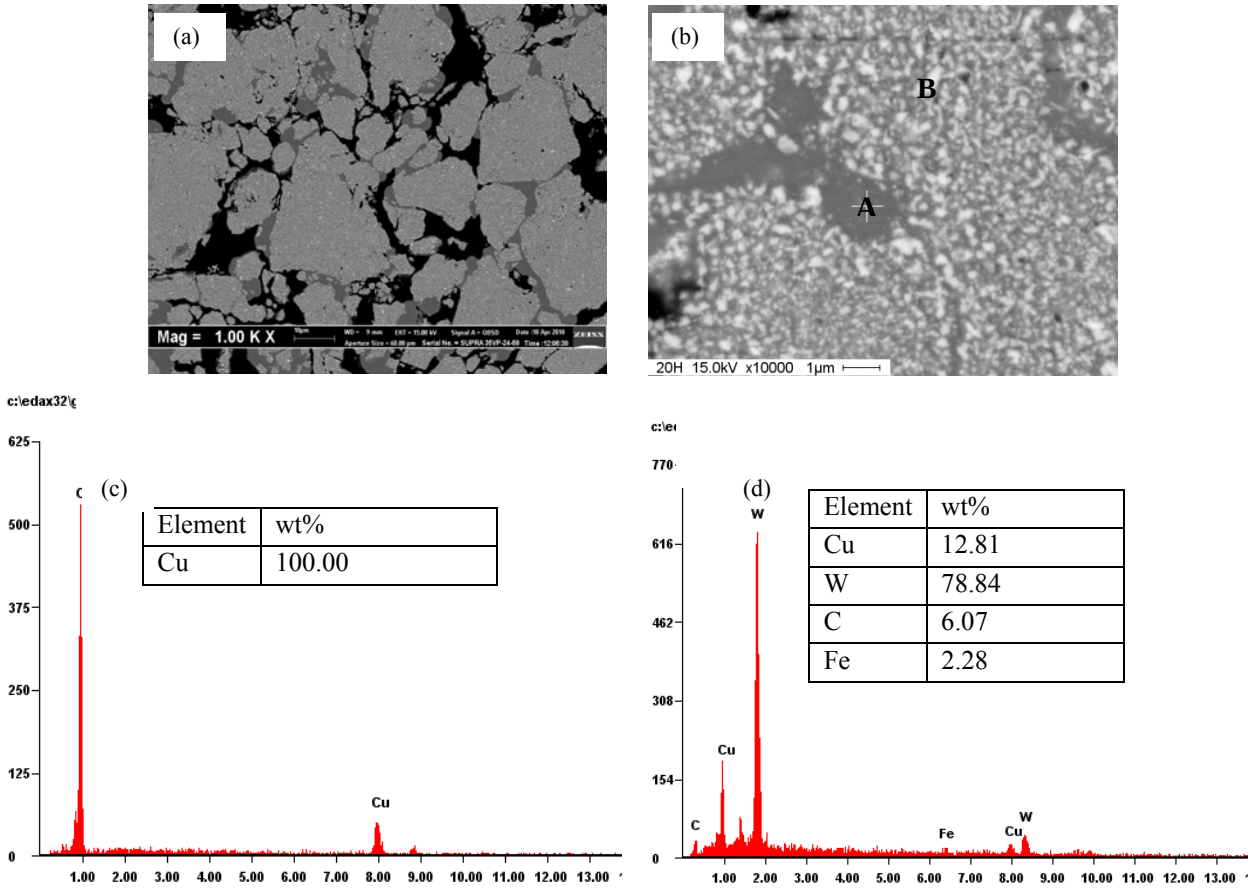

Figure 5: (a) and (b) SEM images with different magnification and corresponding X-ray energy dispersive (EDX) analysis at (c) 'A' area and (d) 'B' area of sintered Cu-W-C composite milled for 400 rpm of milling speed

IV. CONCLUSIONS

This study has found that at lower milling speed, metastable of W_2C phase was formed in sintered composite whereby WC phase only obtain after increasing the milling speed. The crystallite size of copper is decreased whereas internal strain is increased with increasing milling speed due to generating high density of defects. Heat applied during sintering convert W and C to W_2C and WC and they were found to be surrounded by copper matrix.

REFERENCES

[1] S.C. Tjong, Z.Y. Ma, "Microstructural and mechanical characteristics of in situ metal matrix composites", Materials Science and Engineering: R: Reports 29(2000), pp. 49-113.

[2] Y.V. Baikalova, O.I. Lomovsky, "Solid state synthesis of tungsten carbide in an inert copper matrix", Journal of Alloys and Compounds 297(2000), pp. 87-91.

[3] Z.R. Hesabi, S. Kamrani, A. Simchi, S.M.S. Reihani, "Effect of nanoscaled reinforcement particles on the structural evolution of aluminium powder during mechanical milling", Powder Metallurgy 52(2009), pp. 151-157.

[4] F.A. da Costa, A.G.P. da Silva, U. Umbelino Gomes, "The influence of the dispersion technique on the characteristics of the W-Cu powders and on the sintering behavior", Powder Technology 134(2003), pp. 123-132.

[5] D. Gupta, L. Seigle, "Free energies of formation of WC and W2C, and the thermodynamic properties of carbon in solid tungsten, Metallurgical and Materials Transactions A 6(1975), pp. 1939-1944.

[6] S. Bolokang, C. Banganayi, M. Phasha, "Effect of C and milling parameters on the synthesis of WC powders by mechanical alloying", International Journal of Refractory Metals and Hard Materials 28(2010), pp. 211-216.