

# Behavior of Cu-WC-Ti Metal Composite After using Planetary Ball Milling

A.T.Z. Mahamat, A.M. A Rani, Patthi Husain

**Abstract**—Copper based composites reinforced with WC and Ti particles were prepared using planetary ball-mill. The experiment was designed by using Taguchi technique and milling was carried out in an air for several hours. The powder was characterized before and after milling using the SEM, TEM and X-ray for microstructure and for possible new phases. Microstructures show that milled particles size and reduction in particle size depend on many parameters. The distance  $d$  between planes of atoms estimated from X-ray powder diffraction data and TEM image. X-ray diffraction patterns of the milled powder did not show clearly any new peak or energy shift, but the TEM images show a significant change in crystalline structure of copper on titanium in the composites.

**Keywords**—ball milling, microstructures, titanium, tungsten carbides, X-ray

## I. INTRODUCTION

**S**YNTHESIZING of especial material that can combine high electrical conductivity, thermal conductivity and high melting point and good wear resistance reacquire a special technique. Mechanical alloying using ball milling is one of the most promise techniques to synthesize an alloy from material that are immiscible or when the melting temperature vibration of the component is very high such as copper and tungsten. Copper-tungsten exhibits total absence of solubility in both solid and liquid state. Mechanical alloying as a solid state, non-equilibrium process can be beneficial to the processing of such an immiscible system with the added features of refinement of structure. Raghu, et al [1] synthesize an ultrafine Cu-W microcomposite structure by mechanical alloying using ball milling. The milling behavior was found to depend on the composition, milling time and milling atmosphere. Li, et al [2] applied the thermo-mechanical method to produce W-Cu composite powder by applying high temperature oxidation, short time high-energy milling and reduction. The results show that "the oxygen content of W-Cu composite powder decreases with the increase of milling time, while the specific surface of final powder increases with the milling time". Baikalova and Lomovsky [3] investigate Cu-W-C system using high-energy ball milling and then annealing at 820-940°C in an argon atmosphere.

The results of the milled Copper matrix with the composition of W50C50 show that with high tungsten content. W2C was obtained as the dominating product. Meanwhile, Low tungsten content led to the enhancement of WC in the products. Grain size of carbides and copper was less than 0.5  $\mu$ m.

Many other nanocomposite material was synthesized using high energy ball milling. An amorphous of Cu-Ti and Cu-Zr was successfully synthesized by Hen [4]. The structure and thermal behavior of these alloys show that the replacement of Ti by Zr will enhance the amorphization rate. "These may be attributed to a more negative mixing enthalpy by adding zirconium to the Cu-Ti system, providing a larger chemical driving force of solid state reaction". Šebo, et al [5] also studied the influence of Ti and Zr in Cu matrix on the shear strength between Cu-Ti and Cu-Zr alloys and carbon rod (or fiber). The results showed that the interface between carbon fiber with Cu-Ti alloys and Cu-Zr alloys can be controlled by controlling the amount of titanium and zirconium in Cu-Ti and Cu-Zr alloys.

Al-Cu-WC system mechanically alloyed in a high energy milled for 1 h, 2 h, 3 h and 5 h milling duration by [6]. The mechanical alloyed powder were sintered under inert Ar atmosphere at 650 °C for 4 h. The 5 h milled powder showed that the decomposition of WC after 450 °C led to the formation of Al12W intermetallic, which provided high strength composites.

Feng, et al [7] mention that a decrease in the ball milling time will improve the efficiency of mechanical alloying, and prevent the formation of amorphous phase. Meanwhile, the compressibility and sintering behavior is directly related to the properties of milled powder. Gan and Gu [8] studied the relation between powder morphology of the milled powder and compressibility of Cu/SiCp under cold compaction. The results showed that the density of composites decrease with increasing milling time. With increasing milling time, the dendrite copper powder becomes flattened and then changed to spherical composite powder. The morphology and hardening effects during ball milling have important influence on compressibility of the powders".

This research work focused on synthesizing and characterization of a new EDM tool electrode material prepared by using planetary ball milling.

## II. METHODOLOGY

Powder was being characterized before and after milling to reveal the effect of milling variables on the powder characteristics by using SEM, TEM, and X-ray. Milling experiment was designed based on L8 Taguchi orthogonal

A.T.Z. Mahamat is with Mechanical Engineering Department Universiti Teknologi Petronas, Malaysia e-mail: alti33@yahoo.com

A. M. A. Rani is with Mechanical Engineering Department Universiti Teknologi Petronas, Malaysia (e-mail: majdi@petronas.com.my)

Patthi Hussain is with Mechanical Engineering Department Universiti Teknologi Petronas, Malaysia (patthi\_hussain@petronas.com.my)

array as shown in table 1. The selected variables are milling time, tungsten carbide weight present and the titanium weight present. The experiment was conducted using planetary ball milling in a dry air and the ball to powder mass ratios fixed at 10:1. The milled powder characterized using the same equipment. Powder was characterized for as milled particles size, Particle size, particles shape and for any possible new metastable phase.

In order to study and analyze the effect of milling variables (milling time, tungsten carbide content and titanium weight present) on the as milled powder homogenization, as milled particles size and final particle size achieved by milling process, the following three equations used. The total sum of square of all variables is calculated by using equation 1, the total sum of software and the percentage contribution of each variable calculated by using equation 2 and 3 respectively.[9].

$$SS = \frac{k}{N * n} \sum_{i=1}^k T_i^2 - \frac{T_t^2}{N * n} \tag{1}$$

$$SS_t = SS_{IP} + SS_{ON} + SS_{OFF} \dots \dots SS_{error} \tag{2}$$

$$\frac{SS_{IP}}{SS_t} + \frac{SS_{ON}}{SS_t} + \frac{SS_{OFF}}{SS_t} + \dots \dots \frac{SS_{error}}{SS_t} = 100\% \tag{3}$$

Where: The parameters levels is k, N is the total number of run, n the number of replication, T<sub>i</sub> is the total sum of response at ith level and T<sub>t</sub> is the total sum of responses.

TABLE I  
L8 TAGUCHI ORTHOGONAL ARRAY DESIGN AND RESPONSES

Milling Time(h)	WC (Wt. %)	Ti (Wt. %)	milled particles size (µm)	nanosize particle (nm)
5	20	5	200	150
5	20	5	100	70
5	40	10	80	120
5	40	10	100	110
10	20	10	110	50
10	20	10	170	150
10	40	5	140	90
10	40	5	60	250

III. PROCEDURE FOR PAPER SUBMISSION

A. Effects of milling time on as milled particle size and nanoparticle size achieved.

Figure 1 shows that the most effective variables on the milled powder particles size is tungsten carbide and titanium. However, the contribution of milling time on as milled particles size is negligible. The as milled particle size decreases by increasing tungsten carbide and titanium. This can be related to the increase in brittleness and the reduction in copper content. During milling, copper encloses the small tungsten carbide and titanium by elastic deformation and cold welding leading to a bigger particle size. This point is clear in SEM images.

Figure 2 shows the effect of milling time becomes the dominant parameter in particle size reduction followed by tungsten carbide content. This is an expected result because the energy source of particle size reduction is the impact energy of milling. This energy depends mainly on rotation

speed and milling time. The high contribution of tungsten carbide can be referred to the hardness of tungsten carbide particles.

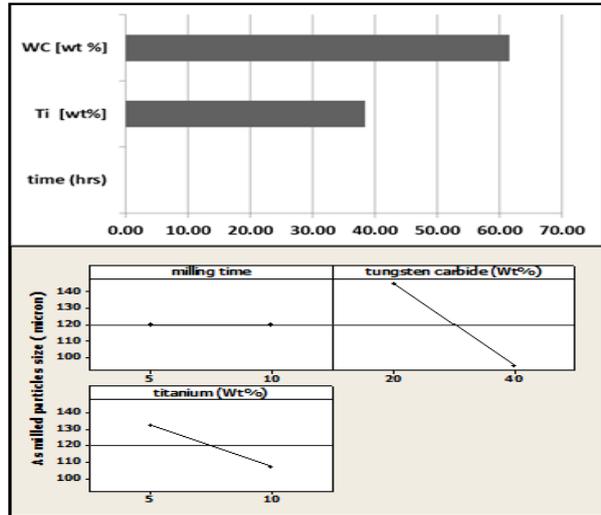


Fig. 1 Percentage contribution and main effects of variables on “as milled powder” as milled particles size

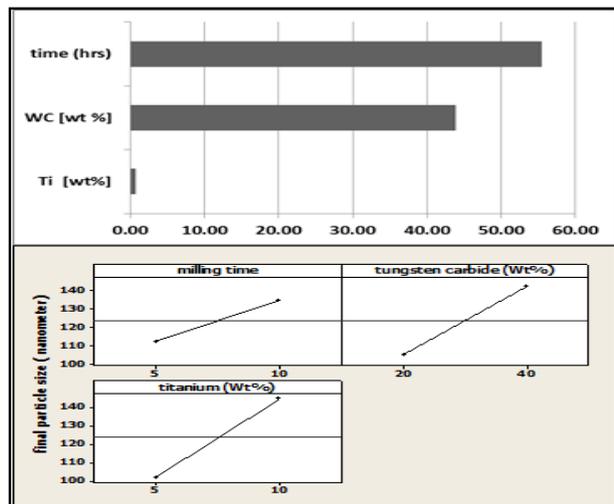


Fig. 2 Percentage contribution and main effects of variables on particle size reduction

B. Microstructure of Cu-WC-Ti composite

Figure 3 (A, B and C) show the SEM images of the as received copper, tungsten carbide on titanium powders. The average particle size is 5µm for the rounded copper, 10µm for the agglomerated tungsten carbide (less than 1µm for a single particle and 20 µm titanium. Agglomeration of tungsten carbide is due to the high surface energy of small particles

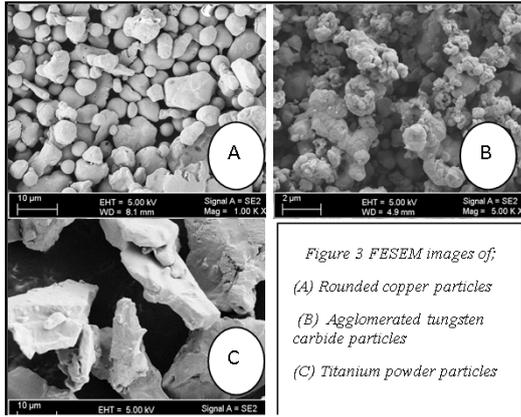


Fig. 3 SEM imaged of (A) copper, (B) tungsten carbide and (C) titanium powders

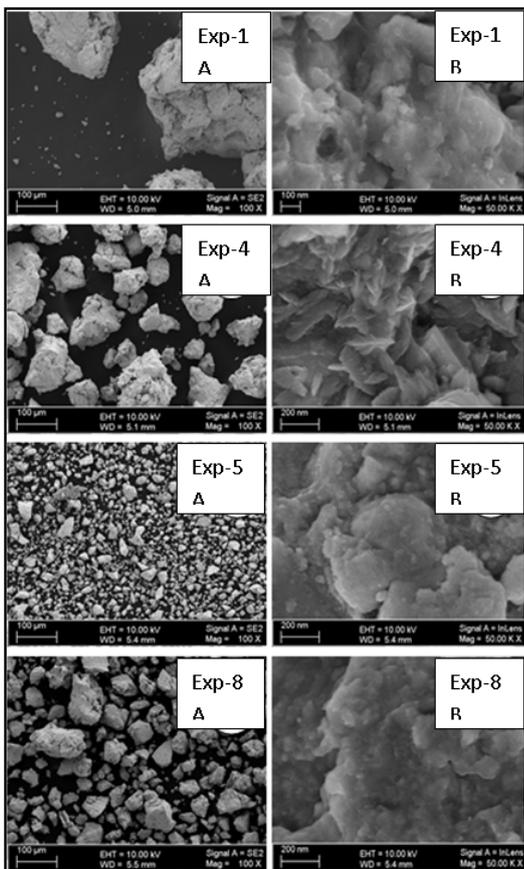


Fig. 4 SEM imaged of EXP-1, EXP-4, EXP-5 and EXP-8 of the “as milled powder” at (100-50000) magnification

Figure 4 is the SEM of the as milled powder at 0.1k and 50k magnifications. The low magnification images taken to show the effect of milling variables on as milled particles size and higher magnification images taken to show the effect of milling variables on particle size reduction and homogenization. The results show that as milled particles size not only depends on copper content as mentioned earlier but also on milling time.

The five hours milled particles size of Experiments 1(A)

and 4(A) is different because of a difference amount of copper (ductile) and tungsten carbide (brittle) contents.

Same phenomenon can be seen from images of experiments 5- 8 (A). Generally, as milling time increase the as milled particles size decrease. The increase of the tungsten carbide amount is due to the erosion of ball and bowl.

On the other hand, the higher magnification images (part B) show that 5 and 10 hours milling time achieve the perfect homogenization. The reduction of particle size by the ball milling process is a function of milling time, milling energy. For all experiments under all conditions, the particle size reductions achieved are in the range of 100 nanometre. This result shows that the reduction in particle size by ball milling is limited by ball size and components. See [10]

*C. X-Ray analysis*

Figure 5 below show the x-ray data profile of the received powder and the ten hours milled Cu+20% WC+10%Ti after milling ( Exp-5). After milling, most of the low intensity peaks disappear and FWHM increases. This distortion can be explained by the effect of perfect homogenization in nanosize and the formation of amorphous structure. At this level, the interference of elemental peaks and the background will increase the error during estimation the thickness of crystal.

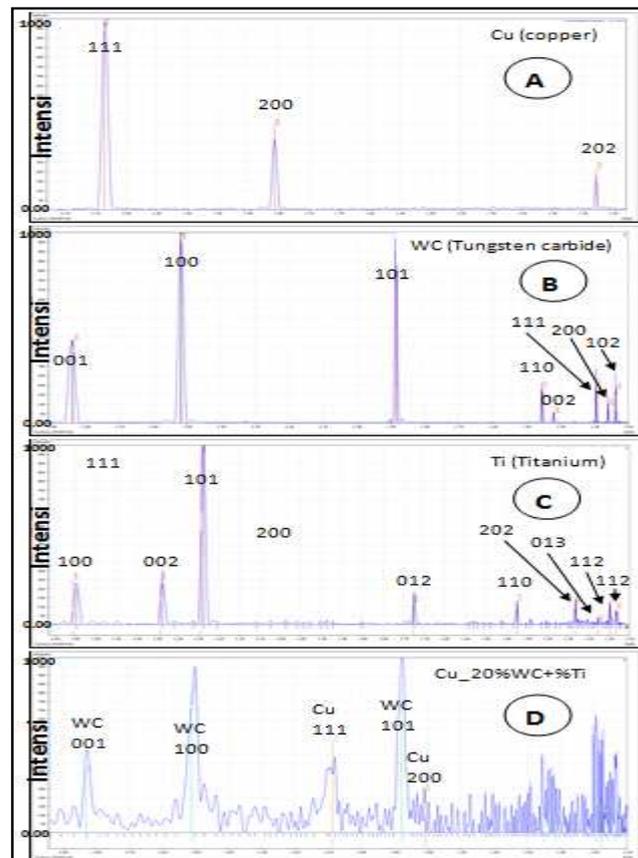


Fig. 5 X-Ray diffraction pattern of the (A) copper, (B) tungsten carbide, (C) titanium, and (D) experiment five

*D.TEM micrograph*

The periodical structure included in TEM micrograph of experiment 5 is limited to localized points, and the structure becomes more amorphous. The d-spacing after milling for 10-hour ranges from 5 to 14 Å. This variation can be referred to the stored energy of the amorphous, and some metastable phase is created. This is agreed with the work done by alam [11].

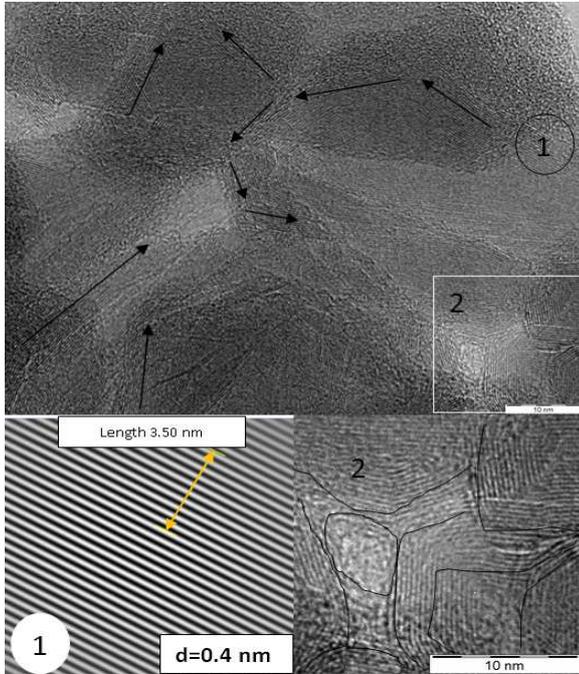


Fig. 6 TEM micrograph of "as received" Cu powder

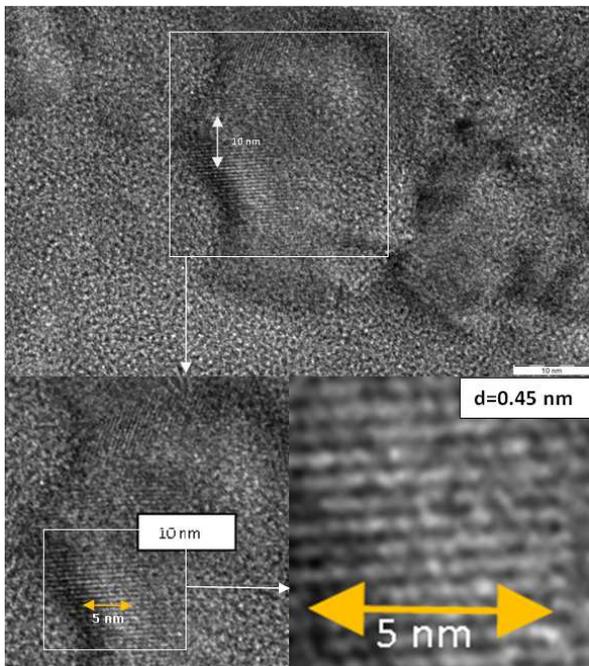


Fig. 7 TEM micrograph of as-received tungsten carbide powder

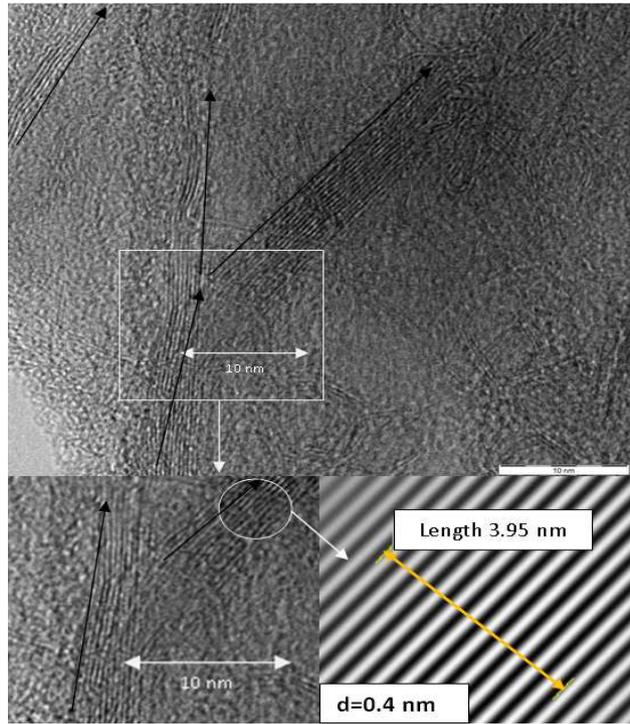


Fig. 8 TEM micrograph of "as received" titanium powder

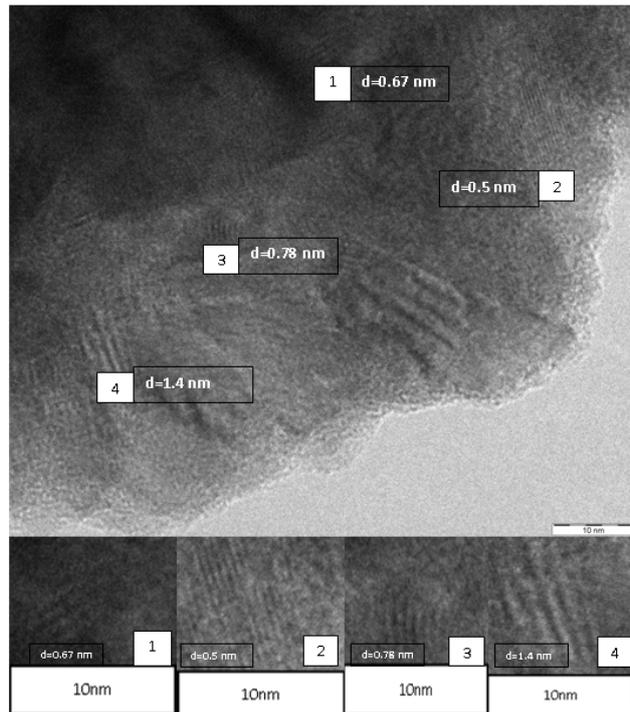


Fig. 9 TEM micrograph of "as milled" Cu-20% WC-5%Ti. Milled for 10 hours in closed air

#### IV. CONCLUSION

The effects of milling condition on the microstructure of Cu-based composites reinforced with WC and Ti particles prepared by ball milling were studied and the following points can be concluded.

Milled particles size mainly depended on composition especially the ductile copper. Meanwhile, particle size reduction not only depends on milling time but also depends on the mechanical properties of the component.

SEM analysis confirms the results concluded from the statistical analysis also reveal that the final particle size achieved by five and ten hours milling time is same. more copper content leads to bigger milled particles size and longer milling time leads to the small as milled particles size.

X-Ray analysis show that after milling, most of the low-intensity peaks of copper, tungsten carbide and all titanium peaks disappear. The peaks detected for copper and tungsten carbide didn't show any energy shifts.

TEM analysis revealed that the final structure shows amorphous structure than crystalline structure. Little periodical structure appears inside the amorphous structure, but the periodicity is different from that of the elemental powder. This can be referred to the effect of stored energy and also to the presence of metastable phase.

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