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STUDIES ON ALUMINA DISPERSION-STRENGTHENED COPPER COMPOSITES THROUGH BALL MILLING AND MECHANICAL ALLOYING METHOD

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Abstract. Oxide dispersion-strengthened copper has the ability to retain most of its properties at elevated temperatures. Among various processes, powder metallurgy route is ideal because of its efficiency in dispersing fine oxide particles. In this study, copper-alumina composites is produced through powder metallurgy route whereby copper powder, which is the matrix, was mixed with alumina powder, which act as reinforcement. Powder mixtures with different compositions of alumina (2.5wt%, 5wt%, 7.5wt% and 10wt%) were prepared. The mixtures were then mixed either by (a) blending process for 45 minutes in a ball mill or (b) mechanical alloying for 45 minutes in a planetary mill. The mixture was then compacted at 200 MPa and sintered under argon atmosphere at 950°C for 1 hour. Results showed that mechanical alloying has produced Cu-Al_2O_3 composite with better hardness and lower electrical conductivity compared to those prepared by ball milling method.

Keywords: Copper-alumina composites, mechanical alloying, ball milling, electrical conductivity

Abstrak. Kuprum diperkuat-serakan oksida berkemampuan untuk mengekalkan sifat-sifatnya pada suhu tinggi. Di antara pelbagai proses, kaedah penghasilan serbuk logam adalah terbaik disebabkan oleh kecekapannya dalam penyebaran partikel oksida yang halus. Dalam kajian ini, komposit kuprum-alumina dihasilkan menerusi kaedah serbuk logam yang mana serbuk matriks kuprum ditambah kepada alumina yang berfungsi sebagai penguat. Campuran serbuk dengan komposisi alumina yang berbeza telah disediakan (2.5% berat, 5% berat, 7.5% berat dan 10% berat). Campuran tersebut kemudiannya dicampur melalui dua kaedah berbeza samada: (a) proses percampuran selama 45 minit dengan pengisar bebola atau (b) proses pengaloian mekanikal selama 45 minit dalam pengisar planet. Ini diikuti dengan penekanan campuran serbuk pada 200 MPa dan persinteran dalam argon pada 950°C selama 1 jam. Keputusan menunjukkan kaedah pengaloian mekanikal telah menghasilkan komposit Cu-Al_2O_3 dengan kekerasan yang lebih tinggi dan kekonduksian elektrik yang lebih rendah berbanding kaedah pengisaran bebola.

Kata kunci: Komposit kuprum-alumina, pengaloian mekanik, pengisaran bebola, kekonduksian elektrik

1.0 INTRODUCTION

Oxide dispersion-strengthened copper has been identified as an effective method to increase copper strength without seriously decreasing its electrical and thermal

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conductivity. Besides high electrical and thermal conductivities, oxide dispersion-strengthened copper also retains its resistance to softening at high temperatures, which is important in electrical applications at elevated temperatures such as resistant welding electrodes, electric motors, high performance switches, and coil material for generating high magnetic field [1]. In this work, alumina has been chosen as the dispersoid due to its good properties such as high toughness and hardness with low bulk density. Furthermore, it is cheaper compared with other ceramic and metallic dispersoids and thermally stable without interaction with the matrix at high temperatures.

Due to the presence of alumina particles with high melting point, conventional melting and casting is not suitable for producing dispersion-strengthened copper composites [2]. Alternatively, powder metallurgy ensures the fine alumina dispersoid is well distributed within the copper matrix, which eventually gives good final mechanical properties to the composite with sufficient physical properties. This paper reports the properties of Cu-Al_2O_3 composites prepared via powder metallurgy with different powder mixing methods, which are by powder blending in a ball mill and by mechanical alloying in a planetary mill.

2.0 MATERIALS AND METHODS

In this study, copper powder of 23.96 μm in size and alumina powder of 0.65 μm in size were used as raw materials in preparing the copper-alumina composite. The composition of the composites was based on the weight percentage of alumina, which are 2.5, 5, 7.5 and 10wt%. The mixtures were then dry milled in a plastic bottle, which contain 20 mm diameter milling balls, by using a ball mill with velocity of 50 rpm. Mixtures of similar composition were also mechanically alloyed in a ceramic jar by using a planetary mill. Ceramic balls of 20, 10 and 5 mm diameter were used as the milling medium. The resultant powders are analyzed in order to determine their particle sizes. Then, the mixtures were compacted into cylindrical pellets at pressure of 200 MPa and green density of the pellet samples was determined according to Equation (1). The green density value was then compared with the theoretical density of each powder mixture and reported as percentage of theoretical density, as shown in Equation (2). The theoretical density of each mixture was calculated according to the rule of mixtures (Equation (3)), by considering the fully dense values for copper and alumina as 8.94 g/cm^3 and 3.9 g/cm^3, respectively.

$$\rho_g = \frac{m}{v} \tag{1}$$

$$\text{Density (\% Theoretical)} = \frac{\rho_g}{\rho_T} \times 100\% \tag{2}$$

$$\rho_T = (\rho_{Cu} \times vol\%Cu) + (\rho_{Al203} \times vol\%Al_2O_3) \tag{3}$$

where ρ_g is the green density of sample, ρ_T is the theoretical density of the composite, ρ_{Cu} and ρ_{Al2O3} are the theoretical densities of copper and alumina, m is the weight of sample, v is the volume of the sample, vol% Cu and vol% Al_2O_3 are the volume percentages of copper and alumina.

Then the pellet samples were sintered at 950°C for 1 hour in argon atmosphere. The heating and cooling rates of the sintering process were 5°C/min and 10°C/min, respectively. The density of the sintered pellets was determined by using Pycnometer Density equipment, which works based on the principle of gas displacement. The measured density values were compared with the theoretical density of each composition and reported as the percentage of theoretical density.

A hardness test was carried out on the pellet samples in order to determine the microhardness of the composite while electrical conductivity test was performed using a Kelvin-Bridge type circuit. Electrical conductivity of each sample was then compared with the value of standard electrical conductivity, which is given by copper compact of industrial grade and reported as percentage of standard conductivity as shown in Equation (4);

$$\text{Electrical conductivity} = \frac{\sigma_s}{\sigma_t} \times 100\% \qquad (4)$$

where σ_s is the electrical conductivity of the tested sample and σ_t is the electrical conductivity of the standard copper. Microstructure study was carried out on the unetched and etched samples using optical and scanning electron microscopes. The etchant used to reveal the composite microstructures was 5 g ferric chloride, 2 ml HCl and 98 ml ethanol solution.

3.0 RESULTS

Figure 1 shows the average particle size for both types of Cu-Al_2O_3 powders prepared by blending with ball mill and mechanical alloying with different alumina content. It is evident that mechanical alloying imparts significant particle size reduction compared to the ball milling process. It is also worth noting that the average particle size for the Cu-Al_2O_3 powders decreases with an increase in alumina content (from 2.5 to 10wt%) for both mixing conditions. Results show that the particle size of the mixture for both mixing methods is lower than the average particle size of as-received copper powder (23.96 µm).

Figures 2 and 3 show, in general the green and sintered densities of the samples prepared by ball milling are lower than the values given by mechanical alloying samples. In addition, it is worth noting that the densities of sintered samples are greater than the densities of unsintered samples. With an increase in alumina content, the green density and sintered density show a gradual reduction.

The microhardness of composite produced from ball milled powder are lower than the microhardness of mechanical alloying compact (Figure 3). For mechanical

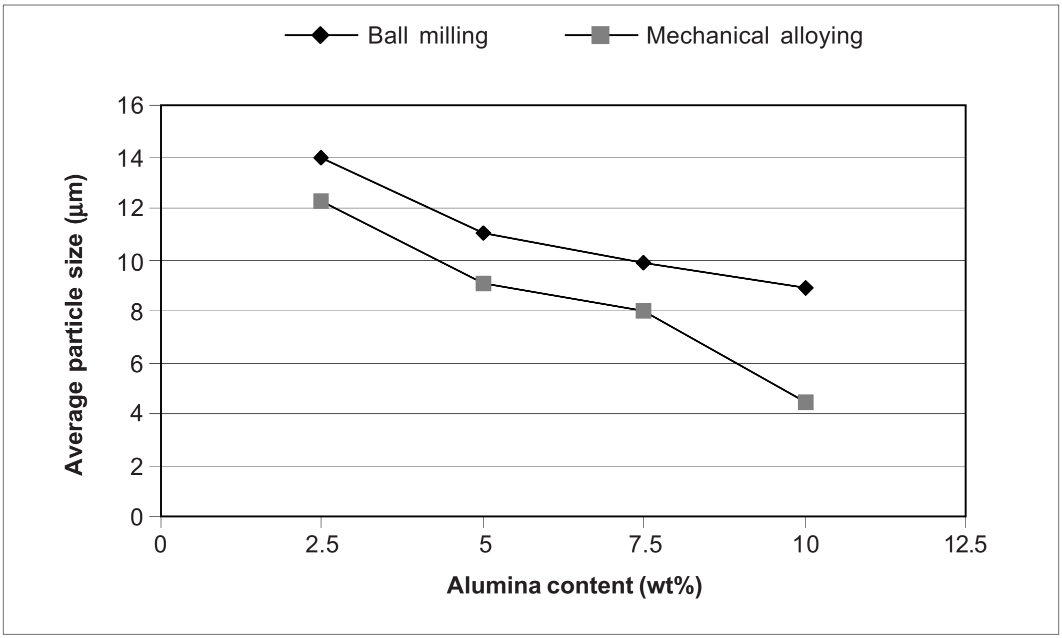


Figure 1 Particle size of Cu-Al_2O_3 powders prepared by ball milling and mechanical alloying

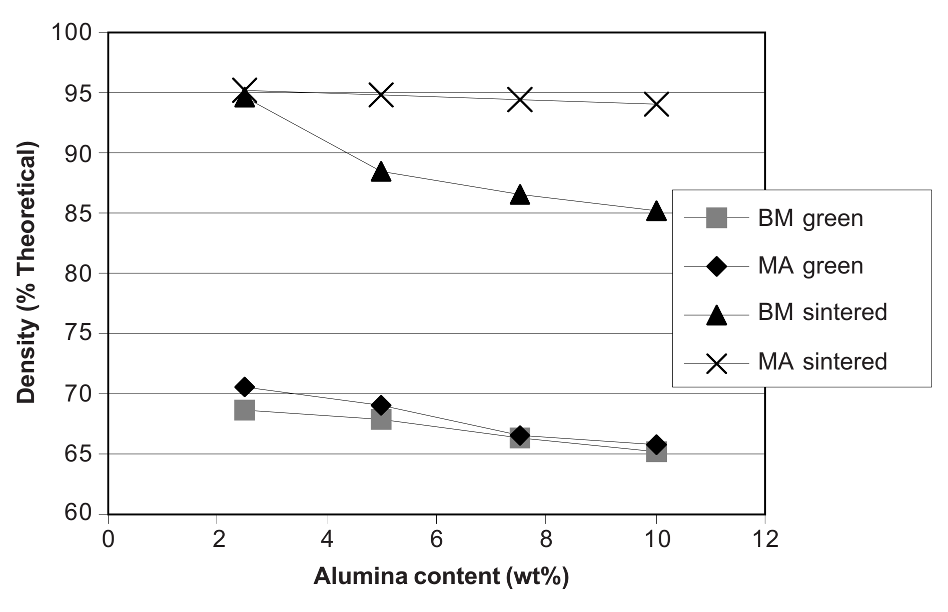


Figure 2 Green density and sintered density of Cu-Al_2O_3 composite prepared by ball milling (BM) and mechanical alloying (MA)

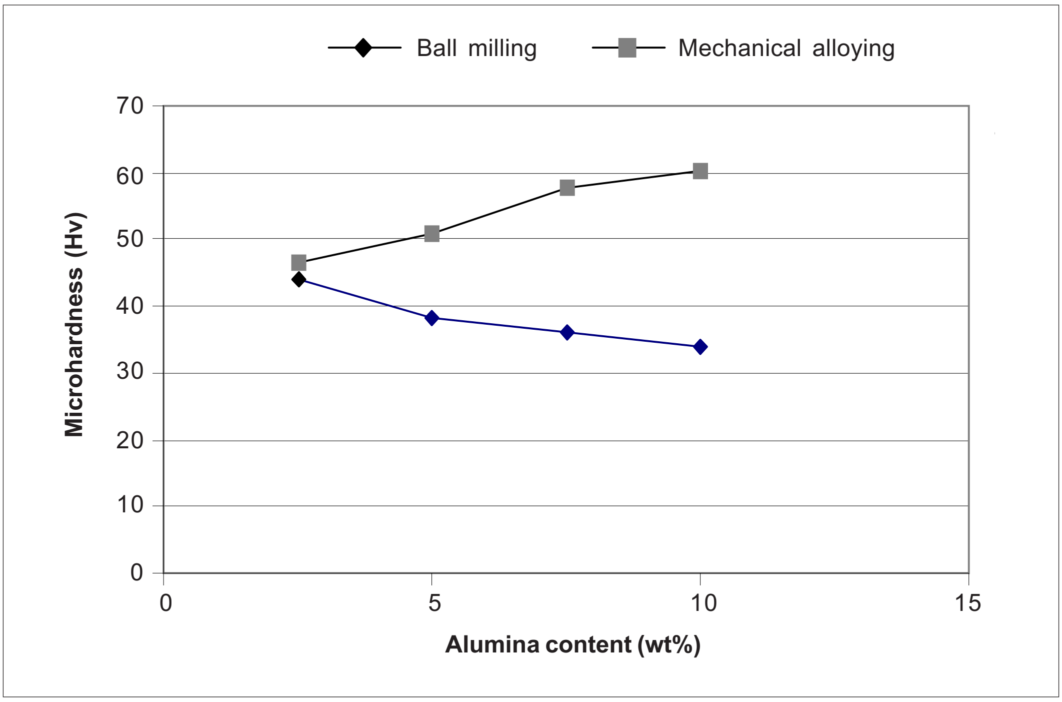


Figure 3 Hardness of Cu-Al_2O_3 composite prepared by ball milling and mechanical alloying

alloying composite, an increase in alumina content from 2.5 to 10wt% has caused a uniform increase in hardness whereas in the case of ball milling, hardness was found to decrease with an increase in alumina content. The variation in electrical conductivity with respect to alumina content in the composite is shown in Figure 4. Obviously, the conductivity of blended composite is higher than that of mechanically alloyed compact. Both types of composite show a uniform decrease in electrical conductivity with an increase in alumina content.

It is worth noting that an increase in alumina content up to 10wt% for ball milled composite causes agglomeration of alumina particle which will lead to pull-out during polishing and induces pores on the surface of the compact samples, which does not occur in mechanically alloyed composite (Figure 5(a) and 5(b)). From optical microstructure of the unetched composite (Figure 6), it is evident that mechanical alloying has caused appreciable grain size reduction. With powder of smaller particle size, more grain boundaries are formed (Figure 6). An increase in alumina content also causes grain size reduction in both mixing methods.

4.0 DISCUSSION

It is evident from the results that mechanical alloying in planetary mill is very effective in reducing the size of mixture powders compared to ball mill, which is in agreement

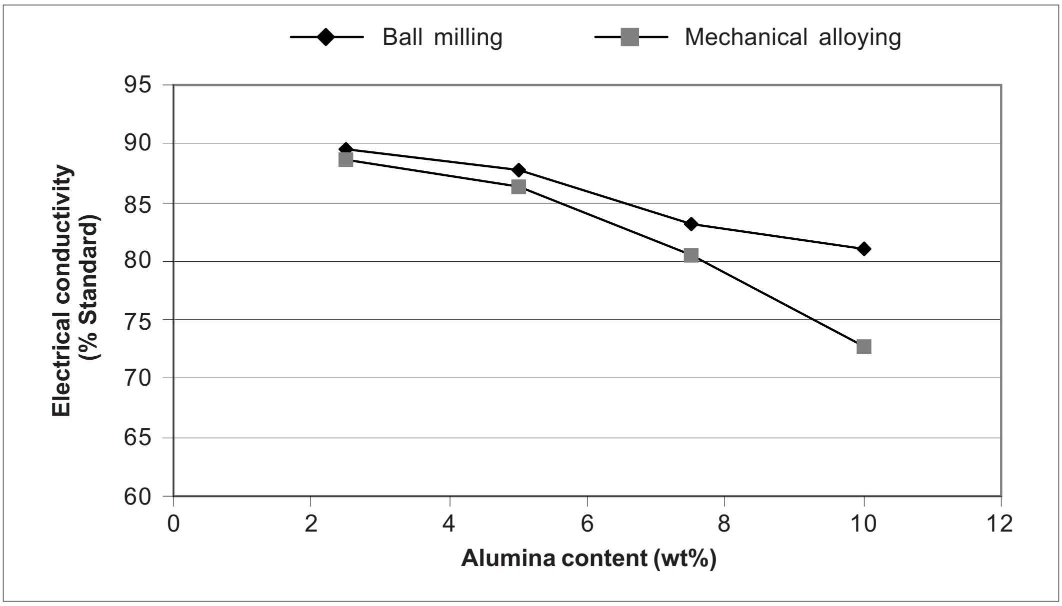
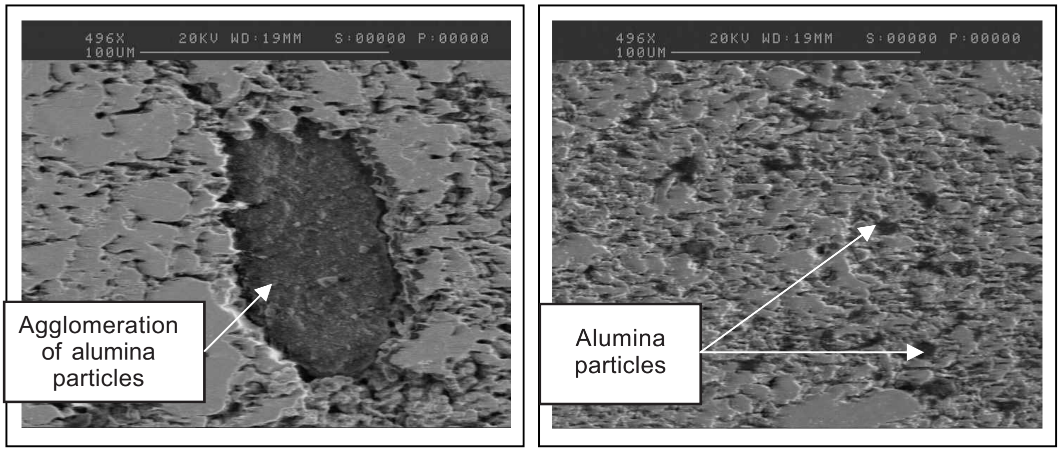


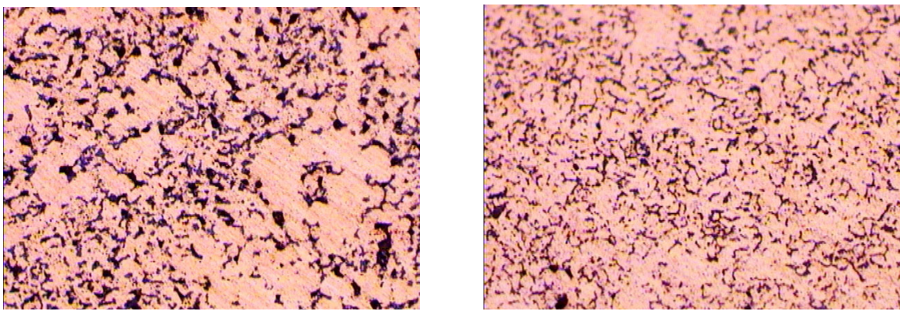
Figure 4 Electrical conductivity of Cu-Al_2O_3 composite prepared by ball milling and mechanical alloying



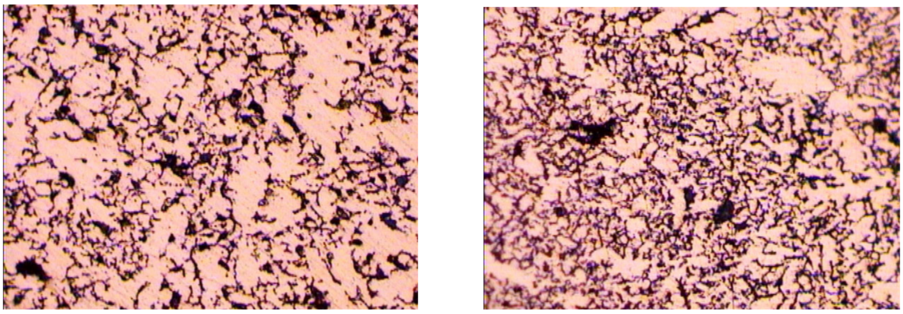
(a) (b)

Figure 5 SEM microstructure (etched) of Cu-10wt% Al_2O_3 composites prepared via (a) ball milling and (b) mechanical alloying (500x)

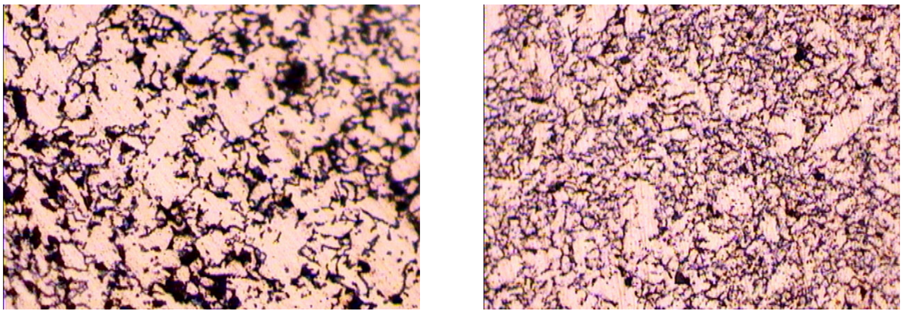
with the results of previous work [2]. The raw powder of copper and alumina become continually intermixed leading to a homogenous material with a uniform second-phase dispersion, i.e. alumina. This is because planetary mill is a high-energy mill, which operates under high velocity, and collision with milling balls enhances the powder refinement. Fine and hard alumina particle also act as milling agent, which



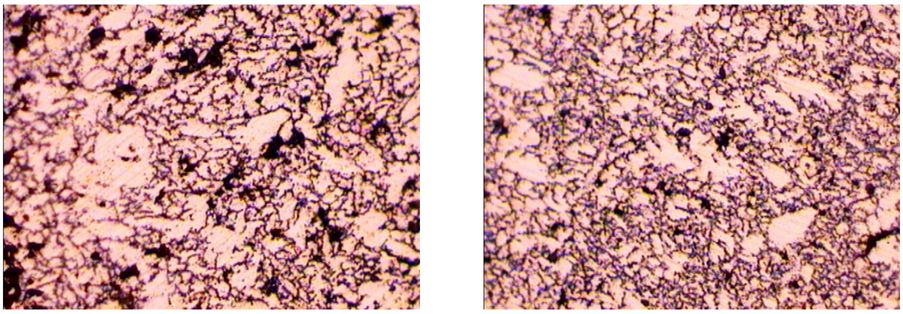
(a) Cu-2.5wt% Al_2O_3 BM (e) Cu-2.5wt% Al_2O_3 MA



(b) Cu-5wt% Al_2O_3 BM (f) Cu-5wt% Al_2O_3 MA



(c) Cu-7.5wt% Al_2O_3 BM (g) Cu-7.5wt% Al_2O_3 MA



(d) Cu-10wt% Al_2O_3 BM (h) Cu-10wt% Al_2O_3 MA

Figure 6 Optical microstructure (unetched) of Cu-Al_2O_3 composites prepared via ball milling (a)-(d) and mechanical alloying (e)-(h). (200x) helps in reducing the powder size [3]. Therefore, the average particle size decreases with an increase in alumina content for both mixing methods.

In general, sintered density of all composites is higher than that of unsintered composites, as shown in Figure 2. This can be explained by the reduction in surface energy, dimension shrinkage of the composite and internal voids during sintering process [4]. The green and sintered densities of composite prepared by mechanical alloying are higher than those prepared by ball milling. This is because composite powder prepared by mechanical alloying is finer, as shown in the result of particle size analysis. Fine powder enables more interparticle contact area to be formed and reduce pore volume during compaction. In addition, inhomogenous alumina distribution in copper matrix also affects composites green density, which gives explanation for the lower green density observed in ball milling composite. This is because the inhomogenous alumina distribution in ball milling samples has caused the fine alumina particles to agglomerate, as shown in Figure 5, which prevents a good contact between copper particles during compaction [5]. This explanation can also be applied for the drop in sintered density with higher alumina content. This is because an increase in alumina content has retarded powder densification during compaction, by preventing the interaction between copper powder, and during sintering as well, by interfering with grain growth [6].

The microhardness of mechanical alloying composite was found to be higher than that of ball mill composite due to its homogenous alumina dispersion in copper matrix. Finer powders produced by mechanical alloying also contributes to greater hardness as agreed by Hall-Petch theory [5]. An increase in alumina content has increased the hardness of mechanical alloying samples but unfortunately induces a decrease in hardness for ball milling samples. This is because for the ball milling process, higher number of alumina particles has created more alumina agglomeration and thus weakens the copper matrix.

Electrical conductivity of mechanical alloying composite is lower than its counterpart because mechanical alloying has produced finer powder and therefore, more grain boundaries are formed which imparts more electron scattering [2]. According to Mathiessen Rules, electrical conductivity is also affected by resistivity due to impurities and plastic deformation [7]. Therefore, mechanical alloying process, which permits more plastic deformation to the composite powders, achieves lower electrical conductivity. An increase in alumina content also decreases the composite's electrical conductivity since alumina is an insulator and act as impurities in copper matrix.

The microstructure in Figure 6 shows that mechanical alloying composite has smaller grains, more grain boundaries and better alumina dispersion compared with ball milling samples. This is due to the high energy process by mechanical alloying which not only decreases the particle size but also helps in dispersing the alumina particles. With smaller particles, more grain boundaries have been formed. This situation does not occur to the ball milling composite because it is a low energy process and the alumina tends to agglomerate particularly for samples with 7.5 and 10wt% alumina.

5.0 CONCLUSION

From the results of this study, a conclusion can be drawn whereby the mechanical alloying process is more suitable in producing alumina dispersion strengthened copper composite. Mechanical alloying is a mixing process, which can refine the composite powder and also disperses the alumina hard particles uniformly within the copper matrix. Both factors contribute to the balance between the hardness and electrical conductivity of the composite. On the other hand, an increase in alumina content has increased the hardness of mechanical alloying composite but their density and electrical conductivity decreased due to matrix hardening and electron scattering, respectively. Therefore, the most suitable composition is that with alumina content equal to or less than 5wt% as to achieve equilibrium in both hardness and electrical conductivity.

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