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Effect Of Ultrasonic And Ball Milling Process On Particle Size, Specific Surface And Its Agglometaion Of Metallic Interconnect Material

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Abstract

Metallic material become interesting field for interconnect application that have high thermal stability and oxidation resistant at high temperature. The main problem is the agglomerates powder and grain growth of metallic material that lead to decrement of properties of interconnect material. Therefore, the main objectives of this research is to develop small size particle up to nano range size and even particle size. The methods of this research was performed though high energy ball milling for 60 h and ultrasonic bath for various holding time of 3 h, 3.5 h, 4 h, 4.5 h and 5 h. The analysis and characterization process will be conducted by Powder size analysis by particle size analyzer (PSA) and Scanning Electron Microscopy (SEM) with magnification of 1000 and 2000 times. The result shows that the particle size decreased gradually from UT samples, Milled 60 h samples and to milled 60 h and UT samples. Its shows that particle size of 38.67µm for raw material, 11.45µm for UT 4.5 samples, 6.27 µm for milled 60 h sample and 5.23 µm for milled 60 h and UT 4.5 h. Fine surface structed and even particle size was shown by UT samples and combination samples due to high energy bubbles through liquid media that collide the material.

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Background, Motivation and Objective

Metallic interconnect has interest material for interconnect application due to their high thermal conductivities (Khaerudini et al., 2012; Dafit Feriyanto and Supaat Zakaria, 2020). In this study, nanocrystalline iron and chromium based alloys is very interesting and it has long been used by many engineering alloys as the basis in high-strength applications such as for fuel cell interconnect.

Developing $Fe_{80}Cr_{20}$ based alloy using mechanical alloying via high energy ball milling and ultrasonic processing in liquid media were successfully carried out by Saryanto et al., (2009) and Juan et al., (2010), respectively. The main objectives of mechanical alloying technique is to synthesize single-phase FeT (T = Cr, Cu and Ni) binary alloys with crystalline size less than 50 nm (Martinez-Blanco et al., 2011). In addition, the ultrasonic technique was performed to improve the reactivity of metals as stoichiometric reagents as synthetic technique for many heterogeneous organic and organometallic reactions. Ultrasonic also very effective to break the agglomerates, aggregates and even primaries (Krisztian et al., 2010). Therefore, it is very challenging to develop nanocrystalline FeCr alloy and develop even powder size because the Cr metal encourage the formation of protective oxide (scale) through alloying process with minimal agglomeration (Quadakkers et al., 2003). The agglomeration may cause the several problems such as oxygen cavitation inside the huge powder size, decrease the thermal stability and decrease the strain of material (Dafit Feriyanto et al., 2020 and 2021).

The challenge of this research is to develop homogenize alloyed powder size. Therefore the high energy ball milling and ultrasonic technique is used. However, the processing techniques involves extreme condition such as high-temperature at longer processing times that lead to the growth rate of crystallite size inevitable. In previous research that conducted by Hendi (2010), grain growth from 5.82 nm to 38.51 nm was occurred. Meanwhile, Ultrasonic technique with fixed frequency develop and achieve homogenous and fine grain structure (Ade, 2012). According to Ade, (2012) that conduct the research by ultrasonic technique with frequency of 18.52 kHz and holding time of 10, 20, and 30 minutes and the result shows that parameter achieve the rough surface and less homogenous. Therefore the main objective of this research is to combine the high energy ball milling and ultrasonic technique with selected parameters to achieve smaller and even powder size.

Methods

Powder blending is conducted by using high energy ball milling process reduce the crystallite size, to develop alloy material and good solid solubility. Ball milling process was prepared by cleaning the ball and the grinding jar using ethanol (95%) for 5 minutes, pause time of 5 seconds and the angular speed of 200 rpm with tolerance of ± 2 rpm. The ration of the ball and powder mass to blend the powder effectively is 13:1(Fnidiki *et al.*, 2005). The iron-chromium was mixed at the composition of 80 wt% Fe (50.4 g) and 20 wt% Cr (12.6 g) and mixed manually for 1-2 minutes and put into 250ml grinding jar. The grinding jar is placed to the glove box (Figure 1) and the pressure tube for glove box was set at 2000 psi with 20 psi of nitrogen pressure. The samples in grinding jar was placed into planetary ball milling (Figure 2) and it conducted by certain parameters such as fixed milling time of 60 h, the rotation speed of 300 rpm$\pm$ 2 rpm, 30 minutes cycle time, 10 minutes pause time.

Figure 1. *Glove box*

Figure 2. *Planetary ball mill*

Ultrasonic technique was carried out by fixed frequency of 35kHz due to conventional power ultrasound is 20kHz – 100 kHz (Krisztian *et al.*, 2010) and the ultrasonic machine only have fixed frequency of 35 kHz. The iron-chromium powder was prepared for 10 g for each parameter. Ultrasonic process was conducted using different holding time of 3 h, 3.5 h, 4 h, 4.5 h and 5 h for each samples as to achieve the optimum times for breaking the agglomerates.

The powder size analysis was conducted by Particle Size Analyzer (PSA) machine where the water is used as liquid media in PSA process. The powder were mixed with water for ± 5 minutes operation and then PSA machine generate the result of particle size.

The agglomeration of the FeCr powder was analysed by Scanning Electron Microscope (SEM) using magnification of 1000 times for raw material and Ultrasonic (UT) samples and 2000 times for milled 60 h and combination technique samples. The SEM was operated at 10 kV to capture images of the $Fe_{80}Cr_{20}$ alloy powder.

Results and Discussions

Sample name in this research was sign as $Fe_{80}Cr_{20}$ for raw material UT for Ultrasonic samples milled 60 h for samples that treated by high energy ball milling for 60 h and milled and UT for combination technique of high energy ball milling and ultrasonic technique.

The particle size, specific surface and distribution of particle size of treated and untreated samples is listed in Table 1. Raw material shows the highest particle size of 38.67µm and smallest specific surface of 865.22cm^2/g. UT samples shows the relatively smaller particle size and specific surface but the smallest particle size and highest specific surface of UT samples is UT 4.5 h sample for 11.45µm and 1694.58cm^2/g, respectively. Milled 60 h samples shows the significant reduction of particle size and significant increment of specific surface for 6.27 µm and 1980.92 cm^2/g, respectively. The combination technique was performed to decrease the particle size and improve the homogeneity and it approved with the data that the combination technique produces the smallest particle size of 5.23 µm and 2376.03 cm^2/g. Ball milling and UT treatment process are significant in reducing the particle size and improving homogeneity. However, the UT is less effective to reduce the particle size compared with ball milling process. It is caused by UT treatment which is focused on improving the homogenous of powder size.

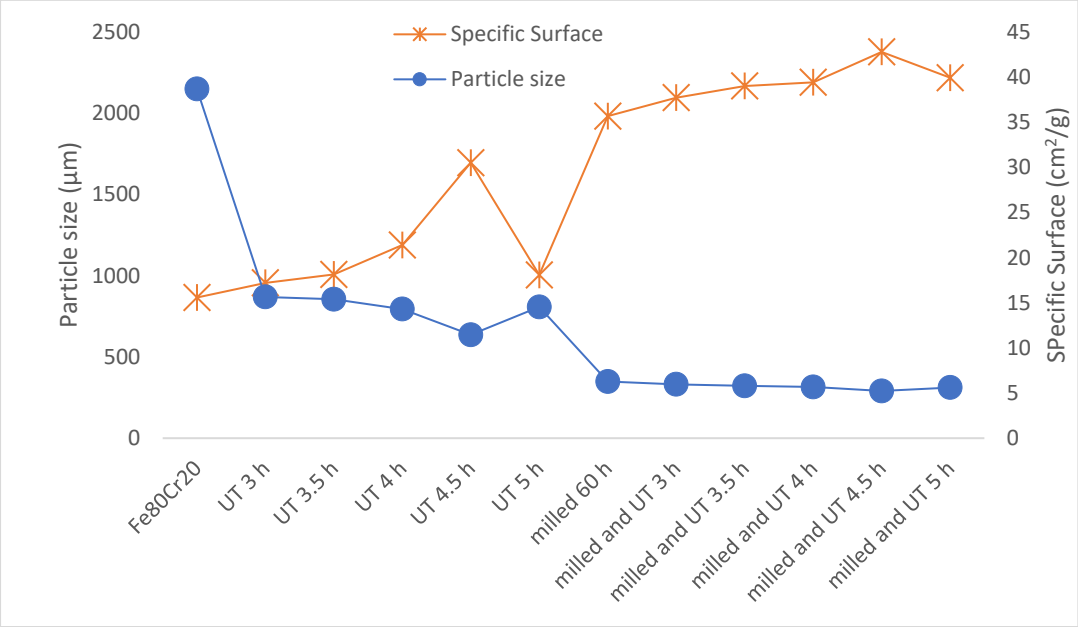
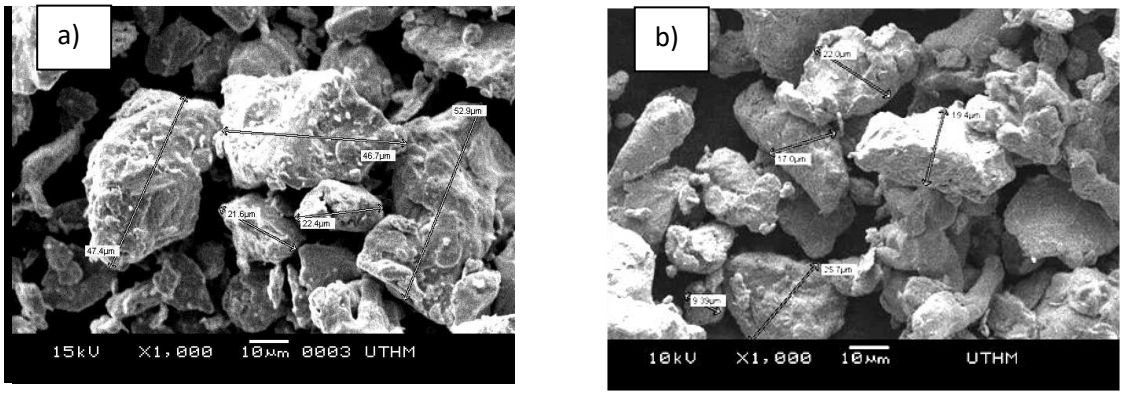


Figure 3. *Particle size and specific surface of treated and untreated samples*

Surface Morphology Analysis

Surface morphology of the raw material and UT samples is shown in Figure 4. The raw material have rough surface structure and not homogenous powder size. The particles consist of irregular round shapes because it was beneficial since it provides the full inter-particle bonding during the solidification process (Tokita, 2006 and Hendi, 2011).



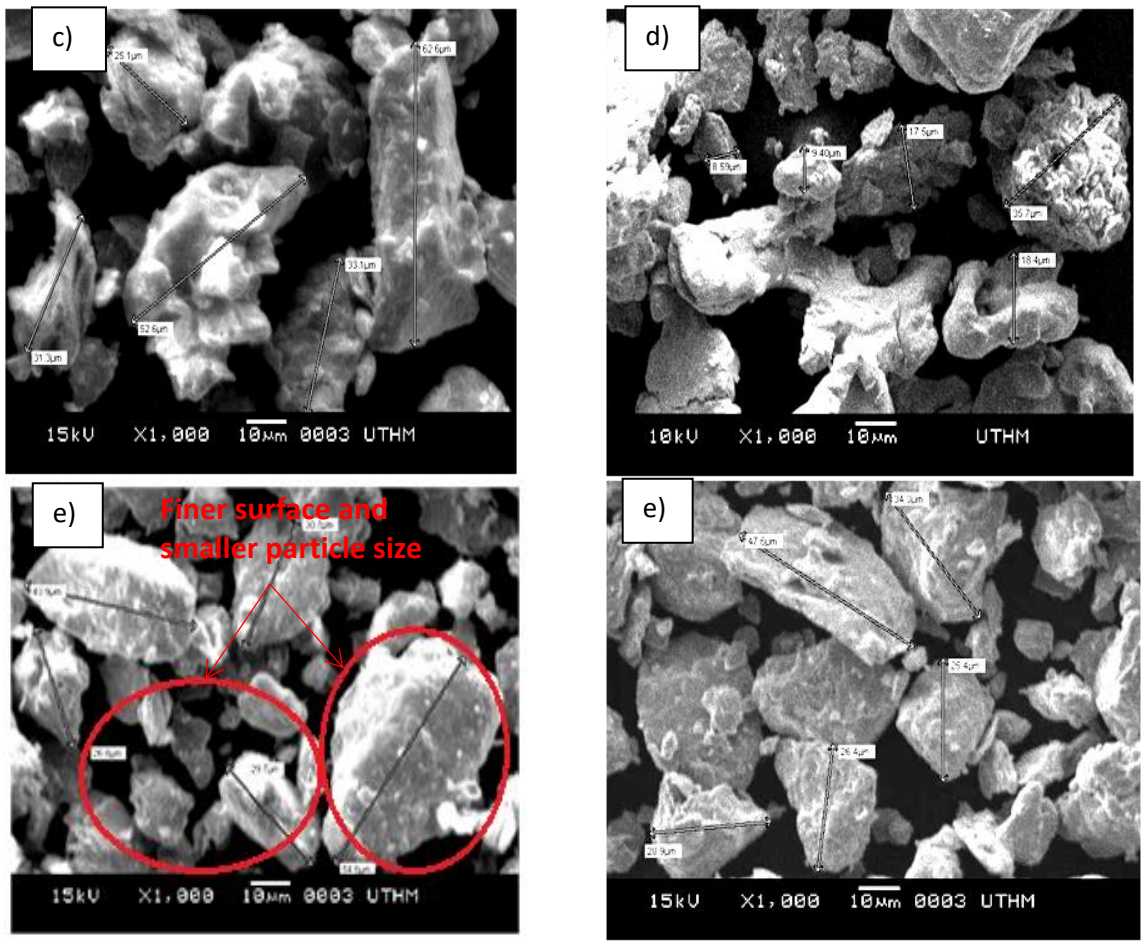


Figure 4. *Surface morphology of a); Raw material; b): UT 3 h; c): UT 3.5 h; d): UT 4 h; e): UT 4.5 h; f): UT 5 h*

The homogeneity $Fe_{80}Cr_{20}$ alloy powder increased by using ultrasonic treatment due to. inside cavitation bubbles which lead to shock out wave into the liquid. Metal particles can be pushed together with a high speed to melt effectively at the point of collision. That phenomena supported by Puga *et al.*, (2011) that the ultrasonic treatment lead to surface modification through cavitation as a result of high speed bubbles that generates jet slugging upon the surface of the each sample powders. Good surface morphology is shown by fine surface of $Fe_{80}Cr_{20}$ alloy powder as shown by UT samples where the ultrasonic treatment generates good surface morphology and effective for breaking the agglomerates as compared to raw material. That result was supported by data after ultrasonic treatment that the particle size was decreased from 38.67µm to 11.45µm.

Surface morphology characterization of the milled 60 h and Milled and UT samples with high magnification of 2000 times is shown in Figure 5. Ball milling process aimed to decrease the powder size through the ball pressure since ball collision to the powders. Meanwhile, the ultrasonic process focused on breaking the powder agglomerates and refine the surface morphology. Increasing ultrasonic time increased the homogenous and surface morphology of the milled and UT samples as shown in Figure 5(c), (d) and (e). However, in the milled and UT 5 h sample, the agglomerate starts to increase. It is showed clearly in Figure 5(f).

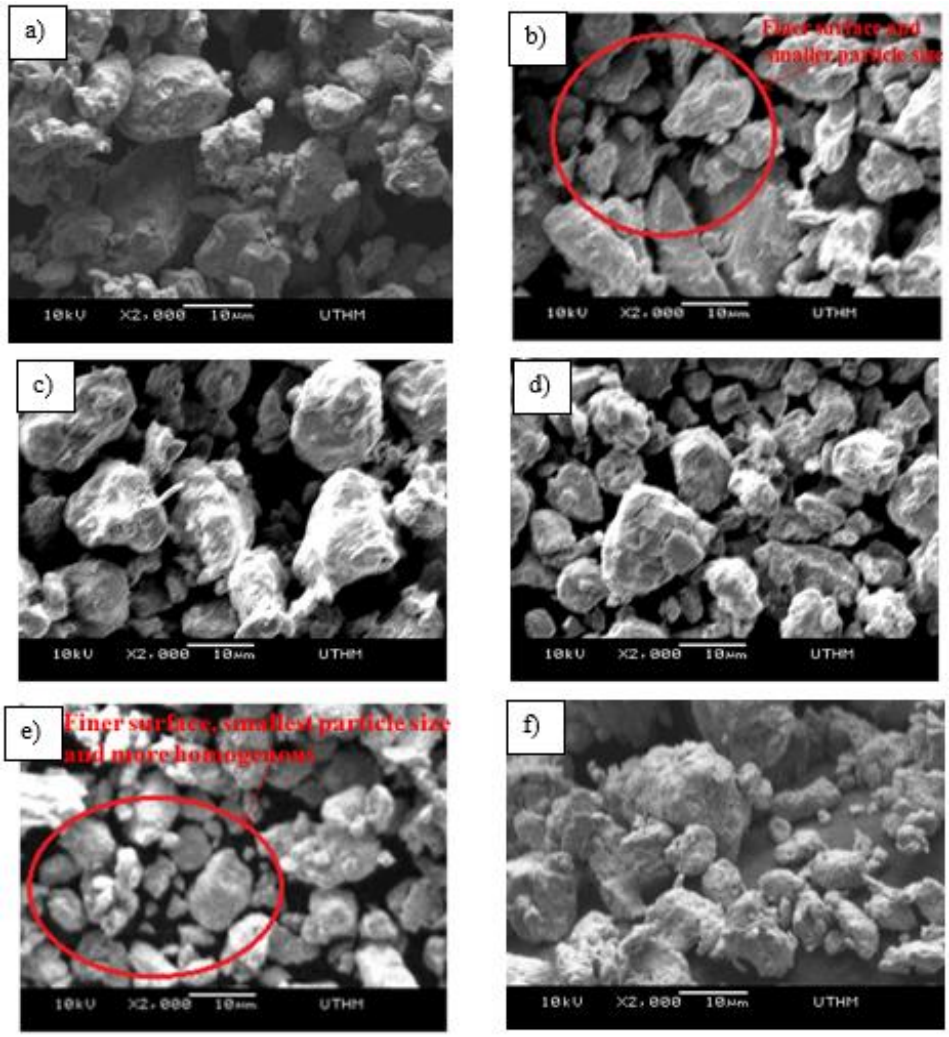


Figure 5. *Surface morphology of a); milled 60 h; b) milled and UT 3 h; c): milled and UT 3.5 h; d): milled and UT 4 h; e): milled and UT 4.5 h; f): milled and UT 5 h*

In the ball milling process, distribution of deformation is not even in powder body. Therefore, the $Fe_{80}Cr_{20}$ alloys powder of milled 60 h sample is not a homogenous powder and need to generate ultrasonic treatment to improve the homogeneity of the powder size. Even though the control agents used during the ball milling process, the agglomeration is still developed because it did not maximum inhibits the agglomeration. The materials after milling process are consisted of nanometer range size, so it was easy to agglomerate as mentioned by Redjdal et al., (2013), shows that the agglomeration of the metallic alloy increased when the materials in nanometer range size. According to Suryanarayana (2001) and Akira Tayamal et al., (2006) stated that the distribution crystallite size is ease increase when the control agent in liquid form as compared to solid or gas form.

A new method of developing nanocrystalline alloy was conducted by using combination technique to reduce the crystallite size, refine the surface morphology, and more homogenous or break agglomerate. Figure 5(e) shows that the agglomerates of milled and UT 4.5 h has decreased. However, the agglomeration still occurs in smaller size. Milled and UT 4.5 h is optimum parameter to reduce crystallite size, increase homogenous and surface of the alloy powder as compared to the other parameter.

Conclusions

Developing $Fe_{80}Cr_{20}$ interconnect material was achieved by high energy ball milling and ultrasonic technique and the results shows that the ball milling was very effective to decrease the particle size from 38.67 to 6.27 µm and improve the specific surface from 865.22cm^2/g to 1980.92 cm^2/g. This research achieves the finer surface structure and even particle size through ultrasonic treatment. In interconnect application, this result gives a significant recommendation regarding to treatment that need to proposed in interconnect material which is combination of high energy ball milling and ultrasonic treatment. Where the optimum parameter was achieved in Milled 60 h and UT 4.5 h.

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