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IN-SITU FORMATION OF NbC IN MECHANICALLY ALLOYED Cu-Nb-C AT DIFFERENT TEMPERATURES

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Abstract

In this study, a high-energy ball milling called mechanical alloying was applied to synthesis *in- situ* copper-based composite reinforced with niobium carbide particle. Cu, Nb and graphite powder mixture were mechanically alloyed for 32 h in a planetary ball with composition of Cu-11.77 %Nb-1.52 %C. The samples of the as-milled powder were compacted at 300 MPa to produce 10 mm diameter pellets. In order to investigate the effect of temperature on the carbide formation, the green compact were sintered in an argon atmosphere at different sintering temperature i.e. 600, 700, 800 and 900 °C. The change of phase and micro-structure of the sintered compact were characterized by X-ray diffraction (XRD) and scanning electron microscope (SEM). The result from XRD shows that the NbC phase was not detected at sintering temperatures lower than 900 °C due to not having enough energy to initiate the reaction of Nb and C in Cu matrix. Cu shows an increase in the crystallite size while the internal strain decreases with increasing sintering temperature. The crystallite size of NbC was unresolved since the NbC peak is poorly defined at lower sintering temperature. From EDX analysis, higher oxide content was observed in Cu matrix at higher temperature which retards the sintering process.

Introduction

In-situ method involves synthesizing reinforcement in metallic matrix by chemical reaction during mixing and the subsequent processing. According to Tjong et al. [1], the formation of both fine *in-situ* reinforcements and metal matrix exhibits superior mechanical properties when compared with the conventional processes. Nanostructured powder particles with well-distributed reinforcement become increasingly synthesized by mechanical alloying (MA). Synthesizing *in-situ* composite by combination of MA and subsequent compaction and sintering is less reported in literatures. Although many primary works on MA of Cu-Nb-C system has been done [2-5], however, the properties after compaction and sintering are not well understood. Botcharova *et al* [6] observed that the properties of Cu-Nb alloy after heat treatment change with different temperature fabricated by MA and powder metallurgy. This work aims to determine the effect of different sintering temperatures on the formation of NbC in *in-situ* Cu-NbC composite.

Experimental Procedure

The elemental powders of Cu, Nb and graphite mixtures were milled in a planetary ball mill for 32 h under an argon atmosphere. The speed and ball-to-powder ratio used was 400 rpm and 5:1, respectively. In order to prevent the oxidation, Cu and graphite powder were first milled for 4 h in stainless steel jar. Milling time start to be taken as Nb was added. The as-milled powder mixture then compacted at 300 MPa in 10 mm diameter pellet. Sintering of green compact was carried out in an argon atmosphere at different temperatures, i.e. 600, 700, 800 and 900°C for one hour.

Result and Discussion

Figure 1 shows the XRD pattern of sintered Cu-Nb-C pellet milled for 32 h with several subsequent sintering temperatures e.g. 600, 700, 800 and 900ºC. In the range of 600 to 800 ºC, only two phases could be resolved which include Cu and CuO. These phases are also observed at 700ºC and 800ºC along with the additional formation of Cu_2O and NbO. In the temperature range of 600 to 800ºC, no peaks of NbC are detected probably due to not having enough energy to initiate the reaction of Nb and C in Cu matrix compared to the reaction at 900ºC. Also, at these temperatures, the fraction of impurities grows. Possible reason is because of the oxidation of powders during milling and the reaction of oxygen and powder during the sintering process.

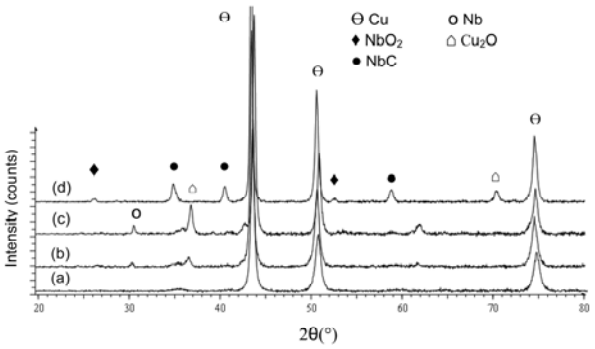

Fig. 1: XRD patterns of sintered pellet of Cu-Nb-C milled for 32 h : (a) 600ºC; (b) 700ºC; (c) 800ºC; and (d) 900ºC

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The crystallite size of Cu increases with increasing sintering temperature as shown in Fig. 2. For instance, the crystallite size of Cu increases from 23.86 nm at 600 ºC to 45.56 nm at 700 ºC. The maximum observed crystallite size of Cu is at 900ºC. The coarsening of the Cu crystallite drastically increases from 600 ºC to 700 ºC but become slow after 700 ºC. This trend could be explained by the influence of sintering temperature on precipitation of NbC particles in Cu matrix [6]. The growth of grains of Cu is much low at 900 ºC due to the formation of NbC reinforcement particle in the Cu matrix. However, the crystallite size of NbC is unresolved since the NbC peak is poorly defined at lower sintering temperature. The percentage of internal strain of Cu as evaluated from XRD patterns also presented in Fig. 2. By increasing of sintering temperature, the internal strain is decreased. For example, the internal strain is significantly decreased at 700 ºC which is half than that at 600ºC.

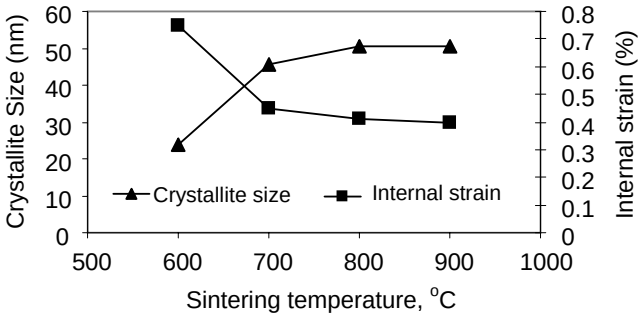


Figure 2. The crystallite size and the internal strain of Cu sintered *in-situ* pellet after 32 h of milling with various sintering temperatures using Williamson-Hall method

Figure 3 shows the SEM micrographs of Cu-Nb-C pellets (prepared from 32 h milled powders) after sintering at different temperatures. As can be seen, there are two regions observed in these micrographs. The grey area is corresponding to the diffusion of Nb in Cu matrix while the black regions contain pores of various shapes and sizes. Possible reason for large distribution of pores within microstructure at 900ºC was due to oxide formation as a result of higher oxygen trapped during compaction and sintering.

Evidently, from EDX analysis (Fig. 4) the presence of Cu and Nb is confirmed at all temperatures. At 700 ºC , the Cu rich region, marked as "d1" (Fig. 4(a)), approximately contains 82.75 wt% of Cu, 4.09 wt% of Nb, 8.5 wt% of O, 3.98 wt of C and 0.68 wt% of Fe. While for "d2" area (Fig. 4(c)) (sintering temperature 800 ^{o}C), the approximate chemical composition is 60.43 wt% Cu, 10.25 wt% Nb, 8.76 wt% O, 19.48 wt% C and 1.08 wt% Fe. Conclusively, sintering at higher temperature leads to higher oxide content in Cu matrix which retards the sintering process. In this work, Fe contamination has been neglected since it exhibits only in small percentage

Conclusion

The result from XRD showed that the NbC phase was not observed at sintering temperatures lower than 900°C. Copper's crystallite size increase while for the internal strain decrease with increasing sintering temperature.

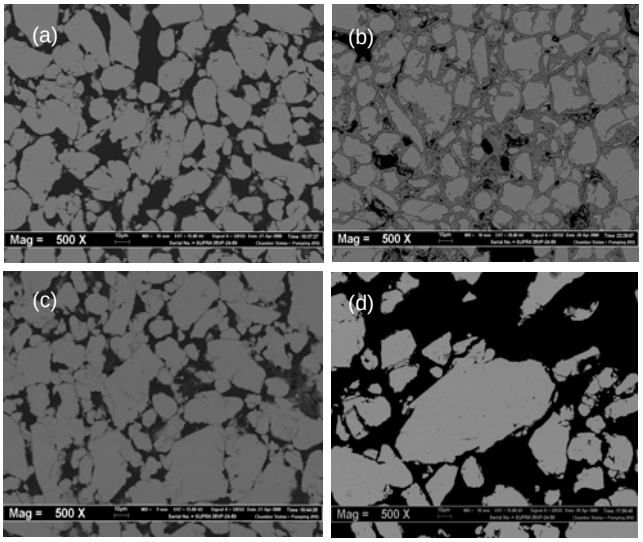
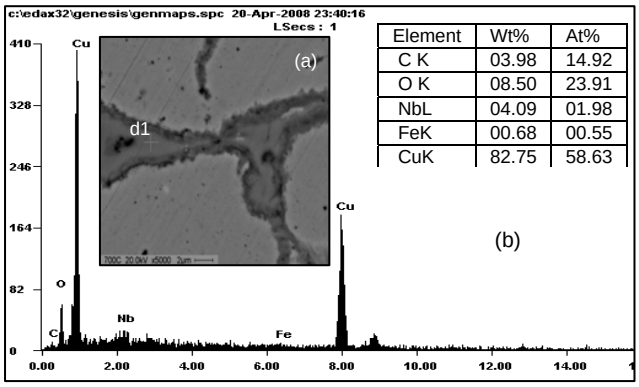


Figure 3. SEM micrographs of sintered *in-situ* pellet after 32 h of milling : (a) 600ºC (b) 700ºC (c) 800ºC and (d) 900ºC at 500X magnification



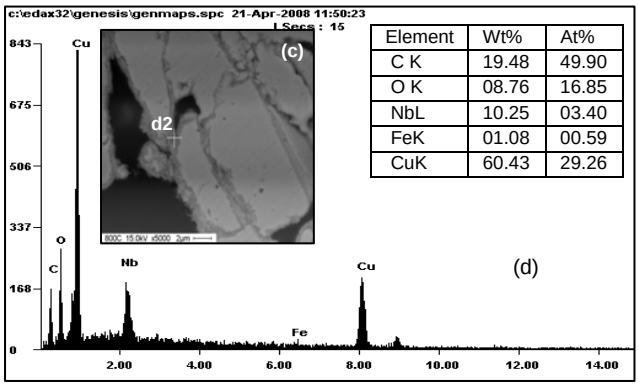


Figure 4. Sintered *in-situ* pellet prepared : (a) SEM image and (b) EDX spectra of "d1" region (sintering temperature 700°C); (c) SEM image and (d) EDX spectra of "d2" region (sintering temperature 800 ^{o}C)

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