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THE STUDY OF COPPER ALLOY WITH GRAPHENE ADDITIVE VIA X-RAY DIFFRACTION

This study evaluated the effect of milling speed and compaction pressure on the densification and morphology of the CuZn-Gr composite. The composite was prepared by using the powder metallurgy technique. The effect on the microstructural and compaction was determined based on different milling speeds. The different milling speeds involved were 175, 200, 225, and 250 rpm. Meanwhile, the different compaction pressures used in this study were 127, 250, 374, and 500 MPa. The properties of the milled powder gave the result to green density and densification parameters. The XRD pattern of Cu and Zn broadened as milling time increased.

Keywords: Copper Alloy; Graphene; Powder Metallurgy; Copper; Zinc

1. Introduction

Composite materials have two or more different elements with various chemical, physical, and mechanical properties. Composite materials are variance by polymer matrix composite (PMC), ceramic matrix composite (CMC), and metal matrix composite (MMC). Most composites consist of bulk material, a ‘matrix’, and the reinforcement added increases the strength and stiffness of the matrix [1-5]. Conventional metals and ceramics in MMC get outstanding properties such as high-temperature properties (300°C), lightweight, high strength (480 MPa), and good wear resistance. MMC applications have been extensively using as structural materials in aerospace, automotive, marine, and military industries [6].

Copper zinc (CuZn) materials are well-known as “brass.” CuZn is commonly employing in both the wrought and cast condition used for tubing, fitting and carrying water and other fluids [7]. Commonly, brass is a material with high strength, good heat and electrical conductivity, and ductile. There are numerous approaches to prepare the CuZn, such as liquid-state, solid-state, in-situ, and spray-forming particulates. Powder metallurgy (PM) is an alternative technique for composite fabrication. PM consists

of three paths which are mixing powder elements, compacting, and sintering. Besides, it can also use various metal or non-metal types and can create an excellent finish surface [1-2].

With powder metallurgy (PM) development, the studies of fabrication Cu-matrix composite have attracted increasing interest. For example, CuZn (brass) fabrication has been widely used as an industrial material because of its excellent high corrosion resistance, non-magnetism, and good plasticity [8]. CuZn is significantly less expensive than copper but has low strength properties that negate brass’s economic advantage.

To increase the strength of CuZn, the addition of one or more alloying elements such as Tin (Sn), Manganese (Mn), Nickel (Ni), Aluminum (Al), and Cobalt (Co) have worked by previous researchers [9]. Addition Al or Ni to CuZn makes the alloy heat treatable for only a limited extent and the resulting increase in strength is achieve when large quantities of Ni are present. As a result, the alloy is more expensive than conventional CuZn.

Furthermore, CuZn alloy containing a small amount of Co can also thermally hardened and the increase in strength is also limited. However, for great in number applications, CuZn-Co is offset by the higher cost of the alloy. As a result, there is

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a need for low-cost, high strength, corrosion resistance and suitable machinability alloy. In this work, CuZn will be reinforced with graphene (Gr) [1]. Gr is the most suitable material to be reinforced in CuZn because it has good physical and mechanical properties. It has a huge theoretical specific surface area ($2,630 \text{ m}^2 \text{ g}^{-1}$), high intrinsic mobility ($200,000 \text{ cm}^2 \text{ v}^{-1} \text{ s}^{-1}$), high Young's modulus (~ 1.1 to 2.0 TPa), thermal conductivity (up to $4,300 \text{ Wm}^{-1} \text{ K}^{-1}$), and good electrical conductivity [10-11]. The sequence of these exceptional properties makes graphene an outstanding candidate for applicant potential products. However, there has been no previous report of any CuZn-Gr composite. Thus, this research occurs to characterize CuZn-Gr by using x-ray diffraction (XRD) technique.

2. Methodology

The materials and experimental in this research will explain detail in this section. Copper zinc (CuZn) with graphene (Gr) composite was prepared with different milling speeds and compaction pressure parameters. The experimental work of producing the CuZn-Gr composite by powder metallurgy (PM) method was conducted through milling and compaction. Then, the CuZn-Gr was characterized by (XRD) to evaluate the effect of milling speed and compaction pressure.

Cu and Zn were mixed by using ratio Cu:Zn (65%:35%). Then, the mixture of CuZn was mixed with 0.3 wt% of graphene (Gr). Before the milling process, 2% of N-heptane was added to the mixture CuZn-Gr powder as a control agent. N-heptane was used to reduce die wall friction and agglomeration during compaction. The CuZn-Gr powders were mixed with different milling speeds (175, 200, 225, and 250 rpm) within 40 h.

Software DIFFRAC.EVA was used for phase identification to perform quantity and quality analysis on XRD pattern of the composite. It is used to identify crystallite size and internal strain. William-Hall method (WH) evaluated the crystallite size and internal strain after compacted of powder. The assumption is that the whole line broadening, B° is the total broadening of size, lattice strain, and instrument [1].

$$B^\circ = B_i - B_{crys} + B_{strain} \quad (1)$$

The B_i broadening of instrumental, B_{crys} broadening due to crystallite size and B_{strain} broadening due to strain. Subtracting the instrumental effect becomes [1].

$$B_r = B_{crys} + B_{strain} \quad (2)$$

The B_r is overall broadening after eliminating the instrument broadening. Therefore, due to crystallite size and internal strain, WH method is given [1].

$$B_r \cos \theta = k\lambda/D + \eta \sin \theta \quad (3)$$

where k is a constant (with a value of 0.9); λ is a wavelength of the x-ray radiation; D and η are the grain size and internal size, respectively; and θ is the Bragg angle.

3. Results and discussion

XRD pattern of the powder mixture of CuZn-Gr composite after milled with 175, 200, 225 and 250 rpm are shown in the Figure 1. The peak positions are almost identical for all milling speeds. The peak-height appears to decrease with increasing of milling speed. However, there was no visible formation of new phase.

The peak of the powder mixture does not show any shift to the left or to the right thus this can be assumed that there is no expansion or reduction of lattice parameter happened. This is due to the low energy milling which generates less energy to change the internal structure of the composite. The highest intensity of the powder mixture in Figure 1 was located at 43.3° , which identified as Zn crystalline.

When the milling speed increased, the broadening of the peak was decreased. There is difference in the area of the peak. This can be shown that the Zn peak broadened at 43.3° and the broadened peak of Cu located at 50.5° . The broadening of the CuZn-Gr composite is virtue from the refinement of structure which can be contributed from the changes in crystalline size and internal strain.

Additionally, the peak located at 43.3° which is highest resulted from the overlap peak Cu and Zn. As can be seen in Fig. 1, the peak of Zn is higher than to Cu peak. This is because of Zn has a larger radius than Cu atom. This result suggests that higher gas pressures decrease the Zn volatility. Moreover, the peak of Gr is extraordinarily short because Gr was added in small quantity in powder mixture.

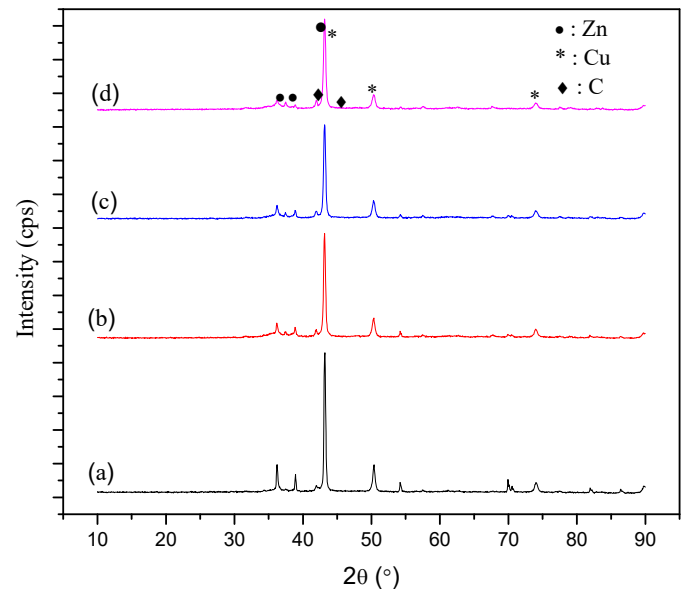


Fig. 1. XRD patterns of milled powder at (a) 175 rpm (b) 200 rpm (c) 225 rpm, and (d) 250 rpm

In order to calculate crystallite size and internal strain of milled powder composite, the WH method was used (Fig. 2). The data derived from the method were plot $B_r \cos \theta$ against $\sin \theta$ is plotted as shown in Figure 2. When the milling speed

is increased, all of intercept and slope resulted in positive values. The linear plot shows that the structure refinement was derived from crystallite size and internal strain.

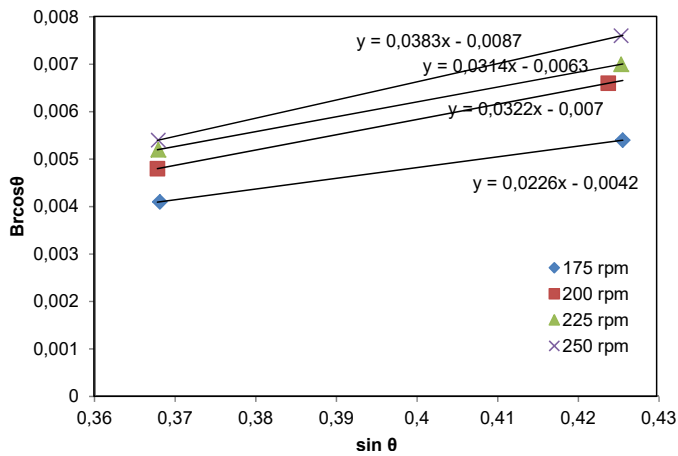


Fig. 2. Plot of $B_r \cos \theta$ against $\sin \theta$ for calculating crystallite size and internal strain for the powder milled

Fig. 3 shows that the graph plotted of crystallite size and internal strain against different milling speeds. Based on the graph, the crystallite size of CuZn-Gr decreases significantly as the milling speed increased from 175 to 250 rpm. This is due to utilization of kinetic energy of high-density alumina ball as a function of their mass and velocity with varying milling speed has affected the crushing and amorphization of the crystallite size reduction as mentioned with previous finding [12]. This is the evidence of the smaller microstructure of CuZn-Gr milled composite that was produced in this work.

Fig. 3 shows the internal strain increased when the milling speed is increased. Increasing milling speed resulted increasing the milling efficiency in reducing particle size and introducing the internal strain. In addition, when milling speed increase, the microstructural evolution and chemical reactions also increases can be obtained from this work.

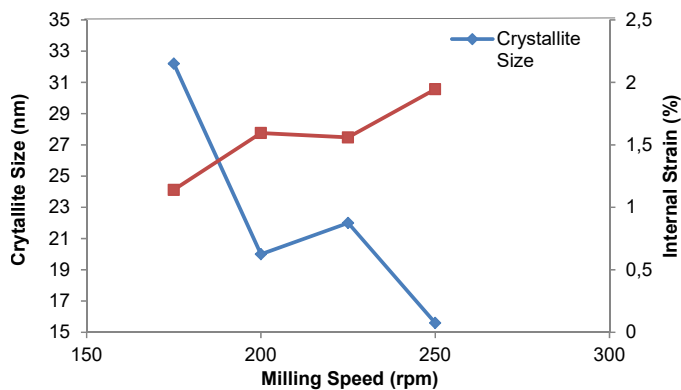


Fig. 3. CuZn-Gr crystallite size and internal strain of milled powder with different milling speed

4. Conclusion

Milled powder that milling at 200 rpm and 225 rpm provided highest green density and densification parameter of CuZn-Gr composite with increasing of the compaction pressure.

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REFERENCES

- [1] M. Yusoff, M. Mohamad, M.B.A. Bakar, M.N Masri, N.Z. Noriman, O.S. Dahham, AIP Conference Proceedings **2213** (1), 020264 (2020).
- [2] M.N. Masri, M. Yusoff, N.N. Nordin, IOP Conference Series: Materials Science and Engineering **440** (1), 012021 (2018).
- [3] I.H. Abdullah, M.N. Masri, M.H. Razali, M. Yusoff, AIP Conference Proceedings **2068** (1), 020052 (2019).
- [4] S. Kahar, M.N. Masri, M. Mohamed, M.H.M. Amini, SA Sobri, WS Hashim, IOP Conference Series: Earth and Environmental Science **765** (1), 012116 (2021).
- [5] M.N. Masri, M.B.A. Bakar, N.W. Rusli, M.F. Ismail, M.H.M. Amini, A.A. Al-Rashdi, International Journal of Integrated Engineering **13** (4), 172-179 (2021).
- [6] V. Srinivasan, S. Kunjiappan, P. Palanisamy, International Nano Letters. 1-25 (2021).
- [7] S.A. Shabalovskaya, Bio-Medical Materials and Engineering **12**, 1, 69-109 (2002).
- [8] S. Li, H. Imai, H. Atsumi, K. Kondoh, Materials & Design **32**, 1, 192-197 (2011).
- [9] C. Jun, D. Lifeng, L. Qiang, C. Chongyan, W. Ruonan, K. Xiangpeng, S. Zhengwei, Z. Xiaohong, N. Yulan, Journal of Applied Electrochemistry **50**, 4, 475-488 (2020).
- [10] S.V. Morozov, K.S. Novoselov, M.I. Katsnelson, F. Schedin, D.C. Elias, J.A. Jaszczak, A.K. Geim, Phys. Rev. Lett. **100**, 016602 (2008).
- [11] J.W. Jiang, J. Wang, B. Li, Phys. Rev. B **80**, 113405 (2009).
- [12] H. Ahamed, V.S. Kumar, Journal of Minerals & Materials Characterization & Engineering **10**, 507-515 (2011).