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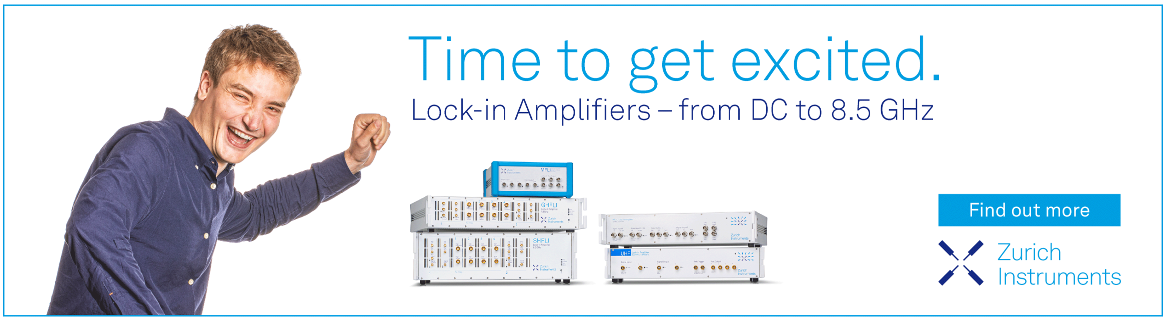

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Effect of Copper Addition on Aluminum Matrix Composite by Powder Metallurgy Method

Aswar[1, 2, b)], Muhammad Syahid[1, a)], Azwar Hayat [1, c)]

[1]*Department of Mechanical Engineering, Faculty of Engineering, Hasanuddin University, Makassar, Indonesia*
[2]*Mechanical Engineering, Akademi Teknik Soroako, Soroako, Indonesia*

[a)] Corresponding author: syahid@unhas.ac.id
[b)] aswar.tekno@gmail.com
[c)] azwar.hayat@unhas.ac.id

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Abstract. Lightweight metals with optimal properties are in great demand in the automotive and aerospace fields. The manufacture of composites using the powder metallurgy method offers both process and product advantages. A simple process with a product shape close to the final shape provides an economic advantage. This study discusses copper-reinforced aluminum matrix composites made by the powder metallurgy route. A mixture of Al-1.5%Mg-5%Graphite-x%Cu powder with the addition of copper reinforcement (0, 4, 7, and 10 % mass fraction) to produce composites. The cold uniaxial pressing method at a pressure of 150 MPa produce green compacts. Sintering process at 600 ^{o}C for 90 minutes to get strength and rigid shape. An optical microscope to observe powder form and mixture uniformity, and a Laser Scanning Microscope to observe the composite's microstructure. The physical properties test includes porosity and Archimedes density of the composite. Vickers hardness and wear rate tests to evaluate the mechanical properties of composites. The results showed that adding copper to the alloy initiates hardness and increases the wear resistance of the composite. In this study, higher hardness reached 81.5 HV at a density of 3.04 g/cm^3 with the highest porosity level of 7.30 %. A lightweight, lead-free composite with optimized properties offers reliable properties as an alternative material in automotive applications.

INTRODUCTION

The development of the global industry is currently growing along with the increasing need for materials for various purposes. The result of metal-based composite materials is in great demand because of their superior properties and ease of engineering [1,2]. Various engineering techniques were carried out to obtain composite materials with the best specifications. Lightweight material with optimal properties is a dream in various applied fields, especially the automotive and aerospace industry. The use of lightweight materials can reduce fuel consumption and improve performance which has an impact on environmentally friendly systems [1,3,4]

Currently, the danger of lead is a critical issue related to health and the environment. In 2020, Charkiewicz et al. reported that lead exposure could accumulate in the body and pose a risk of disease and disorders because lead is difficult to be excreted by the body [5]. The use of lead in machine element applications has received attention in recent years. Sakai et al. succeeded in synthesizing lead-free composites in bearing bush applications for piston pins with Cu6Sn3Ni1.5Mo2C composites as a substitute for Cu10Sn10Pb with high performance and meeting market requirements [6]. Development of alternative materials for the application of lead-free machine elements needs to be done to optimize the system and reduce the risk of exposure to hazardous substances. Aluminum matrix composites made by powder metallurgy techniques offer a solution. Aluminum has lightweight properties, corrosion resistance, non-magnetic, good thermal conductivity, low mechanical properties can be improved by alloying with hard ceramics such as SiC, Al_2O_3, or other elements such as zinc, silver, copper, magnesium [2,7]. The powder metallurgy method provides many advantages, including low process energy, product shape approaching the final shape, no wasted material during the process, high production speed with a more straightforward process route [4].

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Research in recent years has reported various methods and treatments for optimizing the properties of aluminum matrix composites through the powder metallurgy route. Gokce et al. conducted a study on AlCu0.5Mg alloy at a magnesium concentration of more than 0.15 %, causing an increase in the bond between particles and densification after the oxide was disrupted and the hardness value was 60 HV after sintering at 590 ºC for 1.5 hours [4]. Du et al., in their study, the pre-sintered treatment was given at 400 ºC, and the sintering temperature was 620 ºC on the Al-Cu (4.3 %)-Mg (1.2 %) composite, which the optimal conditions were reported at a holding time of 120 minutes with a maximum relative density value is 98.46 % and maximum hardness of 87.5 HBW [2]. Min et al. reported in the study of Al-Cu composites with Mg variation up to 2.5 %. It was reported that the addition of Mg concentration destroyed the oxide layer during sintering and increased the interfacial area between Al and Cu, which had an impact on improving the mechanical properties [8]. Many studies related to the optimization of wear resistance have been reported with materials that provide a self-lubricating effect. Akhlaghi et al. reported in a study of Al2024 composite processed by powder metallurgy technique, then the addition of 5 % graphite gave superior wear resistance properties under dry sliding conditions [9]. Narayan has also studied Mg-Graphite/MoS2 composites. It was reported that both materials at a composition of 5 - 10 % by weight of composites act as self-lubricating capable of reducing wear loss in tribometric tests [10].

The development of process technology supported by advanced equipment encourages the advancement of better composites. It is necessary to develop lightweight, lead-free and economical materials with optimized properties. This research focuses on the synthesis of aluminum matrix composites and the characterization of composite properties. Aluminum matrix composites with variations of copper powder were added to observe the behavior of the composites on physical, mechanical, and microstructure properties.

RESEARCH METHODOLOGY

Aluminum matrix composites are produced from high purity powder raw materials and processed by the powder metallurgical process route. Aluminum, magnesium, graphite, and copper powders were prepared. The raw powder properties are shown in Table 1. The composites were produced with an alloy composition of Al-1.5%Mg-5%Gr-x%Cu (0, 4, 7, and 10 %) weight of copper. The composition of the mixture with copper powder addition is shown in Table 2.

The raw material powder is weighed according to the weight fraction of the composite. Raw material powders were mixed on a high-speed mixer at 1500 rpm for 2 hours in a dry mixing method. Then, the powder mixture's uniformity was observed. Lubrication of the mold walls with liquid paraffin reduces friction and facilitates the sample ejection process. The homogeneous powder mixture was compacted using the cold uniaxial pressing method at a pressure of 150 MPa and held for 5 minutes using a Hydraulic Press Type 16T (Krisbow) machine. Furthermore, the green compact sample was sintered using a Lindberg Blue M Furnace (Thermo Scintentif, US) at a temperature of 600 ºC for 90 minutes. The sample was cooled in the furnace until it reached room temperature.

Characterization is done to see the behavior of the composite to the treatment given. The porosity and density tests were carried out using the Archimedes method to observe the physical properties of the composite. According to ASTM E92 standard, Hardness test with Vickers hardness tester using Wilson Hardness UH250 Buehler with a load of 5 kg and dwelling time of 15 seconds. The wear test refers to the ASTM G99 standard. The wear rate is investigated using the pin on disk test method with 0.4 m/s speed using #1000 grit abrasive paper on the disc. The metallographic process uses an acid solution (Keller Reagent) for etching. Micrograph observation using Laser Scanning Microscope Olympus Type OLS4100 to observe the microstructure of the composite.

TABLE 1. Properties of raw powder

Symbol	Purity	Shape, Average Size	Melting Point (ºC)	Density (g/cm^3)
Al	Aluminum powder, 96.8 %	Rounded, 45 μm	660	2.70
Mg	Magnesium powder, 98.87 %	Irregular, 100 μm	650	1.74
Gr	Graphite powder, 97.24 %	Angular, 55 μm	4,830	2.26
Cu	Copper powder, 97.45 %	Dendritic, 85 μm	1,083	8.95

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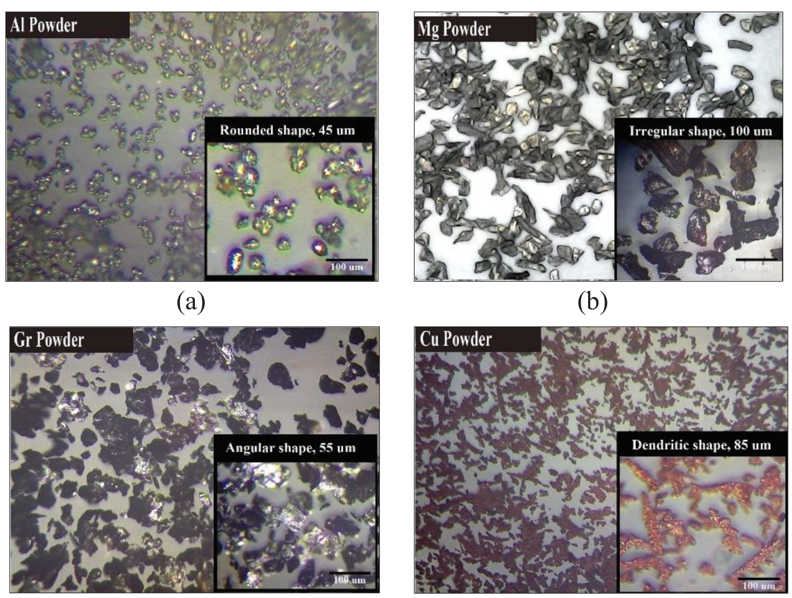
TABLE 2. Mixture powder composition

Sample	Composition	Cu Addition (x %wt)	Theoretical Density (g/cm^3)
C0	Al Pure (100% Al)	-	2.70
C1	Al-1.5%Mg-5%Gr-xCu	0	2.66
C2	Al-1.5%Mg-5%Gr-xCu	4	2.91
C3	Al-1.5%Mg-5%Gr-xCu	7	3.10
C4	Al-1.5%Mg-5%Gr-xCu	10	3.29

RESULT AND DISCUSSION

Powder selection is highly dependent on the needs of the final product. The powder form of the raw material is shown in Fig.1, then the homogeneity of the mixture was observed with an optical microscope. The supplied aluminum powder is rounded (45 μm), irregular shaped magnesium powder (100 μm), angular shaped graphite powder (55 μm), and dendritic copper powder (85 μm). Production powder particles with various methods, the characteristics of the shape and size of the powder are vital because they are related to the flowability of the compaction process and support the success of the sintering process [11]. The powders were mixed according to the composition. The distribution of constituent particles after mixing is shown in Fig.2.

The loose powder form is molded to form a solid sample (green compact) with 20 mm diameter and 7 mm thickness. The bond strength of the green compact relies on mechanical bonds and is still weak. During compaction, the distance between the particles is getting closer, so that there is an increase in density, the relative density of the sample obtained is at a value of 84.0 - 89.2 %, the Al-Cu-Mg powder mixture is optimally pressured in the pressure range of 100 - 500 MPa, compaction pressure is 100 - 200 MPa, or In the range of elastic deformation of the material, the relative density of the green compact is below 94 % [8]. Green compacts are brittle with low strength, and several pores are formed as a function of the shape and size of the constituent particles, so to achieve optimum properties, sintered treatment is required. The increase in temperature in the sintering process will be followed by forming adjacent particle bonds through a diffusion mechanism to increase the strength, density, ductility, and conductivity but shrinkage of the product geometry occurs [11]. The shape of the sample before and after sintering is shown in Fig.3.



(c) (d)

FIGURE 1. OM of raw powder preparation (a) Aluminum powder, (b) Magnesium powder, (c) Graphite powder, and (d) Copper powder

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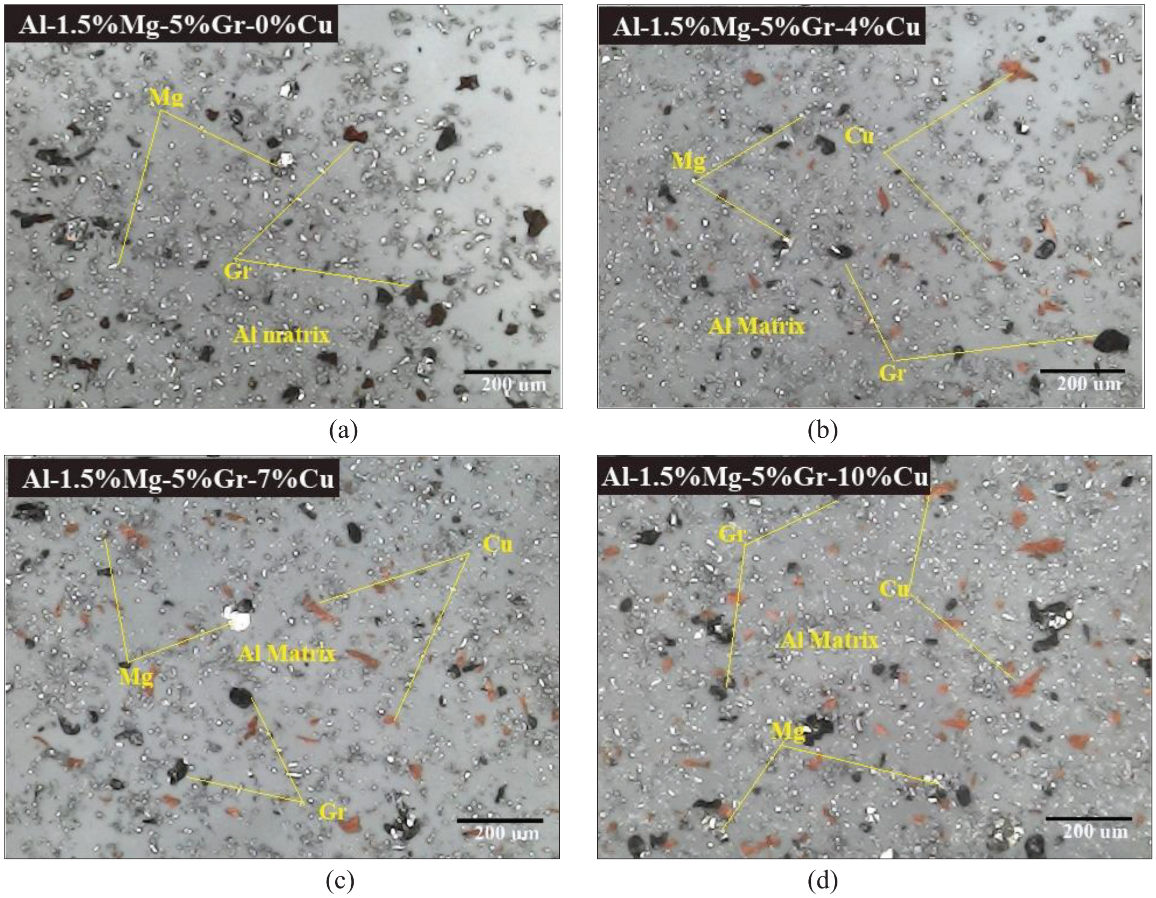

FIGURE 2. OM of mixture powder preparation (a) C1 mixture with 0%Cu, (b) C2 mixture with 4%Cu, (c) C3 mixture with 7 %Cu, and (d) C4 mixture with 10%Cu

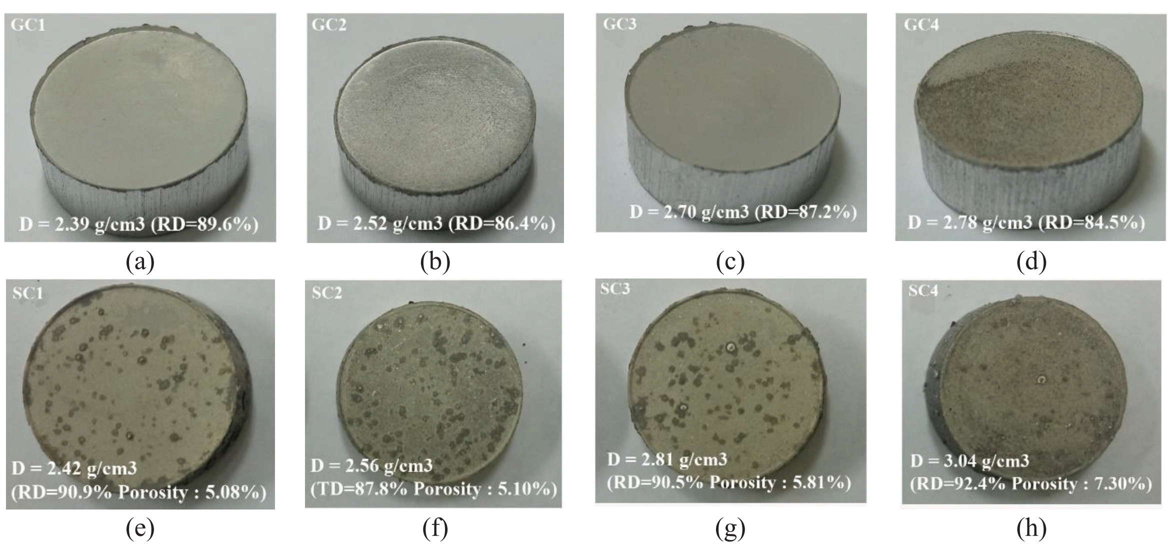

FIGURE 3. Sample of green compact (GC) and sintered compact (SC) (a) GC1 with 0%Cu, (b) GC2 with 4%Cu, (c) GC3 with 7%Cu, (d) GC4 with 10%Cu, (e) SC1 with 0%Cu, (f) SC2 with 4%Cu, (g) SC3 with 7%Cu, and (h) SC4 with 10%Cu

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Effect of Copper Addition on Physical Properties

Al-Mg-Gr precursor mixture with variations in the addition of copper affects the physical properties of the composite. Fig. 4 shows the density and porosity values of the post-sintered composite. The trend shown in Fig. 4a shows an increase in density with the addition of copper fractions in the composite. The lowest density in the Al-1.5%Mg-5%Gr-0%Cu composite was 2.42 g/cm^3, and the highest density was 3.04 g/cm^3 in the Al-1.5%Mg-5%Gr-10%Cu composite. Copper has a high density (8.95 g/cm^3) in the alloy, which plays a vital role in increasing the weight of the composite. A more significant copper fraction indicates a heavier composite.

The porosity level of the composite after sintering treatment is shown in Fig.5b. The values shown show a tendency to increase porosity in more significant copper fractions. The lowest porosity was seen in the Pure Al composite at 4.14 %, while in the Al-1.5%Mg-5%Gr-0%Cu composite, it was 5.08 %. The most significant porosity value was shown in the Al-1.5%Mg-5%Gr-10%Cu composite of 7.30 %. The porosity level that occurs can be related to the form factor and the size of the constituent particles. The process of compacting particles with a uniform shape and smaller size will result in closer particle spacing, the combination of compound constituent particles forming a wider inter-particle distance to trigger larger pores. The shape of spherical particles with a uniform size (monodisperse) is an ideal constituent condition, in packing spherical particles can fill 52 - 74% of the space achieved when the arrangement is close to a hexagonal shape, the space between particles will change with particle shape proportional to particle size [12]. In addition, the mechanism of pore formation has been reported in previous studies that pore formation is caused by the migration of several particles to the grain boundaries, a more prolonged sintering temperature dominates the diffusion of elements in a more uniform alloy, so that the migration of Cu and Mg in the Al phase triggers the accumulation of vacancies which increases the pores. [2]

Fig.5 shows the relative density values of the samples before and after sintering. The displayed values indicate a significant increase in sample density after sintering treatment. The relative density of the green compact obtained was 84.5 - 89.6 %, while the relative density of the post-sintered composite was 90.3 - 92.4 %. The increase in relative density due to sintering treatment in this study can be associated with the aluminum alloy sintering mechanism, which increased dramatically in the first 30 minutes due to shrinkage of pores, an interval of 30 - 120 minutes, relative density increased slowly due to grain growth, this increase will be followed by increased strength of composite [2]. The sintering density indicates how well the composite is sintered. The degree of geometric shrinkage occurs as a response to sintering at a temperature of 590 - 610 ^{o}C, which can increase the relative density [7].

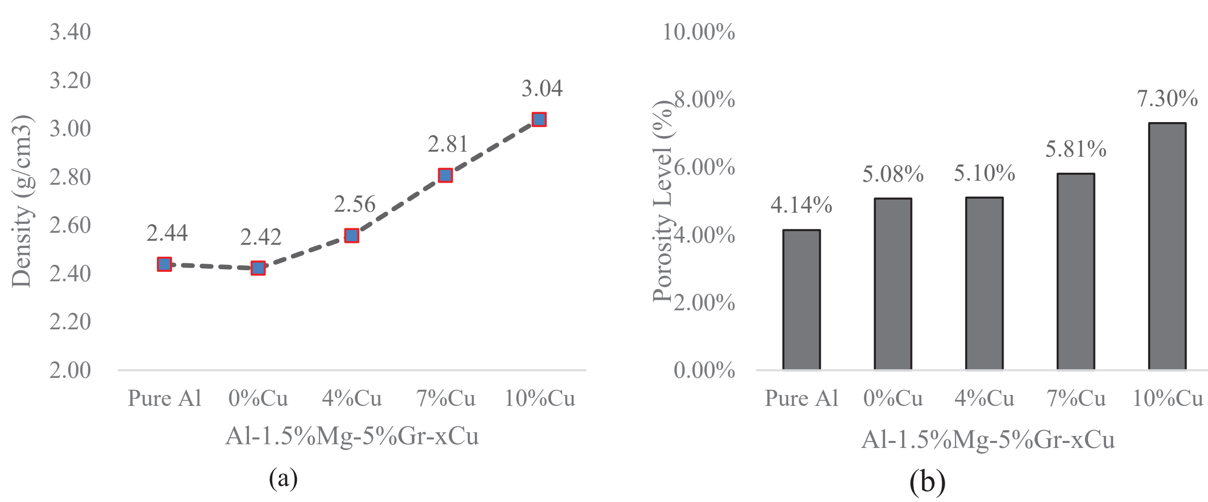

FIGURE 4. Physical properties of composite (a) Density and (b) Porosity level

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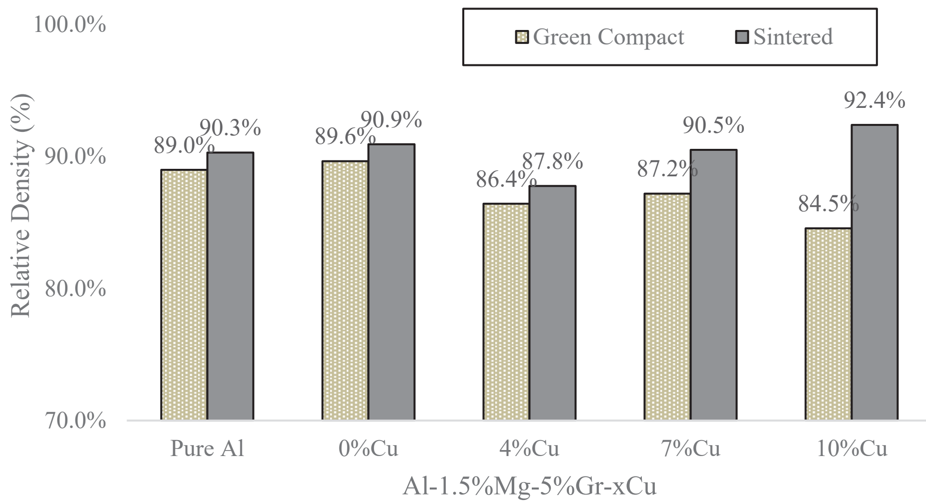

FIGURE 5. Relative density of green compact and sintered compact

Effect of Copper Addition on Mechanical Properties

The surface hardness of the post-sintered composite was tested by the Vickers hardness method. The test results are shown in Fig.6. The graph shows a pattern of the significant increase in hardness that is influenced by the addition of copper fractions. The increase in hardness is due to Mg and Cu particles in the Al matrix contributing to the formation of Al_2Cu precipitate in the more significant copper fraction. The mechanism of increasing the density of the green compact ensures positive contact of Al – Cu, the interdiffusion rate of Cu in Al is fast, $1.1x10^{-14}$ $m^2.s^{-1}$ at 450 ^oC and $2.1x10^{-13}$ $m^2.s^{-1}$ at 560 ^oC, when the temperature is reached, the oxide layer is destroyed by Mg, which increases the positive contact of Cu to diffuse in Al. A certain amount of liquid phase Al_2Cu precipitate will fill the particle boundary when the eutectic temperature is reached [2]. The highest hardness of 81.5 HV in the Al-1.5%Mg-5%Gr-10%Cu composite indicated the formation of more Al2Cu deposits in the more significant copper fraction. The lowest hardness value was 23.8 HV in the Al-1.5%Mg-5%Gr-0%Cu composite, but this value was still better than the Pure Al (16.1 HV) composite. Hardness increases with the addition of Mg to the matrix trigger the toughness nature of the alloy [13]. In this study, the hardness of the composite Al-1.5%Mg-5%Gr-10%Cu is 81.5 HV composite was lower than the Al4.3Cu1.2Mg hardness value 87.7 HB or 92.1 HV [2] and higher than the Al5Cu0.5Mg at 60 HV [4].

The results of the abrasive wear test are shown in Fig.7. The lowest volume loss rate indicates the best wear resistance. The wear resistance characteristic shows a relationship pattern with the composite hardness level. Wear resistance is indicated by a decrease in the wear rate due to the increase in the hardness of the aluminum matrix composite resulting from the contribution of reinforcing elements in the composite [3]. This condition can be attributed to the ability of the composite to resist particle delamination on the friction surface [14]. Composites with high hardness can withstand particle release better than composites with low hardness. In addition, the level of wear resistance is influenced by the graphite content in the composite. Previous studies reported that the wear resistance of 5 %wt graphite composites on the Al matrix showed the lowest wear rate in the dry sliding test. The graphite content in the composite triggered the formation of a graphite-rich thin layer on the mating surface, which reduced the wear rate [9]. The increase in test load shows an increasing trend of wear rate on all composites. The load variation is associated with an increase in the surface contact between the counter material and the sample surface, followed by an increase in the wear rate [15]. Composite Al-1.5%Mg-5%Gr-10%Cu with the highest hardness showed superior wear resistance in each variation of the addition of loads in this test.

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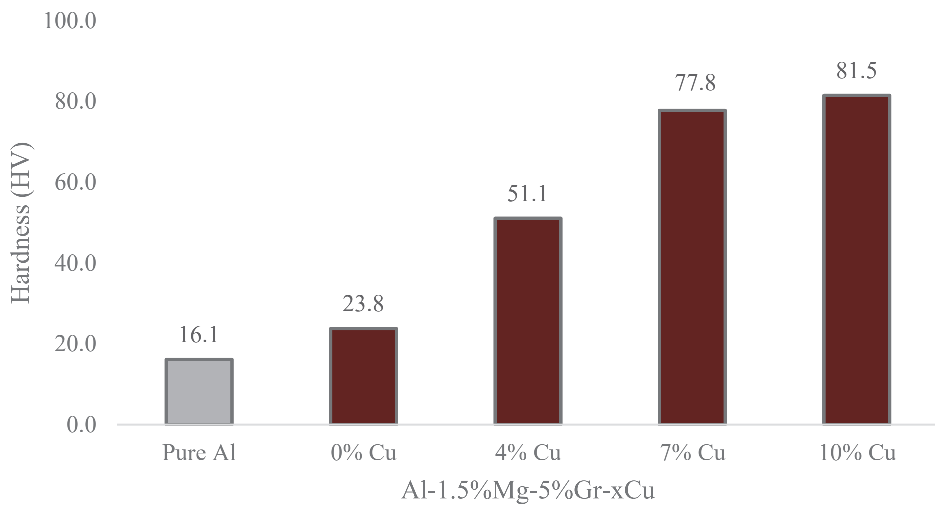

FIGURE 6. The hardness level of composite

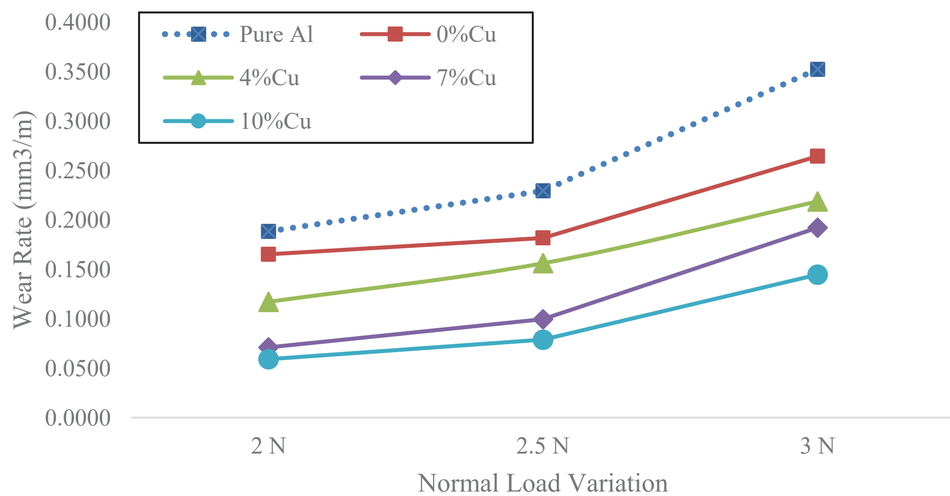

FIGURE 7. Wear rate level of composite

Effect of Copper Addition on Microstructure

Compaction is carried out at room temperature to achieve the shape, density, and contact between particles with sufficient strength to further processing. The micrographic of the green compact is shown in Fig.8. The figure shows the morphology of the green compact. It can be seen that the green compact is composed of sub-particles that are compacted during compaction. Particles appear to be deformed after the compaction process. The bonds between particles rely on mechanical bonds, and the distance between the particles is open. This condition indicates that the green compact sample has low strength and relative density. This study's relative density of green compacts was lowest in the Al-1.5%Mg-5%Gr-10%Cu composite (Fig. 8d), of 84.5 %, which could be observed with a more open distance between particles. In comparison, the highest relative density was found in the Al-1.5%Mg-5%Gr-0%Cu composite

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of 89.6 %, which was seen with slightly denser particle distances than the other samples. Sintering treatment on green compact is needed to improve the composite properties.

Figure 9 shows a micrograph of the composite after sintering. The figure shows that the shape of the enlarged particles is due to grain growth and pore shrinkage. This phenomenon occurs in the sintering process, the sintering temperature increases, and microstructural transformation occurs. Then, the contact between particles increases, the distance between particles, pores, and volume shrinks. The primary identity as a single particle disappears and begins the growth of new grains. The contact area between the particles becomes the grain boundary interface forming a polycrystalline microstructure. During the microstructural transformation, the density increases, and dimensional shrinkage occurs [12]. Sintering at high temperatures shows a fast grain growth rate, and many pores are shrunk with a longer holding time [16]. Figure 9a shows the shape of the composite micrograph without copper reinforcement, while Figures 9b, 9c, and 9d show the composite micrograph with copper reinforcement. The copper matrix and reinforcement form a good bond at the interface area. It is seen that the distance between the particles is closer, indicating an increase in density, then a reduction of pores followed by an increase in mechanical properties. Cu-Al particles are in positive contact at the grain boundaries, indicating the contribution of Mg as a wetting agent which decays the oxide during sintering. The wetting mechanism increases the sintering of the liquid phase through solid capillary forces between the liquid and solid substrates, which increase the interface area [17,18]. Interdiffusion of Cu on Al will initiate the formation of Al_2Cu deposits at the interface area, increasing the strength of the composite [16]. Agglomeration of copper particles in the Al-1.5%Mg-5%Gr-10%Cu composite (fig.9d) due to a high fraction with a deviation of the distribution, as reported in the previous study that the system with smaller particles, a significant deviation of particle distribution, lower viscosity at higher sintering temperature triggers more substantial particle agglomeration [19]. Several dominantly dispersed graphite particles at the grain boundaries act as self-lubricating, reducing the wear rate of the composite.

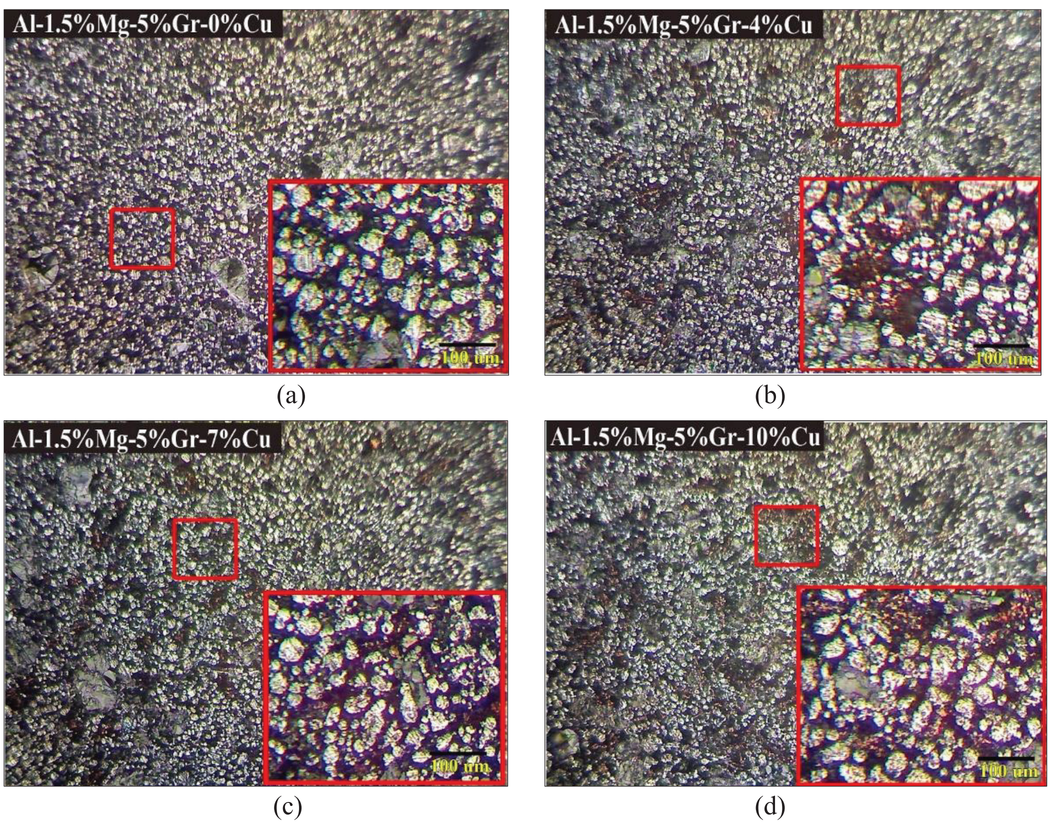

FIGURE 8. Optical micrograph of green compact (GC) (a) GC1 with 0%Cu, (b) GC2 with 4%Cu, (c) GC3 with 7%Cu, and (d) GC4 with 10%Cu

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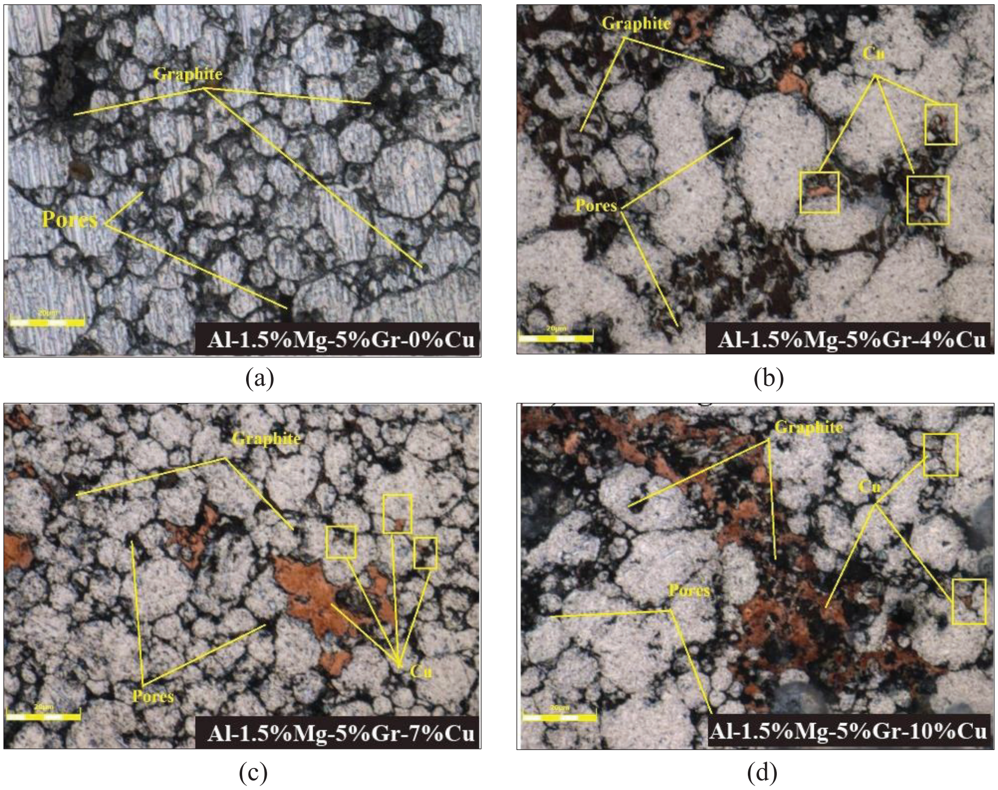

FIGURE 9. Optical micrograph of the sintered compact (SC) (a) SC1 with 0%Cu, (b) SC2 with 4%Cu, (c) SC3 with 7%Cu, and (d) SC4 with 10%Cu

Bushing products are available in various shapes and sizes in the global market. Bearing manufacturers Jiashan DYB Bearing Co. Ltd and CSB Bearings Industry provide a wide range of composite bearing products for automotive applications. According to SAE J460 [20], the available lead composites include CuSn10Pb10, CuSn4Pb24, CuPb24Sn, and CuPb30 with a density value of 10.72 g/cm^3 and minimum hardness at 30 HB (32 HV) [21]. In this study, the properties of aluminum matrix composites with 4, 7, and 10 % copper reinforcement met the minimum hardness values and were 3.5 times lighter than lead composites available in the global market.

CONCLUSION

Copper-reinforced aluminum matrix composite has been successfully synthesized by the powder metallurgy method. Observations of physical properties, mechanical properties, and microstructure have been investigated through experiments. The results can be concluded as follows:

- The addition of copper affects the physical properties. The addition of a more significant copper fraction increased the density and porosity of the composite. Besides that, the sintering treatment was able to increase the relative density of the composite. The highest density value is 3.01 g/cm^3, the porosity value is 7.3 %, and the relative density value is 92.4 %, were obtained in the Al-1.5%Mg-5%Gr-10%Cu composite.
- The addition of copper affects the mechanical properties. The addition of a more significant copper fraction can increase the hardness of the composite, which is followed by good wear resistance. The highest hardness value is 81.5 HV, with the best wear resistance was found in the Al-1.5%Mg-5%Gr-10%Cu composite.
- The microstructure shows the distribution of the dominant constituent particles at the grain boundaries. Shrinkage of the pores after sintering, followed by a closer bond between grains, indicates an increase in mechanical properties.
- The lead-free aluminum matrix composite properties with 4, 7, 10 % copper reinforcement promise alternative materials to automotive applications.

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