Effect of Mechanical Properties with Addition of Graphene Nano-Platelets in Cu/GNPs Composite- Green Part

Muhammad Omar Abdul Rashid*, Norhamidi Muhamad, Abu Bakar Sulong, Nor Nabilla Kadiman Department of Mechanical and Materials Engineering, Faculty of Engineering & Built Environment, Universiti Kebangsaan Malaysia, Malaysia

*Corresponding author: m4d0m4r@gmail.com

Received 18 October 2019, Received in revised form 25 June 2020 Accepted 1 July 2020, Available online 30 November 2020

ABSTRACT

Graphene is one of the best reinforcement materials but have a strong tendency to agglomerate and will affect the final properties of the composite. Effect of mechanical properties with addition of Graphene Nano-platelets (GNPs) in copper through powder injection molding (PIM) process was investigated. 2 feedstocks was prepared, one is Cu 100% and Cu/0.5 vol.% GNPs. Both feedstocks use same powder loading, 62% powder and 38% binder and the binder system used for this study consist of polyethylene glycol (PEG), polymethyl methacrylate (PMMA) and stearic acid (SA). The feedstocks were sonicated, and ball milled using the same parameters and then continue with until injection process. The produced green part strength and density were tested. The main reason to for density testing is to determine either addition of GNPs will affect the composite density The average green density for Cu 100% is 5.835 g/cm3, which is 99.84 % from the theoretical density. Meanwhile for Cu/GNps 0.5%, the average density is 5.874 g/cm3, which is 99.91 % from the theoretical density. For the green strength, the average maximum flexural strength for Cu 100% is 15.29 MPa and 14.32 MPa for Cu/0.5 vol.% GNPs. Even though the density of Cu/GNPs composites decrease with addition of 0.5 vol% GNPs but both sample strength indicates good mechanical properties and suitable for debinding and sintering process.

Keywords: Copper, Graphene, powder injection molding, flexural strength

INTRODUCTION

With the current increased demand for higher performance electronic product in terms of speed and graphic, higher thermal conductivity and heat dissipation component is needed to support the merging of new devices (Saboori et al. 2018). Most of the traditional metal and alloys cannot meet the requirement of this device. Thus the solution is to develop metal matrix composite (MMCs) (Qu et al. 2011). Commonly, Aluminium (Al) and Copper (Cu) been chosen as metal matrix because of their good thermal conductivity and reinforcement used were ceramic, carbon and diamond (Sidhu, Kumar, and Batish 2015). Among other reinforcement, graphene is considered a perfect reinforcement for MMCs due to its exceptional properties: high thermal conductivity, high young modulus and high tensile stress (Hu et al. 2016). Copper (Cu) nanocomposite was chosen due to its enhanced mechanical, electrical and thermal properties that were leading to numerous electronic application (Pavithra et al. 2014). Copper is more difficult to extrude, stamp, cast or machine but it is more

commonly processed with powder metallurgy techniques (Johnson et al. 2005). There are several techniques in powder metallurgy; some of them are metal injection molding (MIM), hot isostatic pressing (HIP), powder forging (PF) and powder injection molding (PIM).

PIM is one of the favourite technique due to its near-net shape forming technology with suitable to producing small parts with complex shape (Qu et al. 2011)(Emeka et al. 2017). PIM also has the potential to produce high in volume at low cost while can meet the geometry requirement (Johnson et al. 2005). Furthermore, through PIM process, the product already achieves the final form and not need further machining process (Md Ani et al. 2017). Basically there are four steps in PIM, the process start with mixing metal powders with binders to form a feedstock, then injection molding of green part, after that debinding process consists of solvent and thermal that purposely to extract the binder from material and lastly will be sintering process to obtain the final material (Ramli et al. 2019). Polyethylene glycol (PEG) is the most extensively reported main binder used in previous

research due to its non-toxicity and commercial availability (Hayat et al. 2017). Polymethyl methacrylate acrylic (PMMA) often used as a secondary binder in PEG-based binder system due to its excellent compatibility (Hayat et al. 2015). To improve powder wetting, stearic acid (SA) often used as surfactant and lubricant (Irwan Ibrahim et al. 2011).

Most of the recent material used for thermal application is a combination of two or three material due to traditional metals and cannot meet the current requirement of thermal application products (Hu et al. 2016). Moreover, until now true potential of graphenes are yet discovered. The processing and dispersion method constantly upgrading thus there still need for promising methods for fabrication. Most common processing techniques for GNPs composites are semipowder metallurgy (SPM), molecular-level mixing (MLM), electrochemical deposition (ELD), spray forming (SF), powder injection molding (PIM) and selective laser melting (SLM). (Naseer et al. 2019). Yue et al. 2017 fabricated GNSs/CU using ball milling and hotpress sintering, reported that on 0.5 wt% the UTS is highest and decreasing with addition of more GNSs. To date, there are not many reports conducted Cu reinforced with graphene through the PIM process. This study is focusing on the effect of mechanical properties with the addition of GNPs in Cu/GNPs composite and the moldability of the composites

METHODOLOGY

Materials

Cu used for this study is a gas atomized copper powder and was supplied from Sandvik Materials Technology. Based from particle size analyzer, average powder size for D (0.1) is 11.784 µm, D (0.5) is 38.194 µm and for D (0.9) is 103.817 µm. The bulk density of Cu powder is 8.93 g/cm3. While the fillers used for this study is Graphene Nanoplatelets (XGNp - M grade) and was supplied by XG Sciences, Inc. Based from particle size analyzer, average powder size for D(0.1) is 6.423 μ m, D(0.5) is 14.598 μ m and for D(0.9) is 418.881µm. According to manufacturer datasheet, bulk density for GNps is 2.2 g/cm3. FESEM micrograph of Cu and GNps are shown in figure 1, it shows that Cu powder is spherical, meanwhile GNps powder are in flake shapes.

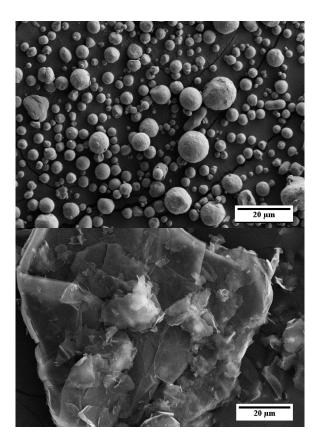


FIGURE 1. FESEM micrograph of (a) Copper powder (b) Graphene Nanoplatelets

Methods

The sample was prepared accordingly from the premixing, mixing and injection process. The process starts with the pre-mixing process. Sonication and ball milling methods were used in the pre-mixing process and its functioned to disperse the GNps (Kadiman et al. 2018), where the GNps were sonicated at 55°C for 1 hour in distilled water, the sonicated GNps were left dried in oven for 18 hours at temperature 100°C in order to remove any moisture on the sample. After that, the dried GNps was mixed with Cu powder using planetary ball mill Pulverisette 6 with milling speed of 100 rpm for 4 hours. Dispersion process is essential for nano-filler materials to prevent agglomeration in the next process (German 2012). Next, the ball milled GNps was mixed with Cu powder and binders at 150 °C for 1 hour using a Brabender mixer. Both feedstocks use same powder loading, 62% powder and 38% binder and the binder system used for this study consist of polyethylene glycol (PEG), polymethyl methacrylate (PMMA) and stearic acid (SA). Selection of binder system is important to control the homogeneity level of feedstocks and control the quality of injection molding output (Ismail, Muhammad, and Omar 2008). The feedstock was then injected into a tensile bar using Boy 22A injection molding machine.

The parameter used to inject both feedstocks are same, mold temperature is set to room temperature or 30°C, holding time is 7 sec and injection pressure is 8 bar. Next is to set injection temperature, the temperature for the nozzle is set to 170°C, 125°C for the front, 120°C for the middle, 115°C for the rear 1 and 110°C for rear 2. Figure 2 shows the sample of 100 vol.% Cu and Cu/0.5 vol.% GNPs green part.

The density of both green parts were determined by using Archimedes method based on MPIF standard 42. The measured density will be compared to the theoretical density in order to determine the different percentage. The theoretical density is measured by added weight percentage of powder and binder use with respect of the weight of the sample. Next the green part will undergo flexural strength test using three-point bending test. The three-point bending are conducted in line with MPIF standard 15. Each specimen of both samples was supported transversely to the load over a span of 30mm and the mid-span was loaded gradually until fracture of the specimen. A crosshead velocity of 0.2mm/min was utilised. The average width and thickness of the samples were 5.66mm and 3.49mm respectively.

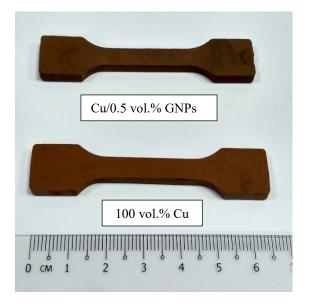


FIGURE 2. Cu 100% and Cu/0.5 vol.% GNPs greenpart

RESULT AND DISCUSSION

Figure 3 shows scanning electron microscopy (SEM) for 100 vol.% Cu and Cu/0.5 vol.% GNPs feedstock. The optical zooming is 2.k and the size is 30um. From the SEM image, it shows the evidence of well-dispersed powder particles in the binder matrix indicating a good mixture of feedstock.

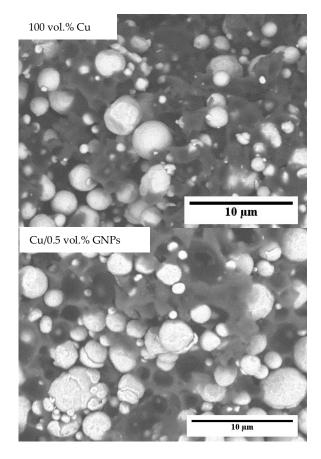


FIGURE 3. SEM for 100 vol.% Cu and Cu/GNps 0.5% feedstock

Furthermore, SEM image as shown in figure 4 also indicated a good mixture and compacted sample for both 100 vol.% Cu and Cu/GNps 0.5% green part.

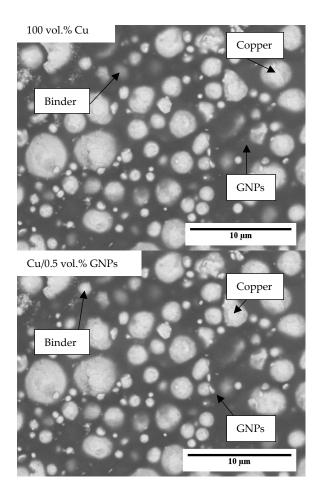


FIGURE 4. SEM for 100 vol.% Cu and Cu/GNps 0.5% green part

The density and green strength test have been done in this study. Table 1 shows the density result of 100 vol.% Cu and Cu/0.5 vol.% GNPs green part. The average green density for 100 vol.% Cu is 5.835 g/cm3, which is 99.84 % from the theoretical density.

 TABLE 1. Measured and theoretical density of 100 vol.% Cu and Cu/0.5 vol.% GNPs green part

Materials	Sample	Density (g/cm ³)	Theoretical Density (g/cm ³)	Different (%)
100 vol.% Cu	1	5.868	5.989	0.129
	2	5.839		0.15
	3	5.877		0.112
Cu/0.5 vol.% GNPs	1	5.823	5.956	0.136
	2	5.903		0.056
	3	5.876		0.076

Meanwhile for Cu/0.5 vol.% GNPs, the average density is 5.874 g/cm3, which is 99.91 % from the theoretical density. Density of Cu/GNPs composites decrease with addition of 0.5 vol% GNPs, this is due to the relative density of graphene (2.2 g/cm³) are lower than copper (8.9 g/cm³) (Hidalgo-Manrique et al. 2019). From the measured density, the value is nearly the same to the theoretical density; these indicate that binder and composite materials are uniformly mixed.

Green strength refers to the mechanical strength of the parts where a compacted powder should have to withstand mechanical operation and handling, it is subjected after pressing and before sintering, without damaging its details and sharp edges (Irwan Ibrahim et al. 2011). Total three specimen were tested for each sample. The average maximum flexural strength for 100 vol.% Cu is 15.29 MPa and 14.32 MPa for Cu/0.5 vol.% GNPs, as shown in figure 5. The flexural strength of both is above 5MPa which according to (German and Bose 1997) is allowable for mechanical strength of injection molded parts.

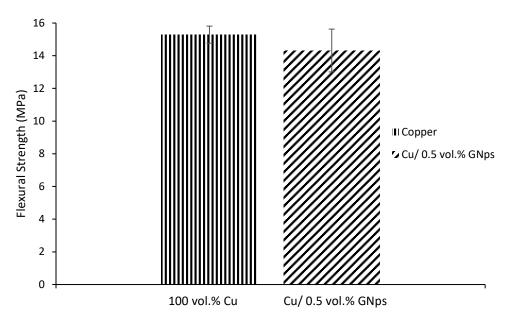


FIGURE 5. Flextural Strength for 100 vol.% Cu and Cu/ 0.5 vol.% GNps

CONCLUSION

In summary, the density and strength of both 100 vol.% Cu and Cu/ 0.5 vol.% GNps green part was determined. The main reason to for density testing is to determine either addition of GNPs will affect the composite density. Even though the density of Cu/GNPs composites decrease with addition of 0.5 vol% GNPs but both samples density is nearly similar to the theoretical density, where 100 vol.% Cu is 99.84% and Cu/ 0.5 vol.% GNps is 99.91% compared to their theoretical density. In term of green strength, for 100 vol.% Cu the average flexural strength is 15.29 Mpa and Cu/ 0.5 vol.% GNps is 14.32 Mpa. Both sample strengths are over 5 MPa which indicates good mechanical properties and suitable for next processing steps such as debinding and sintering. Furthermore, both green parts were defect free, and SEM evaluations of both samples revealed good mixing and compaction of the materials.

ACKNOWLEDGMENTS

The authors would like to thank the Universiti Kebangsaan Malaysia for the financial support from grants FRGS/1/2015/TK03/UKM/1/3 and MyBrain15 programme.

DECLARATION OF COMPETING INTEREST

None.

REFERENCES

- Emeka, U.B., Sulong, A.B., Muhamad, N., Sajuri, Z. and Salleh, F. 2017. "Two Component Injection Moulding of Bi-Material of Stainless Steel and Yttria Stabilized Zirconia – Green Part." Jurnal Kejuruteraan, 29 (1): 1–9.
- German, R. M. 2012. Metal Powder Injection Molding (MIM): Key Trends and Markets. Handbook of Metal Injection Molding. 2nd ed. Elsevier Ltd.
- German, R M, and A Bose. 1997. *Injection Molding* of Metals and Ceramics. Metal Powder Industries Federation.
- Hayat, M.D., Goswami, A., Matthews, S., Li, T., Yuan, X. and Cao, P., 2017. Modification of PEG/PMMA binder by PVP for titanium metal injection moulding. *Powder Technology*, 315:243-249.
- Hayat, M.D., Li, T., Wen, G. and Cao, P., 2015. Suitability of PEG/PMMA-based metal injection moulding feedstock: an experimental study. *The International Journal of Advanced Manufacturing Technology*, 80(9-12): 1665-1671.
- Hidalgo-Manrique, P., Lei, X., Xu, R., Zhou, M., Kinloch, I.A. and Young, R.J., 2019. Copper/graphene composites: a review. *Journal of Materials Science*, 2019: 1-54.
- Hu, Z., Tong, G., Lin, D., Chen, C., Guo, H., Xu, J. and Zhou, L., 2016. Graphene-reinforced metal matrix nanocomposites-a review. *Materials Science and Technology*, 32(9): 930-953.
- Ibrahim, M.H.I., Muhamad, N., Sulong, A.B., Jamaludin, K.R., Mohamad Nor, N.H. and Ahmad, S., 2011. Optimization of Micro

Metal Injection Molding SS 316L For The Highest Green Strength By Using Taguchi Method. In *Advanced Materials Research*, 264: 135-140.

- Ismail, M.H., Muhamad, N. and Omar, M.A., 2008. Characterization of metal injection molding (mim) feedstock based on water soluble binder system. *Jurnal Kejuruteraan*, 20: 11-18.
- Johnson, J.L., Tan, L.K., Bollina, R., Suri, P. and German, R.M., 2005. Evaluation of copper powders for processing heat sinks by metal injection moulding. *Powder Metallurgy*, 48(2): 123-128.
- Kadiman, N.N., Romli, J.E., Muhamad, N., Sulong,
 A.B. and Mohd Foudzi, F. 2018.
 Pengoptimuman Parameter Sonikasi Dan
 Pengacauan Magnetik Bagi Mendapatkan
 Penyerakan Sebati Komposit Kuprum-Grafin
 Berdasarkan Sifat Morfologi. Sains
 Malaysiana 47(5): 1039–43.
- Md Ani, S., Muchtar, A., Muhamad, N. and A. Ghani J. 2017. Kesan Suhu Pensinteran Terhadap Sifat Mekanik Dan Mikrostruktur Alumina-Zirkonia Yang Difabrikasi Dengan Kaedah Pengacuan Suntikan Seramik. *Sains Malaysiana*, 46 (10): 1979–86.
- Naseer, A., Ahmad, F., Aslam, M., Guan, B.H., Harun, W.S.W., Muhamad, N., Raza, M.R. and German, R.M., 2019. A review of processing techniques for graphene-reinforced metal matrix composites. *Materials and Manufacturing Processes*, 34(9): 957-985.
- Pavithra, C.L., Sarada, B.V., Rajulapati, K.V., Rao, T.N. and Sundararajan, G. 2014. A New Electrochemical Approach for the Synthesis of Copper-Graphene Nanocomposite Foils with High Hardness. *Scientific Reports* 4: 4049.
- Qu, X.H., Zhang, L., Mao, W.U. and Ren, S.B. 2011. Review of Metal Matrix Composites with High Thermal Conductivity for Thermal Management Applications. *Progress in Natural Science: Materials International* 21 (3): 189–97.
- Ramli, M.I., Sulong, A.B., Muhamad, N., Muchtar, A. and Zakaria, M.Y. 2019. Effect of Sintering on the Microstructure and Mechanical Properties of Alloy Titanium-Wollastonite Composite Fabricated by Powder Injection Moulding Process. *Ceramics International*.
- Saboori, A., Dadkhah, M., Fino, P. and Pavese, M. 2018. An Overview of Metal Matrix Nanocomposites Reinforced with Graphene Nanoplatelets; Mechanical, Electrical and Thermophysical Properties. *Metals*, 8 (6).
- Sidhu, S.S., Kumar, S. and Batish, A. 2015. Metal Matrix Composites for Thermal Management: A Review. *Critical Reviews in Solid State and Materials Sciences*, 41 (2): 132–57.

Yue, H., Yao, L., Gao, X., Zhang, S., Guo, E., Zhang, H., Lin, X. and Wang, B. 2017. Effect of Ball-Milling and Graphene Contents on the Mechanical Properties and Fracture Mechanisms of Graphene Nanosheets Reinforced Copper Matrix Composites. Journal of Alloys and Compounds, 691: 755– 62.