EFFECT OF T6 HEAT TREATMENT ON MICROSTRUCTURE AND HARDNESS PROPERTIES OF THIXOFORMED GRAPHENE-REINFORCED A356 COMPOSITES

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ABSTRACT: This study reports the development of graphene nanoplatelets reinforced A356 alloy matrix composites. In this investigation, a graphene nanoplatelet reinforced A356 alloy matrix composite was fabricated using stir casting and thixoforming processing, followed by T6 heat treatment. The microstructure and hardness characteristics of alloys and composites were analyzed using optical microstructure, field emission scanning electron microscopy, and microhardness testing. The outcomes show that mechanical stir casting can produce a non-dendritic structure in the composite. The microhardness of cast composites increased by 35.8% from the base alloy sample due to the graphene addition to the base alloy, which also resulted in grain refinement in the structure of the composite. The microhardness of composites is improved by 51.4 % and 70.5 %, compared to the A356 alloy after undergoing the thixoforming process and T6 heat treatment. The field emission scanning electron microscopy shows the coarsened eutectic Si particles, and the Si particles become more spheroids after the T6 heat treatment. The thixoformed composite also shows an increase in density and

treatment. The thixoformed composite also shows an increase in density and less porosity after the T6 heat treatment compared to the base alloy and cast composite. The subsequent T6 heat treatment technique significantly enhanced the composite hardness owing to the incorporation of graphene and spheroidization of eutectic Si particles.

KEYWORDS: *Composite microstructure; Density, Thixoforming; T6 heat treatment; and Hardness properties*

1.0 INTRODUCTION

Aluminium matrix composites are broadly employed in industries owing to their outstanding strength, excellent resistance to wear, and high-temperature durability applications, especially in the automotive field [1], [2]. Its benefits have led to its increased implementation, and currently, it has become a potential metal matrix composite for usage in various industries [3]. However, as a result of advances in technology and science development, the demand for the performance requirements of aluminium matrix composites in all prospects is expanding in related fields.

The conventional particle as reinforcing phase and ceramic fibre [4] no longer fulfil the advanced requirements of industrial technology development because of the existence of limitations in material performance. In recent decades, graphene has drawn a significant amount of interest from researchers owing to its exceptional mechanical properties, intrinsic thermal, and electrical; in addition, graphene possesses a high specific surface area [5], [6]. Therefore, it is recognized as the appropriate reinforcement material for high-performance aluminium matrix composites [7]. The structure of the interface between the metal matrix and graphene as reinforcement material was also investigated and controlled in order to enhance the interfacial adhesion [8]. Moreover, numerous studies have attempted to investigate the impact of graphene-reinforced aluminium matrix composites on strength [1], [8]–[10]. For instance, Borand and Uzunsoy [11] reported the fabrication of 0.7 wt.% graphene/Al-4.5Cu, increasing the hardness by 37.11%.

While Ghazanlou et al. [12] added 2.0 wt.% of graphene, hardness was increased to 93% compared to pure Al due to an enhanced strengthening mechanism. Nonetheless, the observation reported that the hardness started to decline beyond 2.0 wt.% of graphene. Liquid state methods like casting are acceptable for developing composites owing to their flexibility, large-scale production capacity, economics,

and nearly net shaping [13]. However, due to the difference in density between aluminium and graphene, graphene particles are attracted to float at or close to the surface layer of the molten liquid. Additionally, the weak wettability between the aluminium matrix and graphene can prevent the interaction of the interfacial and reduce the mechanical characteristics of the composite, resulting from the agglomeration of graphene in the matrix [14]. In contrast, stir casting is employed to manufacture casting parts to reduce porosity, and improve homogeneity, resulting in outstanding mechanical qualities [15].

Nevertheless, methods combining different processing steps have been proposed to improve and enhance strength, such as semi-solid metal (SSM) processing. Thixoforming is one type of SSM where the process begins with reheating the billet of feedstock with a non-dendritic structure to a semi-solid temperature and then delivering the billet using the required hydraulic pressure into the die [16]. In addition, thixoformed components provide better mechanical properties and near-net shaping of complicated geometries compared to conventionally cast parts [17], [18]. Furthermore, the employment of heat treatment techniques helps promote the stability of the composite and its mechanical properties, which can be used in some applications that require certain criteria [19]. In addition, heat treatment significantly influences the modification of the characteristics and microstructure in accordance with the specified function [15], which requires solution treatment, quenching, and artificial ageing.

The primary objective of solution-treating composite materials is to enhance their strength by creating a more consistent arrangement of structure, dissolving the brittle non-equilibrium particles, and generating supersaturated solid solutions. Artificial ageing is the regulated disintegration of supersaturated solid solutions, creating finer diffused precipitates [20]. Hence, the objective of this study is to fabricate a graphene reinforced A356 aluminium alloy composite using mechanical stir casting. The samples underwent the thixoforming process and T6 heat treatment technique to enhance their hardness properties. Furthermore, the effect of each stage on the microstructure, density, and hardness properties of alloy and composite were investigated.

2.0 EXPERIMENTAL PROCEDURE

This study used anA356 alloy ingot as presented in Table 1. Graphene nanoplatelets with a 500 m2/g surface area from Sigma-Aldrich was employed as reinforcement material. The composites were prepared by

placing 400 g of A356 alloy in a graphite crucible and heating it to 700 °C in an induction furnace. Then, the molten temperature was decreased to 650 °C and remained constant for 5 minutes. In addition, GNP was preheated to remove the moisture. The mixture of preheated GNP (0.3 wt.%) and Mg powder (1 wt.%) was wrapped in aluminium foil and injected into the melted alloy at the base of the crucible by a steel plunger. Magnesium (Mg) powder was supplemented to the molten mixture in order to reduce the poor wettability of graphene in the melt.

The molten liquid was mechanically stirred for 5 minutes at 500 rpm with a three-blade propeller to incorporate the reinforcement into the base alloy. The tablets of hexachloroethane were added to the melt after it had been stirred to degas the melt. The molten mixture was poured into a preheated mould (150 °C) to produce a feedstock of composite as a billet. Then the composite billet was thixoformed using a hydraulic cylinder press that has a maximum compression velocity of 85 mm/s with a load of 20 kN. Next, the billets were immediately reheated at a rate of 130 °C/min until the temperature reached 581 °C at 50% liquid fraction, as shown in Figure 1. The reheating process was operated using a 30-80 kHz, 35 kW high-frequency induction coil stationed beneath the die, and the temperature of the composite billet was monitored by a K-type thermocouple. Subsequently, the thixoformed samples were subjected to the T6 heat treatment in an effort to enhance their properties. The thixoformed composite underwent solution treatment at 540 °C for 8 hr and was quenched directly in 60 °C of warm water. The completion of a cycle in T6 heat treatment involved artificial ageing, where the quenched samples were required to age at 160 °C for 4 hr and cooled with air temperature.

Table 1: Averaged chemical composition of A356 alloy used in wt.%

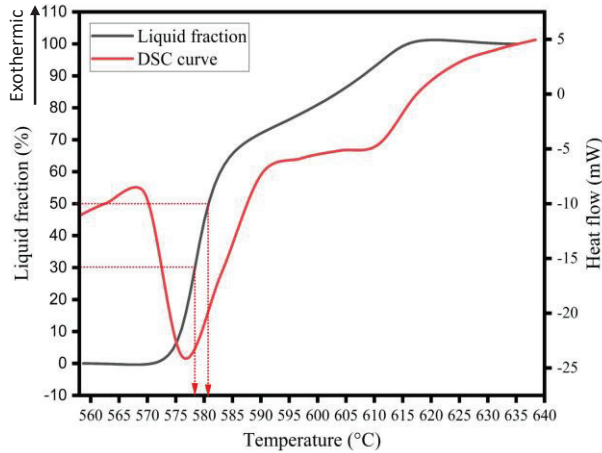| Si	Mg	Fe	Cu	Mn	Zn	Al
7.50	0.250	0.425	0.045	0.185	0.0035	Balance

Figure 1: The curve of liquid fraction and DSC of composite.

The microstructural characterization and distribution of GNP of as-cast, thixoformed and heat-treated composites were performed by Nikon optical microscope and FESEM (ZEISS Sigma 500, Germany) after grinding (400, 600, 800, and 1200 grits), polishing with diamond solution (6, 3 and 1 μ) and etching with Keller's reagent solution. Microhardness measurements (ASTM E384-17) were obtained using the VH testing Shimadzu machine (test load = 9.807 N and dwell time = 10 s). Ten measurements were conducted to get reliable data for hardness.
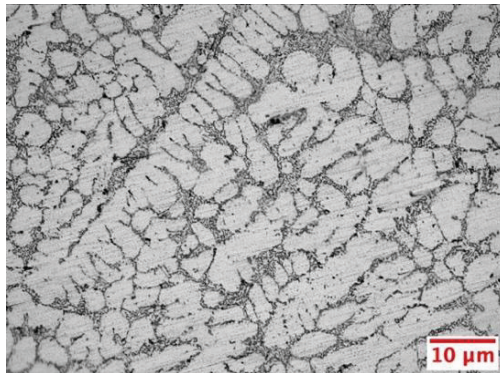

Figure 2: Optical microstructure image of as-received A356.

3.0 RESULTS AND DISCUSSION

3.1 Microstructure of the composite

The GNP-A356 composites were fabricated by mechanical stir casting, followed by thixoforming and T6 heat treatment. According to Figure 2, there is a common dendritic α-Al structure along with interdendritic

eutectic microstructures for the as-received A356 alloy after being poured at 650 °C. Figures 3(a) and 3(b) demonstrate the formation of globular and non-homogeneous rosette-like of α-Al phase after the stirring operation for A356 alloy and GNP-A356 composite, respectively.

The dendritic arms were broken using a blade driven by an external mechanical force, and α-Al islands developed. However, the addition of reinforcement has increased the heterogeneous nucleation in the aluminium melt [9] and has helped in refining the microstructure of the composite, as shown in Figure 3(b) compared to Figure 3(a). As the composite solidifies, GNP is restricted from moving in the direction of globular and rosette-like α-Al grains. Furthermore, the irregular and non-uniform shape of the α-Al might also have occurred because of the existence of GNP nanoparticles in the alloy.

Hanizam et al. [21] revealed that the development of a homogeneous globular microstructure was disrupted during stirring as a result of the turbulent flow in the molten metal. However, this outcome was acceptable for the subsequent thixoforming process. Furthermore, Figure 3(c) demonstrates the microstructures of the thixoformed composite. It reveals that heating the billet at 50% liquid fraction resulted in the development of globules and the coarsening of the morphology of the α-Al. The α-Al particle remained solid at a reheating temperature of 50% liquid fraction, while the eutectic Si melted during reheating and repositioning, forming plate-like structures around the α-Al globules [22], [23]. In addition, some of the deposition processes of Si and Al on the α-Al and primary Si particles occurred during reheating in the semi-solid state and generated the process of coarsening in the α-Al particles.

Furthermore, the T6 heat treatment has exposed some microstructure alteration to the treated composite, as shown in Figure 3(d), where the plate-like and needle-like eutectic Si become coarsened and transformed to spheroid-like. In addition, it is well known that Si atoms diffuse more rapidly than Al atoms during solutionizing [25], and some dispersed fine Si particles were precipitated in the α-Al matrix.
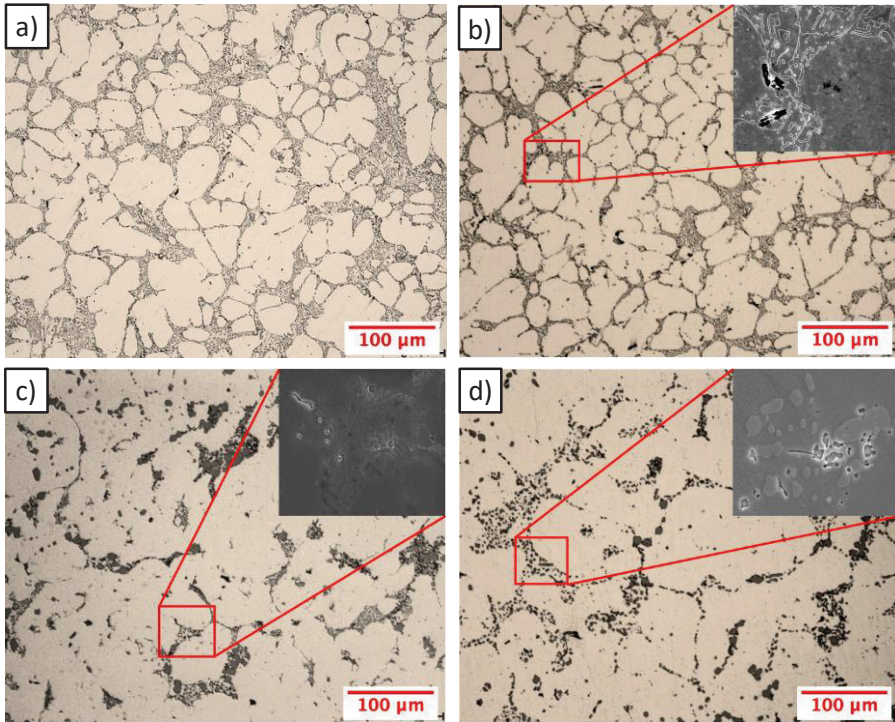
Figure 3: Optical microstructure images of (a) mechanical stirring of A356 matrix; (b) mechanical stirring, (c) after thixoforming, (d) after thixoforming and T6 heat treatment of GNP-A356 composite.

Meanwhile, the observation also reveals that α-Al becomes coarser after the T6 heat treatment. However, the existence of GNP nanoparticles in the regions hindered the formation of homogeneous α-Al structures. It is because GNP has the interaction capability with the matrix to become a firm bonding surface and prevent the dispersion of atoms during solution treatment owing to its exceptionally thin two-dimensional structure [24].

3.2 Density measurement

Figure 4 compares the experimental density of base alloy and composite using the Archimedes rule. The relative density is significant for evaluating the capability of composite and is compared using the experimental density to the theoretical density of alloy and composite. The relative density discloses the escalation in density of the GNP-A356 composites after the thixoforming process and T6 heat treatment.

However, the density of as-cast composites slightly decreases compared to the density of the base alloy. The density of GNP is lower

than aluminium, which led to a tendency in the density reduction of the GNP-A356 composite [26]. Nonetheless, the thixoformed composite density increased to 2.688 ± 0.0020 gcm-3 compared to the cast composite of 2.678 ± 0.0015 gcm-3. Hence, there is a decrease of 0.37 % porosity in the composite after the thixoforming process.
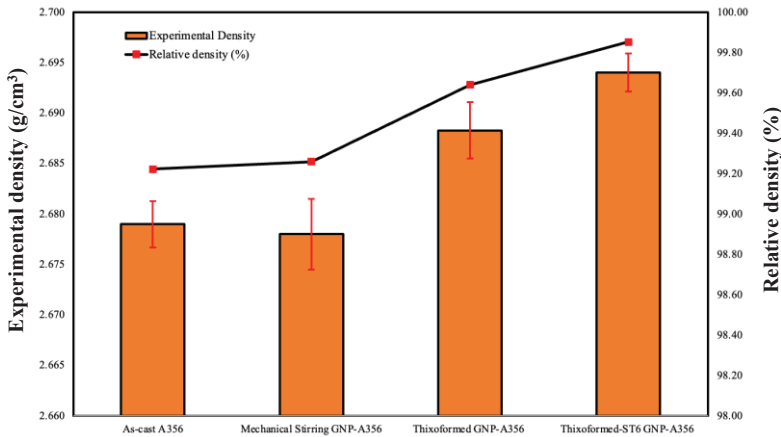

Figure 4: Experimental and theoretical density of GNP-A356

These results are in parallel with the research conducted by Alhawari et al. [23], which reveals that the compression process during thixoforming reduces the porosity amount in the thixoformed samples more than the as-cast alloy. Therefore, a solid composite can be produced through the thixoforming method. Next, the reduction of porosity was 0.48 % after being subjected to T6 heat treatment. Accordingly, the T6 heat treatment technique has a significant effect in terms of reducing the porosity level and increasing the density due to the pressure applied to the thixoformed sample.

3.2   Hardness properties

Figure 4 shows the Vickers microhardness values of alloy and composite. The GNP-A356 composite demonstrated substantially increased hardness values compared to the A356 alloy after being subjected to T6 heat treatment because of the incorporation of GNP in the alloy. The hardness of the as-cast GNP-A356 composite (85.55 ± 3.11 HV) was enhanced by 35.8% compared to the as-cast A356 alloy (63.01 ± 6.55 HV), and this enhancement revealed the significant effect of GNP on the alloy.

Afterwards, the hardness of the composites further improved to 95.39 ± 3.7 and 107.45 ± 4.6 HV after the thixoforming process and T6 heat

treatment, respectively. Numerous determinants contribute to the increment in the composite hardness. Several determinants contribute to the hardness enhancement of the GNP-A356 composite, including almost no voids in the manufactured composites, strong adhesion of the interfacial between the reinforcement and alloy, and the alteration of the required microstructure in the composite [27].
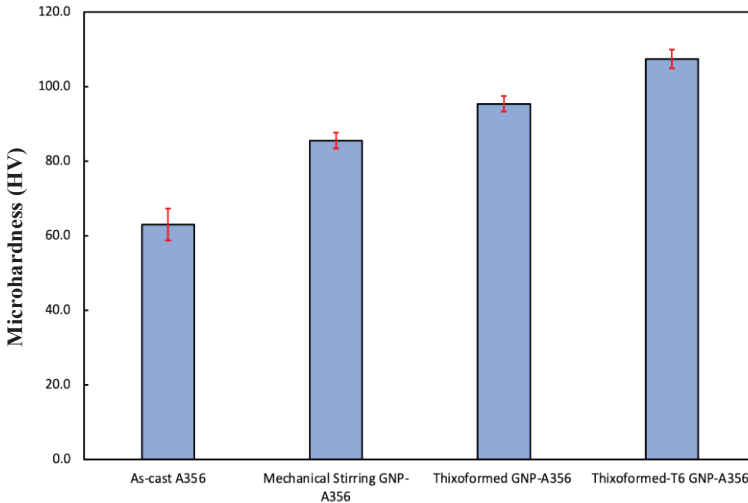

Figure 5: Hardness values of each stage of GNP-A356.

In addition, this finding may also be correlated to the refinement and spheroidal form of eutectic Si after the T6 heat treatment process. The increase in hardness after T6 heat treatment is also caused by the atoms that are dissolved in the solid solution during solutionizing. Hence, this improvement in hardness can be attributed to the influence of adding reinforcement and T6 heat treatment.

4.0 CONCLUSIONS

As a conclusion, a 0.3 wt.% GNP-A356 alloy matrix composite was fabricated using the stir casting method, followed by the thixoforming process, and the T6 heat treatment. The application of mechanical stirring resulted in the transformation of α-Al from a dendritic to a non-dendritic structure. However, incorporating GNP contributes to the refinement of α-Al compared to the base alloy. Afterwards, the large eutectic Si grains and coarse of α-Al are formed after the thixoforming process.

Subsequently, the coarsening and spheroidization of eutectic Si were noticed after T6 heat treatment. Furthermore, incorporating GNP in the

matrix and performing proper fabrication could effectively improve the overall hardness properties and reduce the porosity of the composite. The hardness was increased by 70.52% from 63.01 ± 6.55 HV of the base alloy to 107.45 ± 4.6 HV of the composite after the T6 heat treatment technique.

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AUTHOR CONTRIBUTIONS

N.F.B.W. Anuar: Conceptualization, investigation, Writing - Original Draft; M.S. Salleh: Conceptualization, Writing-review & editing, Supervision; M.Z. Omar: Conceptualization, Writing-review & editing, Supervision; W.F.H.W. Zamri: Conceptualization, writing-review & editing; S.H. Yahaya: Conceptualization, writing-review & editing; A.M. Ali: Conceptualization, writing-review & editing

CONFLICTS OF INTEREST

All authors have approved the review, agree with its submission and declare no conflict of interest in the manuscript.

REFERENCES

[1] L. Zhang, G. Hou, W. Zhai, Q. Ai, J. Feng, L. Zhang, P. Si and L. Ci, "Aluminum/graphene composites with enhanced heat-dissipation properties by in-situ reduction of graphene oxide on aluminium particles", *Journal of Alloys and Compounds*, vol. 748, pp. 854–860, 2018.

[2] A. Lakshmikanthan, S. Angandi, V. Malik, K. K. Saxena, C. Prakash, S. Dixit and K. A. Mohammed, "Mechanical and tribological properties of aluminum-based metal-matrix composites", *Materials*, vol. 15, no. 17, pp. 6111, 2022.

[3] J. W. Li, J. G. Guo, and L. J. Zhou, "Molecular dynamics studies on mechanical properties of graphene/nanotwinned aluminum matrix composites", *Physica E: Low-dimensional Systems and Nanostructures*, vol. 147, pp. 115597, 2023.

[4] J. Li and R. A. Laghari, "A review on machining and optimization of particle-reinforced metal matrix composites", *The International Journal of Advanced Manufacturing Technology*, vol. 100, pp. 2929–2943, 2019.

[5] A. Md Ali, M. Z. Omar, M. S. Salleh, H. Hashim, I. F. Mohamed, and N. F. B. Wakhi Anuar, "Mechanical behaviour and morphology of thixoformed aluminium alloy reinforced by graphene", *Materials*, vol. 15, no. 19, p. 6791, 2022.

[6] N. F. B. Wakhi Anuar, M. S. Salleh, S. H. Yahaya, M. Z. Omar, A. M. Ali, S. Sivarao and M. A. M. Ali, "Characteristics of AlSi7Mg0.3/x-GNPs composites manufactured via stir casting method", *Journal of Advanced Manufacturing Technology*, vol. 16, no. 2, 2022.

[7] Z. Zheng, X. Yang, J. Li, X. Zhang, I. Muhammad, and L. Geng, "Preparation and properties of graphene nanoplatelets reinforced aluminum composites", *Transactions of Nonferrous Metals Society of China*, vol. 31, no. 4, pp. 878–886, 2021.

[8] T. Han, F. Wang, J. Li, C. He, and N. Zhao, "Effect of GNPs on microstructures and mechanical properties of GNPs/Al-Cu composites with different heat treatment status", *Journal of Materials Science & Technology*, vol. 92, pp. 1–10, 2021.

[9] S. Rawal, T. Das, A. M. Sidpara, and J. Paul, "Fabrication and characterization of Al/GNPs composite by bottom pouring stir casting", *Materials Letters*, vol. 327, pp. 133002, 2022.

[10] A. Sharma, Y. Morisada, and H. Fujii, "Interfacial microstructure and strengthening mechanisms of SPSed Al/GNP nanocomposite subjected to multi-pass friction stir processing", *Materials Characterization*, vol. 197, pp. 112652, 2023.

[11] G. Borand and D. Uzunsoy, "Fabrication of functionally graded few-layered graphene reinforced Al-4.5Cu alloy by powder metallurgy", *Journal of Alloys and Compounds*, vol. 923, pp. 166348, 2022.

[12] S. I. Ghazanlou, S. I. Ghazanlou, S. I. Ghazanlou, S. Hosseinpour, Y. Liao, and M. Javidani, "Improving the properties of an Al matrix composite fabricated by laser powder bed fusion using graphene–TiO_2 nanohybrid", *Journal of Alloys and Compounds*, vol. 938, pp. 168596, 2023.

[13] G. Li, Y. Qu, Y. Yang, R. Chen, Q. Zhou, and R. Li, "Effect of mechanical combined with electromagnetic stirring on the dispersity of carbon fibers in the aluminum matrix", *Scientific Reports*, vol. 10, no. 1, pp. 8106,2020.

[14] B. Andilab, P. Emadi, and C. Ravindran, "Casting and characterization of A319 aluminum alloy reinforced with graphene using hybrid semi-solid stirring and ultrasonic processing", *Materials*, vol. 15, no. 20, pp. 7232, 2022.

[15] V. Chak and H. Chattopadhyay, "Fabrication and heat treatment of graphene nanoplatelets reinforced aluminium nanocomposites", *Materials Science and Engineering: A*, vol. 791, pp. 139657, 2020.

[16] S. Samat, M. Z. Omar, A. H. Baghdadi, I. F. Mohamed, and A. M. Aziz, "Mechanical properties and microstructures of a modified Al–Si–Cu alloy prepared by thixoforming process for automotive connecting rods", *Journal of Materials Research and Technology*, vol. 10, pp. 1086–1102, 2021.

[17] M. A. Gebril, M. Z. Omar, I. F. Mohamed, N. K. Othman, and O. M. Irfan, "Microstructure refinement by a combination of heat treatment and thixoforming process followed by severe plastic deformation and their effects on al-si alloy hardness", *Metals*, vol. 12, no. 11, pp. 1972, 2022.

[18] M. A. H. Abdelgenei, M. Z. Omar, M. J. Ghazali, M. A. Gebril, and M. N. Mohammed, "The effect of the rheocast process on the microstructure and mechanical properties of al-5.7si-2cu-0.3mg alloy", *Jurnal Kejuruteraan*, vol. 31, no. 2, pp. 317–326, 2019.

[19] T. Savaşkan, Z. Azakli, and A. P. Hekìmoğlu, "Effect of heat treatment on mechanical and wear properties of Zn–40Al–2Cu–2Si alloy", *Transactions of Nonferrous Metals Society of China*, vol. 31, no. 9, pp. 2651–2663, 2021.

[20] O. Trudonoshyn, P. Randelzhofer, and C. Körner, "Heat treatment of high-pressure die casting Al-Mg-Si-Mn-Zn alloys", *Journal of Alloys and Compounds*, vol. 872, pp. 159692, 2021.

[21] H. Hanizam, M. S. Salleh, M. Z. Omar, and A. B. Sulong, "Effects of mechanical stirring and short heat treatment on thixoformed of carbon nanotube aluminium alloy composite", *Journal of Alloys and Compounds*, vol. 788, pp. 83–90, 2019

[22] M. S. Salleh, M. Z. Omar, K. S. Alhawari, M. N. Mohammed, M. A. M. Ali, and E. Mohamad, "Microstructural evolution and mechanical properties of thixoformed A319 alloys containing variable amounts of magnesium", *Transactions of Nonferrous Metals Society of China*, vol. 26, no. 8, pp. 2029–2042, 2016

[23] K. S. Alhawari, M. Z. Omar, M. J. Ghazali, M. S. Salleh, and M. N. Mohammed, "Microstructural evolution during semisolid processing of Al–Si–Cu alloy with different Mg contents", *Transactions of Nonferrous Metals Society of China*, vol. 27, no. 7, pp. 1483–1497, 2017.

[24] H. Xiao, G. Ma, J. Ye, and Y. He, "Preparation of graphene reinforced AZ31B magnesium-based composites by stirring casting", *Vacuum*, vol. 191, pp. 110281, 2021.

[25] M. Beder and Y. Alemdag, "Influence of Mg addition and T6 heat treatment on microstructure, mechanical and tribological properties of Al–12Si–3Cu based alloy", *Transactions of Nonferrous Metals Society of China*, vol. 31, no. 8, pp. 2208–2219, 2021.

[26] M. Khan, R. Ud Din, A. Wadood, W. H. Syed, S. Akhtar, and R. E. Aune, "Effect of graphene nanoplatelets on the physical and mechanical properties of Al6061 in fabricated and T6 thermal conditions", *Journal of Alloys and Compounds*, vol. 790, pp. 1076–1091, 2019.

[27] H. M. Yehia, P. Nyanor, and W. M. Daoush, "Characterization of Al-5Ni-0.5Mg/x (Al2O3-GNs) nanocomposites manufactured via hot pressing technique", *Materials Characterization*, vol. 191, pp. 112139, 2022.