Effect of Graphene Content on Tensile Strength and Microstructure of Aluminum Matrix Composites

Mahmut Can Şenel, Mevlüt Gürbüz*, Erdem Koç

Mechanical Engineering Department, Faculty of Engineering, Ondokuz Mayıs University, Turkey

Copyright©2018 by authors, all rights reserved. Authors agree that this article remains permanently open access under the terms of the Creative Commons Attribution License 4.0 International License

Abstract In this study, graphene nanoplatelets (GNPs) reinforced aluminum (Al) composites with various GNPs content (0.1, 0.3, 0.5wt.%) were fabricated by powder metallurgy (PM) method. In this method, ultrasonication, mixing, filtering, drying, compacting and sintering processes were performed. The crystal structure and microstructure of powders and fabricated composites were analyzed with X-Ray diffractometer (XRD) and scanning electron microscopy (SEM). With this study, the effects of GNPs content were investigated on the density, Vickers hardness, ultimate tensile strength, and microstructure of Al-GNPs composites. The Vickers hardness (57 ± 2.5 HV) and ultimate tensile strength (120MPa) of Al-0.1%GNPs improved up to +90% and +23.3% when compared with pure aluminum (30 ± 2 HV, 92MPa). From the microstructural analysis, homogeneously distributed GNPs was located at aluminum grain boundaries. The mechanical properties of Al-GNPs composite decreased due to the agglomeration of GNPs above 0.1wt.% GNPs content.

Keywords Powder Metallurgy, Aluminum, Graphene, Composite, Hardness, Strength

1. Introduction

Aluminum-based composites are finding increased use in military, automotive, and aerospace industries due to their advanced properties [1]. In general, these composites can be fabricated through various methods such as powder metallurgy (PM), melting and squeeze casting [2]. PM is a remarkable method due to its ability of more homogeneous dispersion. The samples have been fabricated by this method owing to having its minimal finishing requirement, and economic/technical advantages [3-5].

Graphene is a two-dimensional material, a single layer of graphite consisting sp^2 -hybridized carbon atoms [6]. In addition, graphene is an attractive material owing to having its good tribological behaviors [7-10], high

electrical [11, 12], thermal [13, 14] and mechanical properties [15-23] such as high fracture strength (125GPa) [24], high Young's modulus (1 TPa) [24], extreme thermal conductivity ($5000 \text{ Wm}^{-1}\text{K}^{-1}$) [25], super charge-carrier mobility ($200,000 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$) [26]. One possible way to utilize the graphene's superior properties for application is to incorporate and disperse graphene in different material matrices such as polymers, ceramics, and metals.

Graphene has been used as reinforcement element in metal matrices such as aluminum [27-29], magnesium [30], and titanium [31] to enhance the properties of metal matrix nanocomposites. However, there are a limited number of studies related to mechanical or tribological properties of Al-GNPs composites. Bastwros et al. [32] examined the bending strength of Al-GNPs composites fabricated by PM method. The strength of Al-1%wt.GNPs increased ~47% when compared with Al6061 alloy. Rashad et al. [33] researched the effect of GNPs content on the compressive, tensile strength, Vickers hardness of aluminum composites. Comparing with pure aluminum, the tensile strength, hardness, and compressive strength of Al-0.3wt.%GNPs composite increased nearly +11.1%, +11.8%, -7.8%, respectively. Li et al. [34] investigated the tensile properties of bulk nanostructured aluminum/graphene composites. The results showed that the bulk nanostructured aluminum/graphene composite exhibited increased strength over pure aluminum with an addition of only 0.5wt.% graphene nanoflakes. Above 1.0wt.% of graphene nanoflakes, however, this strengthening effect sharply dropped due to the clustering of graphene nanoflakes.

Most of the given studies concentrated on just one GNPs content and mechanical properties. Also, the sizes of their raw materials are greater than the size of raw materials in this paper. In this study, Al-GNPs composites with various GNPs content (0.1, 0.3, 0.5wt.%) were fabricated by PM method. The effects of GNPs content were deeply investigated on the apparent density, Vickers hardness, ultimate tensile strength, and microstructure of Al-GNPs composites.

2. Materials and Method

2.1. Materials

In this study, the pure aluminum powder was used as matrix material which was purchased from Alfa Aesar (United Kingdom). Also, GNPs powders were used as reinforcement element which was supplied from Grafen Chemical Industries (Turkey). The general properties of purchased Al and GNPs powders were given in Table 1.

Table 1. Properties of aluminum and GNPs powders

Material	Average Particle Size (μm)	Theoretical Density (g/cm^3)	Elasticity Modulus (GPa)
Pure Al	8-15	2.7	68
GNPs	5-8 nm (thickness) 120-150 m^2/g (surface area)	2.25	~1000

SEM images of pure aluminum and GNPs powders were presented in Figure 1. As seen from the figure, aluminum powders contain nearly spherical, rod-like morphology, and the particle size of $\sim 10\mu\text{m}$. On the other hand, GNPs have few-layered morphology and the layer thickness of $\sim 10\text{nm}$.

Figure 1. SEM images of aluminum (a) and GNPs (b) powders

2.2. Method

The experimental procedure and sintering conditions were studied in our previous study [28]. In summaries, Al-GNPs composites were fabricated by PM method as given in Figure 2. In this method, aluminum powders were blended in an ethanol medium using a mixer. Dispersion of GNPs (0.1, 0.3, 0.5wt.%) was carried out for 1 hour using ultrasonic homogenizer. GNPs slurry added drop by drop into the aluminum-ethanol mixture. Mixing performed until homogeneous slurry obtained. After the mixture filtered and dried overnight at 50°C under vacuum. The composite powder was pressed in a mold under 600MPa to form green cylindrical samples. After compacting, the samples were sintered under vacuum in the tube furnace at the constant sintering time and temperature ($t_s = 180\text{ min}$ and $T_s = 630^\circ\text{C}$).

The experimental density of Al-GNPs composites was measured according to the Archimedes principle, by using a balance with the accuracy of 0.1 mg. A micro-Vickers hardness tester (HV1000B micro Vickers hardness tester) under a load of 200g was used to measure the hardness of the samples. The average values of at least six measurements at polished cross-sections for various areas of each sample were considered. Tensile tests were performed by the universal test machine (Mares Test-10 tons) with the tensile rate of 5 mm/min.

The morphology and microstructure of powders and sintered composites were analyzed by scanning electron microscopy (SEM, Jeol JSM-7001F). X-ray diffraction analysis (XRD, Rigaku Smartlab) were used to investigate the phase analysis of powders and composites.

Figure 2. Fabrication scheme of Al-GNPs composites

3. Results and Discussion

3.1. Characterizations of Composites

XRD patterns of pure Al, GNPs powders and Al-GNPs composites were shown in Fig. 3. As shown in the figure, GNPs peaks ($2\theta=26.5^\circ$) are not detected in any composite structure because of the low detection limit of XRD and the low content of GNPs. Also, in-situ formed secondary phase (Al_4C_3) is not detected in all Al-GNPs composite.

Figure 3. X-ray diffraction patterns of powders and Al-0.1%GNPs

Fig. 4 represents the morphology of pure Al and 0.1%, 0.3%, and 0.5%GNPs reinforced aluminum matrix composites by scanning electron microscope. Better bonding and good neck formation between the particles was observed at Al-0.1%GNPs composite. On the other hand, the agglomeration was determined at Al-0.3%GNPs and Al-0.5%GNPs composite. The agglomerated

graphene flakes led to the higher porosity, lower density, and ultimate tensile strength. The analyses carried out which corrected the density analysis and the mechanical properties of the fabricated Al-GNPs composites.

Figure 4. SEM images of fracture surface of pure Al (a), Al-0.1%GNPs (b), Al-0.3%GNPs(c), Al-0.5%GNPs (d) composite

3.2. Density and Hardness Test Results

The theoretical and experimental density variations of pure aluminum and graphene reinforced aluminum composites were given in Table 2. The maximum density value was obtained at Al-0.1%GNPs composite due to the homogeneous dispersion of GNPs in aluminum matrix. After this graphene content (0.1wt%), both theoretical and experimental density were decreased due to the agglomeration tendency of GNPs. In addition, the low theoretical density of graphene ($\rho=2.25 \text{ g/cm}^3$) affected the decrease in the density of Al-GNPs composites after 0.1%GNPs content.

Table 2. The density variation of pure Al and Al-GNPs composites

	Theoretical density (g/cm^3)	Experimental density (g/cm^3)
Pure Al	2.44 ± 0.03	2.43 ± 0.02
Al-0.1%GNPs	2.50 ± 0.02	2.52 ± 0.02
Al-0.3%GNPs	2.48 ± 0.02	2.50 ± 0.01
Al-0.5%GNPs	2.46 ± 0.02	2.49 ± 0.01

The best sintering conditions were determined as given the previous paper ($T_s=630^\circ\text{C}$, $t_s=180 \text{ min}$) [28]. By using these sintering conditions, the variation of Vickers hardness for pure aluminum and Al-GNPs composites was presented in Figure 5. The highest hardness value ($57 \pm 2.5 \text{ HV}$) was detected at Al-0.1%GNPs composite. After 0.1wt.%GNPs content, the poor interface between aluminum and graphene formed due to the large agglomerated graphene clusters which can lead to the higher porosity and the lower Vickers hardness. Also, GNPs act as solid-lubricant which promotes the easy sliding between the particles during plastic deformation.

Figure 5. The variation of Vickers hardness for pure Al and Al-GNPs composites

3.3. Tensile Test Results

The variation of ultimate tensile strength for pure Al and Al-GNPs composites was shown in Fig. 6. Similarly, the ultimate tensile strength (+23.3%) for Al-0.1%GNPs composite increased when compared with pure aluminum. In Al-GNPs composites, GNPs act as a barrier at aluminum grain boundaries in order to preserve the grain growth. The dislocations cannot move easily due to the restricted dislocation movement which increases in strength of aluminum composites. After 0.1wt.% graphene content, the ultimate tensile strength of aluminum composites reduced due to the agglomeration tendency of GNPs. This agglomeration was confirmed by the SEM images of Al-0.3%GNPs and Al-0.5%GNPs composite.

Figure 6. The variation of ultimate tensile strength for pure Al and Al-GNPs composites

4. Conclusions

In this study, pure aluminum and Al-GNPs composites with various graphene content (0.1, 0.3, 0.5wt.%) were fabricated by PM method. The effects of GNPs content on the density, mechanical properties (Vickers hardness, tensile strength), and microstructure of Al-GNPs composites were investigated. The results obtained from the study performed were summarized as follows:

- The highest apparent and experimental densities were determined as $2.50 \pm 0.02 \text{ g/cm}^3$ and $2.52 \pm 0.02 \text{ g/cm}^3$ for Al-0.1%GNPs, respectively. Similarly, the maximum Vickers hardness value was obtained as $57 \pm 2.5 \text{ HV}$ at the same composition. Density and hardness of Al-GNPs composites began to decrease above 0.1wt.GNPs content due to agglomeration tendency and easy-sliding of GNPs.
- The highest ultimate tensile strength was obtained as 120MPa in Al-0.1%GNPs. This increment is due to the superior mechanical properties of GNPs.
- By considering all mechanical test results, The Vickers hardness ($57 \pm 2.5 \text{ HV}$) and ultimate tensile strength (120MPa) of Al-0.1%GNPs improved up to +90% and +23.3% when compared with pure aluminum ($30 \pm 2 \text{ HV}$, 92MPa).
- XRD analysis showed that graphene peaks were not seen in these composites owing to the low content of GNPs and the low detection limit of XRD analyzer.
- From the SEM analyses, bonding between the particles and well neck formation were observed at pure Al and Al-GNPs composites. GNPs were detected along the aluminum grain boundaries in Al-GNPs composites.
- As a result, graphene addition to aluminum matrix enhanced the mechanical properties of Al-GNPs composites up to 0.1wt.%GNPs. After this content, the mechanical properties of Al-GNPs composites decreased due to the agglomeration of GNPs.

Acknowledgements

This work was supported by Ondokuz Mayıs University, Scientific Researched Project Department [grant numbers: PYO.MUH.1902.15.001 and PYO.MUH.1904.16.002]. The authors of this study thank Black Sea Advanced Technology Research and Application Center (KITAM) in Ondokuz Mayıs University (OMU) for SEM and XRD analysis.

REFERENCES

- [1] G. Iacob, V.G. Ghica, M. Buzatu, et al. Studies on wear rate and hardness of the Al/Al₂O₃/Gr hybrid composites produced via powder metallurgy, Compos Part B-Engineering, Vol. 69, 603-611, 2015.
- [2] A. Saboori, C. Novara, M. Pavese, et al. An investigation on the sinterability and the compaction behavior of aluminum/graphene nanoplatelets (GNPs) prepared by powder metallurgy, Journal of Materials Engineering and Performance, Vol. 26, No. 3, 993-999, 2017.
- [3] G. O'Donnel, L. Looney. Production of aluminium matrix composite component using conventional PM technology, Materials Science and Engineering A-Structural Materials, Vol. 303, 292-301, 2001.
- [4] O.G. Neikow, S.S. Naboychenko, G. Dawson. Handbook of Non-Ferrous Metal Powders—Technologies and Applications, Oxford: Elsevier Ltd, 2009.
- [5] B. Ramesh, T. Senthilvelan. Formability characteristics of aluminium based composites – a review, International Journal of Engineering & Technology, Vol. 2, 1-6, 2010.
- [6] M. Rashad, F. Pan, A. Tang, M. Asif, S. Hussain, J. Gou, J. Mao. Improved strength and ductility of magnesium with addition of aluminum and graphene nanoplatelets (Al+GNPs) using semi powder metallurgy method, Journal of Industrial and Engineering Chemistry, Vol. 23, 243-250, 2015.
- [7] D. Berman, A. Erdemir, A.V. Sumant. Graphene: a new emerging lubricant, Materials Today, Vol. 17, No. 1, 31-42, 2014.
- [8] W. Zhai, X. Shi, J. Yao, A.M.M. Ibrahim, Z. Xu, Q. Zhu, Y. Xiao, L. Chen, Q. Zhang. Investigation of mechanical and tribological behaviors of multilayer graphene reinforced Ni₃Al matrix composites, Composites: Part B, Vol. 70, 149-155, 2015.
- [9] W. Zhai, X. Shi, M. Wang, Z. Xu, J. Yao, S. Song, Y. Wang. Grain refinement: a mechanism for graphene nanoplatelets to reduce friction and wear of Ni₃Al matrix self-lubricating composites, Wear, Vol. 310, 33-40, 2014.
- [10] Z. Xu, X. Shi, W. Zhai, J. Yao, S. Song, Q. Zhang. Preparation and tribological properties of TiAl matrix composites reinforced by multilayer graphene, Carbon, Vol. 67, 168-177, 2014.
- [11] Y. Fan, L. Wang, J. Li, J. Li, S. Sun, F. Chen, L. Chen, W. Jiang. Preparation and electrical properties of graphene nanosheet/Al₂O₃ composites, Carbon, Vol. 48, No. 6, 1743-1749, 2010.
- [12] M.H. Al-Saleh. Electrical and mechanical properties of graphene/carbon nanotube hybrid nanocomposites, Syntetic Metals, vol. 209, 41-46, 2015.
- [13] C. Yun, Y. Feng, T. Qiu, J. Yang, X. Li, L. Yu. Mechanical, electrical, and thermal properties of graphene nanosheet/aluminum nitride composites, Ceramics International, Vol. 41, No. 7, 8643-8649, 2015.
- [14] Y. Song, J. Yu, L. Yu, F.E. Alam, W. Dai, C. Li, N. Jiang. Enhancing the thermal, electrical, and mechanical properties of silicone rubber by addition of graphene nanoplatelets, Materials&Design, Vol. 88, 950-957, 2015.
- [15] S.E. Shin, H.J. Choi, J.H. Shin, D.H. Bae, Strengthening Behavior of few-Layered graphene/aluminum composites, Carbon, Vol. 82, 143-151, 2014.

- [16] R. Pérez-Bustamante, D. Bolaños-Morales, J. Bonilla-Martínez, I. Estrada-Guel, R. Martínez-Sánchez. Microstructural and hardness behavior of graphene-nanoplatelets/aluminum composites synthesized by mechanical alloying", *Journal of Alloys and Compounds*, Vol. 615, No. 1, 578-582, 2014.
- [17] S.J. Yan, S.L. Dai, X.Y. Zhang, C. Yang, Q.H. Hong, J.Z. Chen, Z.M. Lin. Investigating aluminum alloy reinforced by graphene nanoflakes, *Materials Science & Engineering A*, Vol. 612, 440-444, 2014.
- [18] A.F. Boostani, S. Tahamtan, Z.Y. Jiang, D. Wei, S. Yazdani, R.A. Khosroshahi, R.T. Mousavian, J. Xu, X. Zhang, D. Gong. Enhanced tensile properties of aluminum matrix composites reinforced with graphene encapsulated SiC nanoparticles, *Composites: Part A*, Vol. 68, 155-163, 2015.
- [19] S.E. Shin, D.H. Bae. Deformation behavior of aluminum alloy matrix composites reinforced with few-layer graphene, *Composites: Part A*, Vol. 78, 42-47, 2015.
- [20] M. Rashad, F. Pan, H. Hu, M. Asif, S. Hussain, J. She. Enhanced tensile properties of magnesium composites reinforced with graphene nanoplatelets, *Materials Science & Engineering A*, Vol. 630, 36-44, 2015.
- [21] M. Rashad, F. Pan, M. Asif, A. Tang. Powder metallurgy of Mg-1%Al-1%Sn alloy reinforced with low content of graphene nanoplatelets, *Journal of Industrial and Engineering Chemistry*, Vol. 20, No. 6, 4250-4255, 2014.
- [22] M. Rashad, F. Pan, A. Tang, Y. Lu, M. Asif, S. Hussain, J. She, J. Gou, J. Mao. Effect of graphene nanoplatelets (GNPs) addition on strength and ductility of magnesium-titanium alloys, *Journal of Magnesium and Alloys*, Vol. 1, No. 3, 242-248, 2013.
- [23] L.Y. Chen, H. Konishi, A. Fehrenbacher, C. Ma, J.Q. Xu, H. Choi, H.F. Xu, F.E. Pfefferkorn, X.C. Li. Novel nanoprocessing route for bulk graphene nanoplatelets reinforced metal matrix nano composites, *Scripta Materialia*, Vol. 67, 29-32, 2012.
- [24] C. Lee, X.D. Wei, J.W. Kysar, J. Hone. Measurement of the elastic properties and intrinsic strength of monolayer graphene, *Science*, Vol. 321, No. 5887, 385-388, 2008.
- [25] A.A. Balandin, S. Ghosh, W. Bao, I. Calizo, D. Teweldebrhan, F. Miao, et al. Superior thermal conductivity of single-layer graphene, *Nano Letters*, Vol. 8, No. 3, 902-927, 2008.
- [26] K.I. Bolotin, K.J. Sikes, Z. Jiang, M. Klima, G. Fudenberg, J. Hone, et al. Ultrahigh electron mobility in suspended graphene, *Solid State Communications*, Vol. 146, No. 9-10, 351-355, 2008.
- [27] M.C. Şenel, M. Gürbüz, E. Koç. New generation composites with graphene reinforced aluminum matrix, *Engineer and Machinery*, Vol. 56, No. 669, 36-47, 2015 (in Turkish).
- [28] M. Gürbüz, M.C. Şenel, E. Koç, The effect of sintering temperature, time and graphene addition on the mechanical properties and microstructure of aluminum composites, *Journal of Composite Materials*, Vol. 52, No. 4, 553-563, 2018.
- [29] B. Oztop, M. Gürbüz. Investigation of properties of composites produced by reinforcement graphene matrix obtained from waste aluminium, *International Journal of Multidisciplinary Studies and Innovative Technologies*, Vol. 1, No. 1, 12, 2017.
- [30] L.Y. Chen, H. Konishi, A. Fehrenbacher, et al. Novel nanoprocessing route for bulk graphene nanoplatelets reinforced metal matrix nanocomposites, *Scripta Materials*, Vol. 67, No. 1, 29-32, 2012.
- [31] M. Gürbüz, T. Mutuk, Effect of process parameters on hardness and microstructure of graphene reinforced titanium composites, *Journal of Composite Materials*, Vol. 52, No. 4, 543-551, 2018.
- [32] M. Bastwros, G.Y. Kim, C.Z.K. Zhang, et al. Effect of ball milling on graphene reinforced Al6061 composite fabricated by semi-solid sintering, *Composites Part B-Engineering*, Vol. 60, 111-118, 2014.
- [33] M. Rashad, F. Pan, A. Tang et al. Effect of graphene nanoplatelets addition on mechanical properties of pure aluminium using a semi-powder method, *Progress in Natural Science-Materials International*, Vol. 24, 101-108, 2014.
- [34] J.L. Li, Y.C. Xiong, X.D. Wang, S.J. Yan, C. Yang, W.W. He, J.Z. Chen, S.Q. Wang. Microstructure and tensile properties of bulk nanostructured aluminum/graphene composites prepared via cryomilling, *Materials Science & Engineering A*, Vol. 626, 400-405, 2015.