

THE COMPARISON OF NANO- Al_2O_3 VS GRAPHENE ADDITIVES FOR THE REINFORCEMENT OF ALUMINUM MATRIX COMPOSITE

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Abstract

Composite materials have been used to replace many conventional materials because of high mechanical strength and lighter weight since several decades. In general, scientists are preparing metal matrix composites with 1 additive at different weight ratios. However, comparison of the effect of different reinforcements such as nano alumina ($n-Al_2O_3$) and graphene with metal casting is not studied before. In this paper, Al-7.0 Si-0.3Mg aluminum alloy known as A356 was used as matrix material and different fractions of $n-Al_2O_3$ (0.5, 1.0 and 1.5 wt. %) and graphene (0.075, 0.15 and 0.25 wt. %) were used as reinforcements. The novel hybrid stirring (mechanical and ultrasonic) was applied to produce samples with homogeneous distribution of additives and all the samples were subjected T6 heat treatment. Uniaxial tensile test was applied to determine the mechanical properties and quality index (QI) was calculated by using these results. The results showed that higher yield strength (YS) and ultimate tensile strength (UTS) values were obtained with 0.5 wt.% Al_2O_3 addition and with 0.25 wt.% graphene addition when compared to the reference sample. In comparison to the reference sample, UTS values increased 8.5% and 5.0% with adding $n-Al_2O_3$ and graphene, respectively. Likewise, QI values also increased as nearly 20.0% and 13.9% with adding $n-Al_2O_3$ and graphene, respectively.

Keywords: Aluminum metal matrix composites (AMMC), metal matrix composite, nano alumina, graphene

1. Introduction

Because of environmental pollution, developed countries began to publish new regulations. For instance, the European Union designated the emission standards of

CO_2 for 2015 as 130 g/km and targeted in 2021, 95 g/km for new passenger cars [1]. For this reason, engineers and scientists focused on investigating the alternative materials to decrease the weight of vehicles owing to fuel

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economy. So, composite materials have been taking a great attention in order to meet lighten in weight and strengthen, especially last decades [2-5].

Composite materials consist of two or more different material and classified according to matrix types (as metal matrix, polymer matrix, ceramic matrix) or reinforcement types (as fiber, whisker, and particulate) [6-7]. In this classification, metal matrix composites, especially aluminum based [8], are critical to weight reduction of products. Reinforcement materials are generally chosen as micron dimension to improve UTS and YS [9]. In recent years, metal matrix nanocomposites have been also started to use of investigation of the increase of ductility [9, 10].

Su et al. [9] examined the effect of n-Al₂O₃ in 2024 aluminum alloy by solid liquid mixed casting technique. Their investigating schedule consisted three steps. First step, they processed n-Al₂O₃ and pure aluminum powder into ball mill in order to obtain n-Al₂O₃/Aluminum composite powder. Second step, they added this composite powder into the melted liquid matrix and applied mechanical stirring for 10 minutes. Third step, they applied also ultrasonic stirring to degassing for 5 minutes. They obtained finer grain microstructure and improved the UTS and YS compared with the matrix as 37% and 81%, respectively. Mehdinia et al. [11] researched the influence of micro and nano aluminum on mechanical features of composite obtained by hot extrusion and mechanical alloying. They educed that the mechanical properties of composite with n-Al₂O₃ were higher than the ones with micron Al₂O₃. Mula et al. [12] tried to determine the effect of 2 wt.% nano-sized Al₂O₃ reinforcement on mechanical properties of aluminum metal matrix composite. They used ball mill to obtain n-Al₂O₃ from Al₂O₃ powder (75µm) and applied the non-contact ultrasonic casting method. After they analyzed the test samples, they elucidated that UTS increased as 57% and also hardness

increased as 92% with adding n-Al₂O₃ reinforcement. Sajjadi et al. [13] studied on different particle size of Al₂O₃ as nano and micro dimensions as well as various processing parameters. They declared that the wettability of particle was decreasing with both decreasing particle size and increasing percentage of reinforcements. Also they proved that samples with increasing amount of n-Al₂O₃ had higher hardness and compressive strength.

After 2010 year, graphene has begun to take a part in composite material area. Researchers have published many articles. Jagadish [14] investigated the effect of graphene amount in aluminum matrix composite by using powder metallurgy. He concluded that the maximum tensile test results were obtained by using composite with 0.75 wt.% graphene. If the amount of graphene increased, mechanical properties decreased. Venkatesan [15] produced samples with different amount of graphene (0.33%, 0.55% and 0.77%) in order to investigate the effect on mechanical properties of aluminum composite. They used stir casting technique at 400 rpm for 5 to 10 seconds at 820 °C. They concluded that hardness of composite samples increased with decreasing amount of graphene and obtained optimum results with 0.33 wt.% additives.

The novelty of current paper is that the comparison of the effect of n-Al₂O₃ and graphene on mechanical properties of composite samples by producing with our hybrid stirring process in order to improve the homogeneity. n-Al₂O₃ and graphene were used as reinforcement materials with 0.5, 1.0, 1.5 wt.% and 0.075, 0.15, 0.25 wt. %, respectively. A356 alloy was used as matrix material. Heat treatment process (as T6) was applied to the whole test samples then uniaxial tensile test applied. QI results were calculated with the help of the mechanical test results by using Equation 1. This equation is useful for engineers to evaluate

the suitable conditions for material selection [16-18].

$$QI = UTS + K * \log(\text{elongation}) \quad \text{Eq. 1}$$

QI; Quality index - MPa

UTS; ultimate tensile strength - MPa

K; constant (for A356 is equivalent to 150 MPa)

Elongation; (%)

Table 1. Chemical composition of A356 alloy

Element	Al	Cu	Fe	Mg	Mn	Si	Ti	Zn
wt. %	Balance	0.1	0.1	0.3	0.05	7.3	0.1	0.05

Particle size of n-Al₂O₃ was analyzed with the help of Malvern Nano ZS in Dokuz Eylul University Center for Fabrication and Application of Electronic Materials (EMUM) approximately 79 nm and it was added as 0.5, 1.0 and 1.5% wt. Graphene was supplied from Nanografi Company (from Turkey, as

2. Experimental

The chemical composition of A356 alloy which was used as matrix material in this investigation is given at Table 1.

4 kg of A356 alloy as matrix metal was put into a graphite crucible to melt at 750°C in a resistance furnace. Al₂O₃ and graphene were added at different conditions given in Figure 1.

Graphene Nanoplatelet, 99.5+%, 6 nm, S.A,150 m²/g Dia: 24µm) and it was added as 0.075, 0.15 and 0.25% wt. To improve the wettability, preheating process of n-Al₂O₃ and graphene reinforcements was applied for 20 min at 700°C and for 30 min at 600°C, respectively.

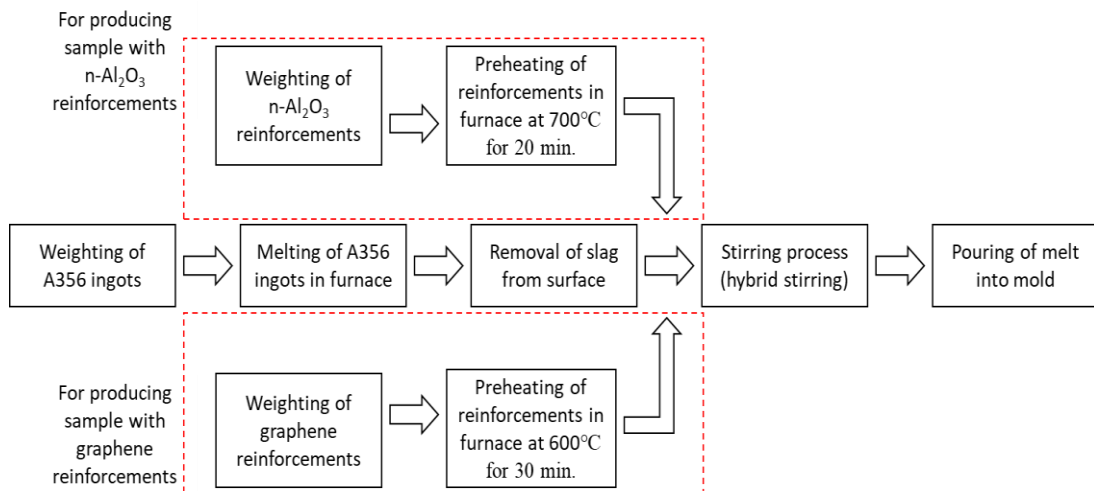


Figure 1. Flow chart of casting process; with n-Al₂O₃ and with graphene

Preheated n-Al₂O₃ and graphene were respectively added into the liquid matrix material throughout the mechanical stirring. Thereafter, ultrasonic vibration was applied for 1 minute. Stirring process was applied in order of mechanical and ultrasonic vibration with the help of Optimum B20-400 V brand machine at 600 rpm and Rtul brand machine (3 kW, 19.8 kHz), respectively. After the stirring process completed, the composite

melt was poured into the mold which was preheated to 320°C, as demonstrated in Figure 2. To compare the effect of reinforcements, reference samples were also produced without any addition. T6 heat treatment was applied to all samples as solutionized at 540°C for 4 hours, quenched in water at 80°C, artificially aged at 155°C for 3 hours.



Figure 2. Preheated mold after pouring

According to DIN EN ISO 6892-1 (in Figure 3a), tensile test samples were prepared. For mechanical analysis, Zwick Z100 brand machine was used in compliance with DIN EN 10002-1. Charpy Impact test samples given in Figure 3b were prepared in compliance with ASTM-E23 and INSTRON CEAST 9050 brand machine was used to measure. The samples were prepared for metallographic analysis with standard sample

preparation method and 0.5% HF and FeCl₃ etching solutions were applied a few seconds to sample surface to obtain micro and macro images, respectively. For microstructure examinations, Clemex S2.0C and Nikon Epiphot 200 brand machines were used. Detailed microstructure analyzes were carried out by using Carl Zeiss 300VP SEM in Izmir Katip Celebi University.



(a)



(b)

Figure 3. a) Machined tensile test bar, b) machined Charpy impact test bar

3. Results and Discussion

The microstructure photos and SDAS (secondary dendrite arm spacing) size measurement results of the samples are given in Figure 4 and Table 2, respectively. If these figures are taken into consideration, it can be said that reference sample has larger porosity and it looks like a shrinkage porosity type. But in these micrographs, the samples with different reinforcements have less porosity than reference. However Figure 4 shows that

increasing the amount of n-Al₂O₃ reinforcement makes the coarse aluminum dendrites finer. But the same situation does not occur for graphene. On the other hand, SDAS measurements (as given in Table 2) are nearly similar for each sample. This infers that there is no mold-based effect for the cooling process. Hence, it can be deduced that the internal structure of the samples are directly related to mechanical test result.

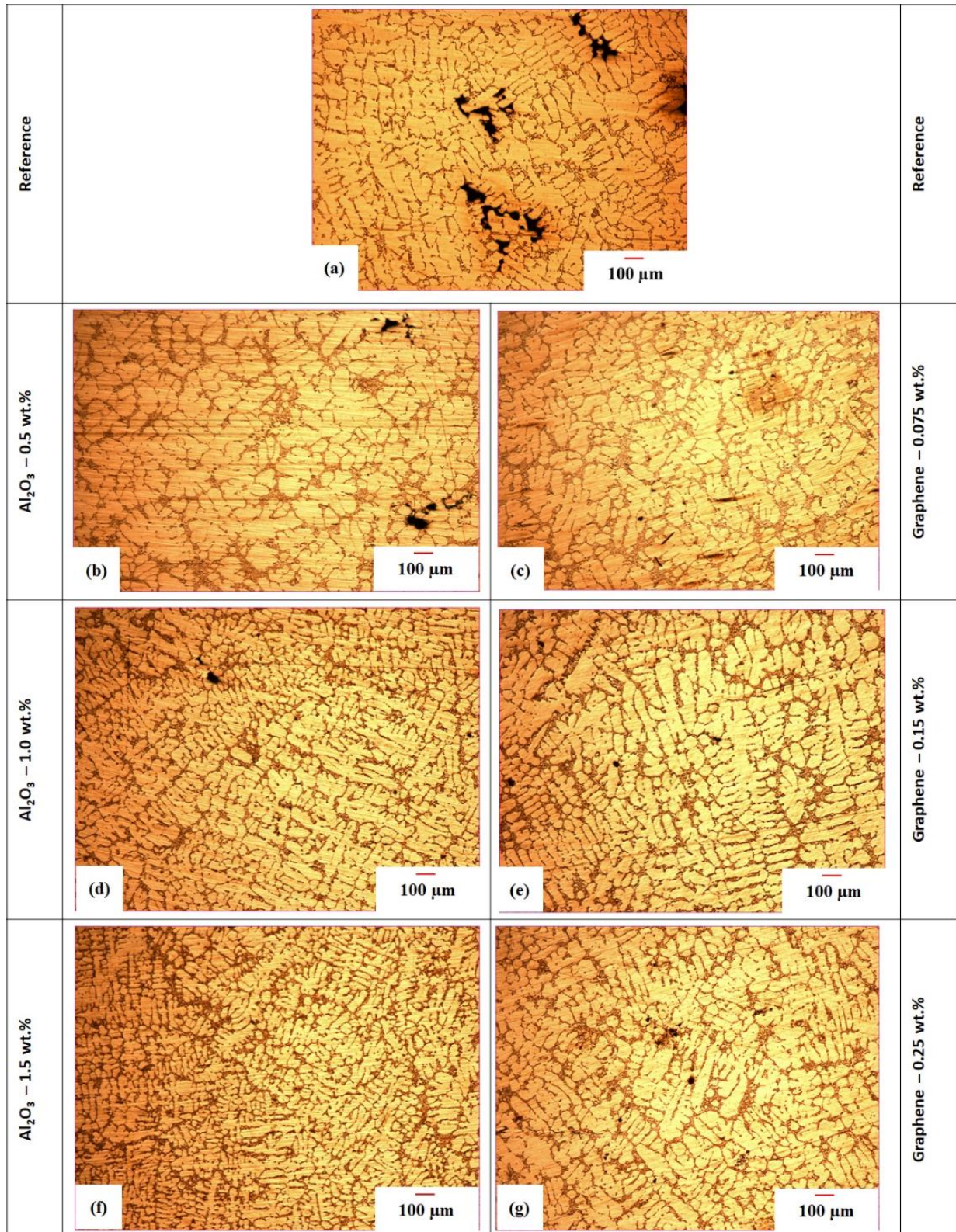


Figure 4. Microstructure images of samples

The macro structure photos and grain size measurement results of all the samples are given in Figure 5 and Table 3, respectively.

In Figure 5, it is seen that there is an increase of porosity and shrinkage in the interior structure of whole samples. But if samples

with n-Al₂O₃ are taken into consideration, density of porosity increases with the increasing amount of reinforcements. For samples with graphene, it can be said that density of porosity is decreasing with the increasing amount of reinforcements. But the porosity value should be quantified to make certain comment.

Table 2. SDAS measurement (μm)

Sample		Mean	St.dev
Reference		51.4	10.4
n-Al ₂ O ₃ (wt.%)	0.5	32.2	6.5
	1.0	33.8	6.1
	1.5	32.4	6.2
Graphene (wt.%)	0.075	38.8	7.7
	0.15	33.1	6.2
	0.25	34.6	5.6

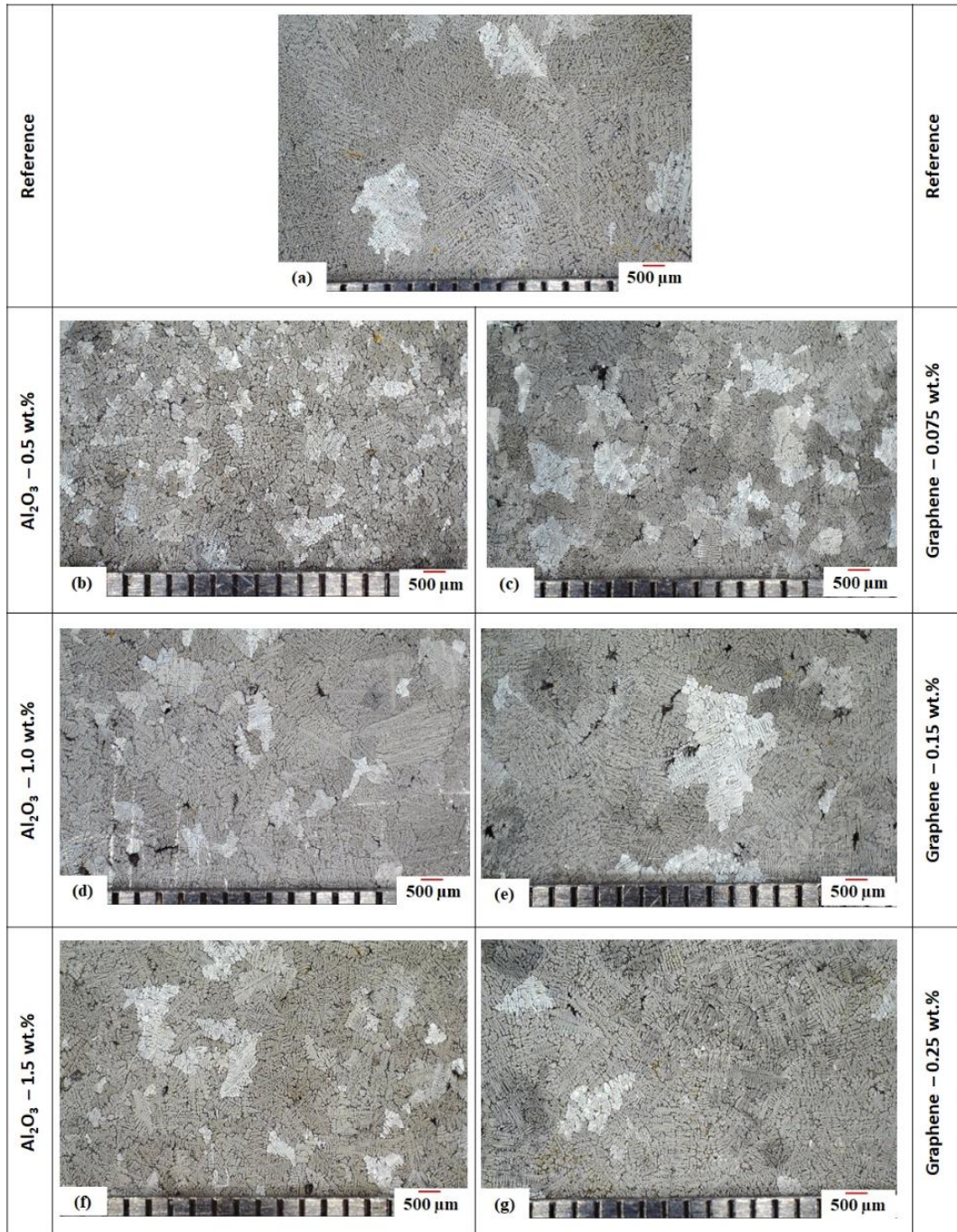


Figure 5. Macrostructures images of samples

Table 3 Grain size measurement (μm)

Sample	Mean	St.dev
Reference	1093	178
n-Al ₂ O ₃ (wt.%)	0.5	755.5
	1.0	781.7
	1.5	887.3
Graphene (wt.%)	0.075	948.4
	0.15	2226.14
	0.25	1925.9

Percentage of porosity was calculated by using differences of density between theoretical and measured as given Eq. 2 and Eq. 3 [19, 21]. The results of both theoretical and measured density were determined with

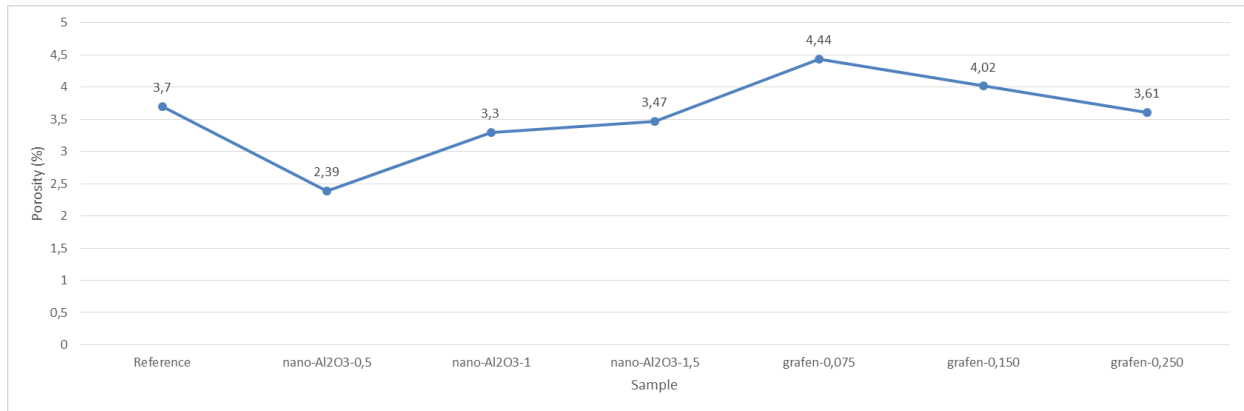
the help of rule of mixtures (seen Eq. 3) and Archimedes' principle, respectively.

$$\% \text{ porosity} = \frac{((Q_{\text{theoretical}} - Q_{\text{measured}}) / Q_{\text{theoretical}}) * 100}{\text{Eq. 2}}$$

$$Q_{\text{theoretical}} = \sum(f_i * Q_i) = f_{\text{matrix}} * Q_{\text{matrix}} + f_{\text{particle}} * Q_{\text{particle}} \quad \text{Eq. 3}$$

q; density of each one material in composite.
f; volume fraction

Percentage of porosity value were calculated by using Eq 2 and Eq 3 and shown in Fig 6.

**Figure 6.** Percentage of porosity

The calculated porosity given in Figure 6 proves the macrographs of samples given in Figure 5. When the percentage of porosity is taken into consideration, it is increasing with the increasing the amount of n-Al₂O₃. Ezatpour et al. [22] deduced that percentage of porosity increases not only with increasing the stirring duration but also with the amount of n-Al₂O₃ reinforcement. They added the same amount of n-Al₂O₃ and get the highest porosity percentage, as nearly 4%, with 1.5 wt.% addition of n-Al₂O₃. In present study, the maximum percentage of porosity was procured with the amount of 1.5 wt.% addition as 3.47%. It could be thought that this result was obtained due to the ultrasonic stirring added mechanical stirring. Babu et al. [23] elucidated that cavitation bubbles

formed by ultrasonic stirring decreases the surface tension between reinforcement and matrix material and improve the wettability. This could be the reason of getting lower porosity values than the literature.

If samples with graphene are considered, the best test results were obtained with the highest amount of additive. The porosity values depend on both the amount of additives and the type and duration of applied stirring process during sample producing. Oztop et al. [19] used waste aluminum for matrix material and different wt.% graphene (0.1, 0.3 and 0.5 wt.%) for reinforcement to investigate the effect of amount of reinforcement. He explained that if the amount of graphene reinforcement increased, the percentage of porosity could be increased.

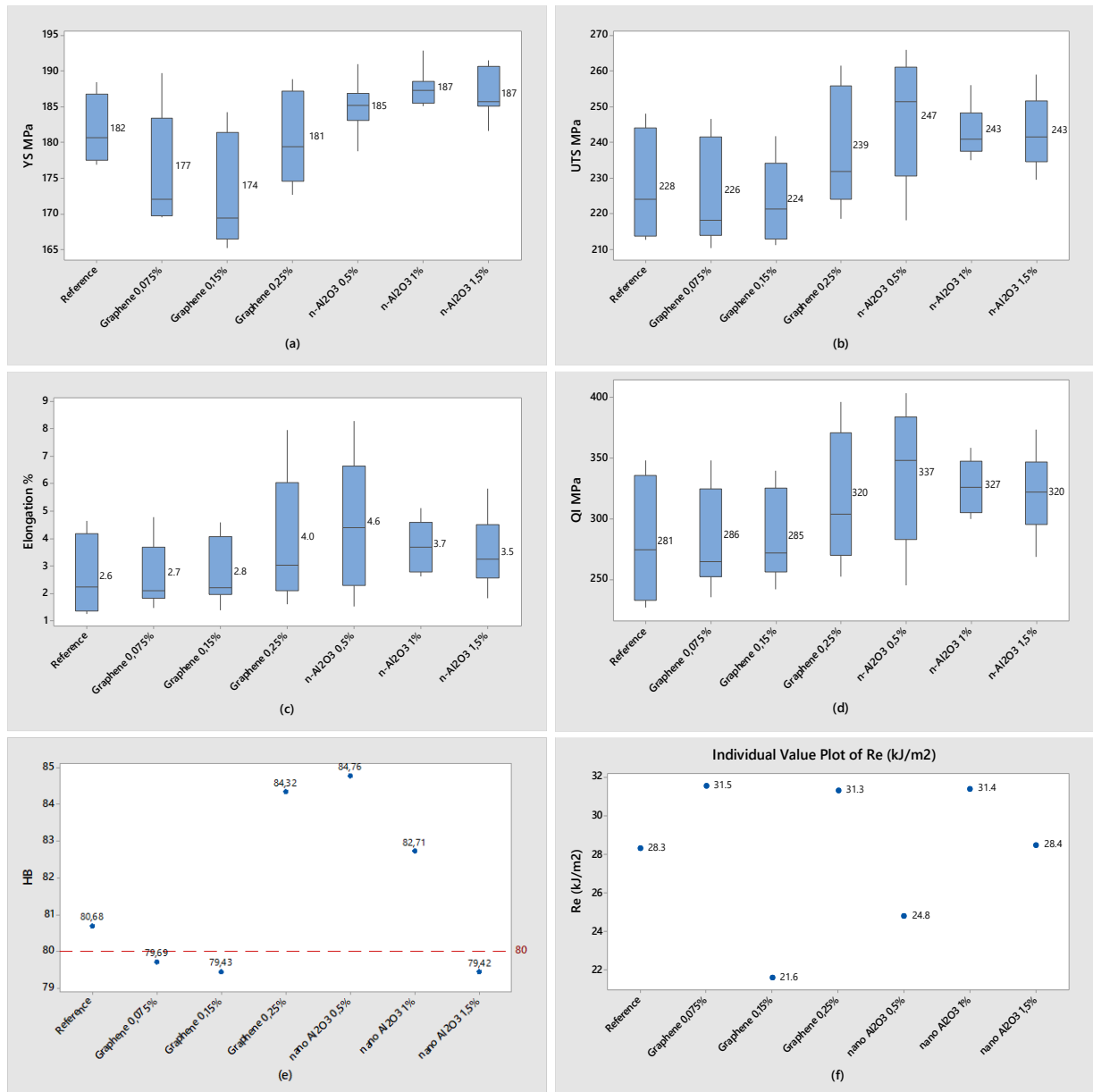


Figure 7. Mechanical test results of samples

But they only used mechanical stirrer for 6 minutes during sample producing. Hashim et al. [20] and Su et al. [9] deduced that the porosity was formed by the ingress of gases during stirring process. Therefore, the increase in the mechanical stirring cycle and the duration increases the amount of porosity because of the inlet of the air into the melt. In current study, the percentage of porosity values of samples with graphene additions is lower than the results of Oztop et al. study. There are two main reasons. One of them is

the duration of stirring as long as Oztop's and the other one is the application of ultrasonic stirring. Because, with the help of applying ultrasonic vibration, the dissolved gases in the molten metal are floated to the surface of melt. Additionally, Table 3 shows the grain size measurements. For n-Al₂O₃ samples, the grain size increases with increasing the amount of additive. But there is no regular change of particle size analysis with increasing amount of graphene and this may be due to the occurrence of agglomeration in

the added particles. Khorshid et al. [24] declared also the similar results in their paper as samples with graphene had higher grain size than samples with n-Al₂O₃. Suthar et al. [25] explained that due to the fact that having more surface area, agglomeration could be seen with small particle sizes. In current paper, the lowest grain sizes of samples with n-Al₂O₃ and graphene were obtained with amount of 0.5 wt.% and 0.075 wt %, respectively.

Fig 7 shows the mechanical test results. In this figure, it can be seen that the higher results were obtained on the samples with n-Al₂O₃ than the samples with graphene. It can be said that the particle size of reinforcement materials is the most important reason for this results. When compared each addition group in itself, the addition of 0.5 wt.% n-Al₂O₃ shows the highest UTS and elongation results. There can be two main reasons. The

agglomeration because of the increased quantity of reinforcement and higher percentage of porosity. Additionally Seretis et al. [26] explained that Al₄C₃ carbide group could be also occurred if graphene was used as reinforcement. They said that if this carbide group settled at the boundaries of interdendritic region, there could be stress accumulation. Due to this phenomena samples with graphene reinforcement got lower mechanical properties indicated a more brittle material behavior [26]. This can be seen also in Figure 8 which SEM analysis of fracture surface is given. Graphene reinforcements are located in boundaries of interdendritic boundaries of aluminum matrix. Oztop et al. [19] also declared that graphene palette looked as bright. Because of this, it can be thought that the mechanical test results of the samples with graphene are lower than the samples with n-Al₂O₃.

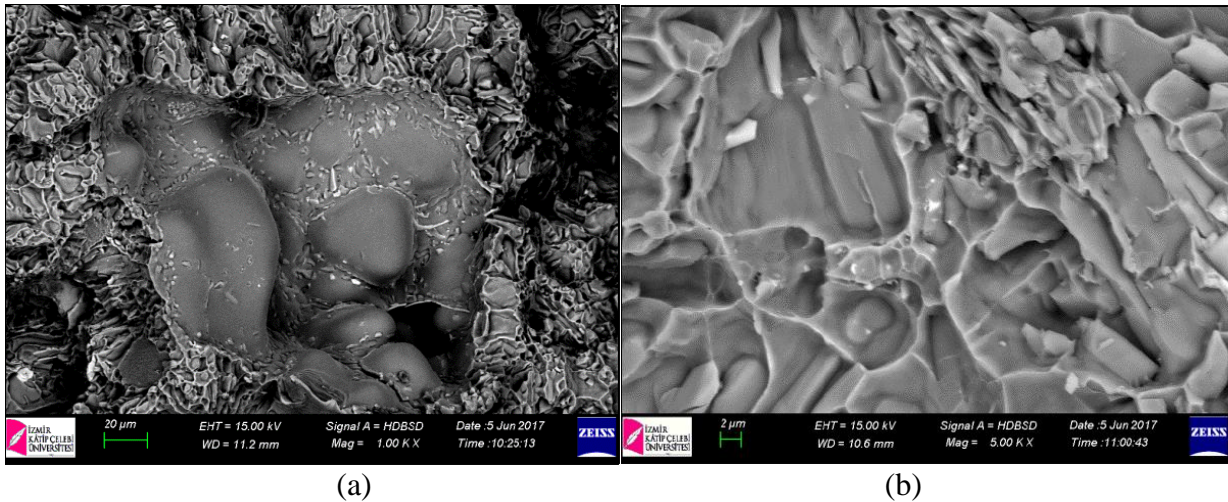


Figure 8. SEM analysis of fracture surface of samples with; a) n-Al₂O₃ and b) graphene reinforcements

Considering the QI, the results of the samples with n-Al₂O₃ additives were higher than the samples with graphene additives (Figure 7d). But increasing the amount of n-Al₂O₃ causes to decrease in the QI. On the other hand samples with graphene, the maximum QI value was obtained with the highest amount of reinforcement. When the hardness test results are examined (Figure 7e), the

maximum result was obtained with minimum amount of n-Al₂O₃ reinforcement as 87.7 BH. On the other hand, for samples with graphene, maximum hardness result was obtained with maximum amount of graphene reinforcement as 85.12 BH. On the other hand, charpy impact test result of some samples with both n-Al₂O₃ and graphene are so close to maximum value (Figure 7f).

Yuksel et al. [27] declared that this situation is correlated with the state of the porosity relative to the plane on which the load is applied.

4. Conclusions

The aim of this study is to comparison the influence of different amount of n-Al₂O₃ and graphene on aluminum metal matrix composites. At first composite samples were produced by using hybrid stirring technique then heat treatment process was applied. To determine the mechanical properties of samples, tensile, hardness and Charpy Impact tests were applied. From whole these analyses, it was concluded that;

- Aluminum metal matrix composite consist of n-Al₂O₃ reinforcements have higher mechanical properties than the one with graphene particles,
- Aluminum composite with 0.5 wt.% n-Al₂O₃ has the highest mechanical properties the reason of having the lowest porosity,
- The increasing of addition of n-Al₂O₃ decreases the result of QI, contrary to this, the result of QI increases with increasing the graphene.
- Aluminum composite with 0.5 wt.% n-Al₂O₃ has the maximum hardness value,
- Charpy impact test result of some samples with both n-Al₂O₃ and graphene are so close to maximum value. This situation can be said that is correlated with the state of the porosity relative to the plane on which the load is applied.

Note

This article is derived from Uğur Aybarç's PhD thesis.

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