Düzce University
Journal of Science & Technology

Research Article

THE EFFECT OF GRAPHENE NANOPARTICLES ON THE MECHANICAL PROPERTIES OF WOVEN ARAMID/EPOXY COMPOSITES

Ömer ŞEN [a], Musa YILDIRIM [b*]

[a] *Department of Industrial Design Eng. Karabük University Technology Faculty, KARABÜK TURKEY*
[b] *Department of Mechanical Eng. Karabük University Engineering Faculty, KARABÜK TURKEY*
** Corresponding author's e-mail address: musayildirim@karabuk.edu.tr*
DOI: 10.29130/dubited.1542336

ABSTRACT

In this study, the effect of the amount of graphene added to aramid fiber/epoxy composites on the mechanical properties of these composites was investigated. In the study, graphene nanoparticles were added to the epoxy matrix at four different rates and mixed by mechanical methods, and then 5-layer aramid epoxy graphene composite plates were obtained using hand lay-up and vacuum infusion methods. Samples were cut from these composite plates according to ASTM 790 for bending test and ASTM D3039 for tensile test and three-point bending and tensile tests were performed. Microstructure examinations were carried out under a macro microscope. After the studies, it was observed that agglomeration occurred in the microstructures of the produced composites. It was determined that graphene added to the aramid epoxy composite increased the bending strength and bending modulus, the highest bending stress was observed in the samples with 1% graphene added. The flexural strength, which compared to the undoped composite, increased about 64 % in this sample. In addition, the highest tensile strength was measured in the undoped sample, and after the 0.25% graphene addition, the tensile strength decreased due to the agglomeration of graphene that occurred in the structure.

Keywords: *Graphene nanoparticles, Mechanical properties, Aramid fibers, Epoxy composites*

Grafen Nanopartiküllerinin Dokuma Aramid/Epoksi Kompozitlerinin Mekanik Özelliklerine Etkisi

Öz

Bu çalışmada aramid fiber /epoksi kompozitlere ilave edilen grafen miktarının bu kompozitlerin mekanik özelliklerine etkisi incelenmiştir. Çalışmada epoksi matris içerisine dört farklı oranda grafen nanopartiküller ilave edilerek mekanik yöntemle karıştırıldıktan sonra el yatırma ve vakum infüzyon yöntemleri kullanılarak 5 katlı aramid epoksi grafen kompozit plakalar elde edilmiştir. Elde edilen bu kompozit plakalardan eğme testi için ASTM 790 ve çekme testi için ASTM D3039 standartlara göre numuneler kesilerek üç nokta eğme ve çekme testleri yapılmıştır. Mikro yapı incelemeleri makro mikroskop altında yapılmıştır. Yapılan çalışmalar sonrasında üretilen kompozitlerin mikro yapılarında aglomerasyonun oluştuğu gözlemlenmiştir. Aramid epoksi kompozite ilave edilen grafenin eğilme

Received: 02/09/2024, Revised: 28/10/2024, Accepted: 31/10/2024

dayanımı ve eğilme modülünü arttırdığı %1 grafen ilave edilen numunelerde en yüksek eğilme gerilmesi gözlenmiştir. Katkısız numunelere göre bu numunede eğilme mukavemeti yaklaşık %64 oranında artmıştır. Ayrıca çekme testleri sonrasında en yüksek çekme dayanımının katkısız numunelerde olduğu, %0.25 den daha fazla grafen ilave edilen numunelerde ise grafenin yapıda oluşturduğu aglomerasyona bağlı olarak çekme dayanımının düştüğü tespit edilmiştir.

Anahtar Kelimeler: *Grafen nanopartikülleri, Mekanik özellikler, Aramid lifler, Epoksi kompozitler*

I. INTRODUCTION

Polymer matrix composites are of great interest today due to the need for high strength and light materials. Fiber-reinforced polymer matrix composites are used in many areas such as the aerospace industry, the defense industry, and the automotive industry due to their low density, high mechanical properties, and high corrosion resistance [1-3]. Epoxy resin is one of the widely used matrix materials in fiber-reinforced polymer composites due to its chemical corrosion resistance and low shrinkage during curing, as well as its low weight and high adhesion properties. Glass fiber, carbon fiber, and aramid fibers are used to reinforce this matrix material [4-6]. Aramid fiber is a type of synthetic fiber that contains at least 85% amide bonds between two aromatic rings. Kevlar, Twaron, and Teijinconex are the most common commercially known aramid fibers. In general, aramid fiber is divided into two groups: meta-aramids and para-aramids. In para-aramids the bonds are aligned in the fiber direction, while in meta-aramids the bonds are formed in a zigzag pattern. Para aramids have higher strength due to different bond direction and type. Aramid fibers are used commercially in the form of yarn and woven fabrics [7-8]. Aramid fiber is a preferred reinforcement element in polymer matrix composites due to its many superior properties such as low density (1.44 g/cm3), high rigidity, high strength (3620 MPa tensile strength), and high specific module. These polymer matrix composites, which are formed by combining with various resins, are used in applications requiring high performance such as ballistic armors in airplanes and helicopters [9-10]. However, composites manufactured using strong fiber and the appropriate matrix may not always exhibit high mechanical properties. A significant parameter affecting the mechanical properties in fiber-reinforced composites is the interfacial properties between the matrix and the fiber. With the high interfacial adhesion between the matrix and the fiber, the load transfer from the matrix to the fiber is easier, thus increasing the strength of the composite [11, 12]. Since aramid fibers have chemical inertia, high crystallinity, and smooth surface properties, the interfacial bond between the matrix and the fiber is weak. To improve this bond, the properties of composites are being enhanced with carbon-based nanofillers such as carbon nanotubes and graphene. Graphene is considered one of the most effective fillers for improving bonding at the fiber/matrix interface among all carbon allotropes [13- 15].

Graphene is one of today's most popular materials, consisting of carbon atoms arranged in a hexagonal lattice to form a 2D layer, large surface area (2630 m2/g), an elastic modulus of 1.1 TPa, and high fracture toughness (125GPa) as well as high thermal and electrical conductivity. Due to these superior properties, it is used as a filler material in fiber-reinforced polymer matrix composites [9, 16, 17]. AF Ávila et al., [18] examined the bending properties of the carbon fiber/epoxy composite by adding graphene at different rates and found that graphene affected the bending strength, the highest bending strength was obtained with the addition of 0.5% graphene and emphasized that the graphene added at higher rates could not be homogeneously distributed in the structure and causes agglomeration. Namdev et al., [16] examined the mechanical properties of the composites produced by adding graphene at different rates into carbon fiber/epoxy composites and reported that the added graphene increased the tensile bending strength but after 0.5%, the graphene nanoparticles agglomerated in the structure and reduced the strength. Wu et al., (9) stated that the composites produced by adding graphene oxide at different rates into aramid/epoxy composites increased the bending strength and bending modulus and strengthened the interfacial bond between the matrix and the aramid fiber. Alsaadi et al., [19] examined the effect of graphene added in epoxy/kevlar composites and epoxy/carbon/kevlar composites on mechanical properties and emphasized that they improved tensile, bending and impact properties.

Various studies are carried out in this regard. However, many issues need to be clarified about the effects of nano-filling materials added in woven aramid fiber epoxy composites on mechanical properties. In this study, the mechanical properties of woven aramid fabric/epoxy composites produced by adding graphene nanoparticles to the epoxy resin at 4 different rates were examined.

II. MATERIALS AND METHOD

In this study, Hexion brand epoxy (MGS Lamination L160), commercially available from Dost Kimya (Istanbul), was used as the matrix material and MGS H160 as the hardener for the curing of this resin. As a reinforcement element, basket type aramid (Twaron® 1680 dtex Type 2040) fiber woven fabrics supplied commercially from the same company were used. Graphene nanoparticles with 99.9% purity, 5 nm diameter, and a surface area of 170 m2/g, commercially available from Nanokar (Istanbul) company, were used as filler material.

After the resin weight is adjusted to be twice the woven aramid fabric to be used, 0.25%, 0.5%, 1%, and 2% graphene was weighed on a precision scale and added to the epoxy resin and mixed in a mechanical mixer for 15 minutes. After the mixing process, the basket type which was cut in 135 mm X 345 mm dimensions, was laid in a plexi mold in 5 layers, and the resin fiber cloth was spread with the help of a roller by hand layup method. After the hand layup process, the vacuum bagging method was applied to obtain equal thickness in each region and to remove the air. The produced composite plates were kept at room temperature for 24 hours for curing. The production phase of composite plates is shown schematically in Figure 1.

The specimens of the obtained composite plates were cut using a water jet according to the standards for the bending (ASTM 790) and tensile tests (ASTM D3039).

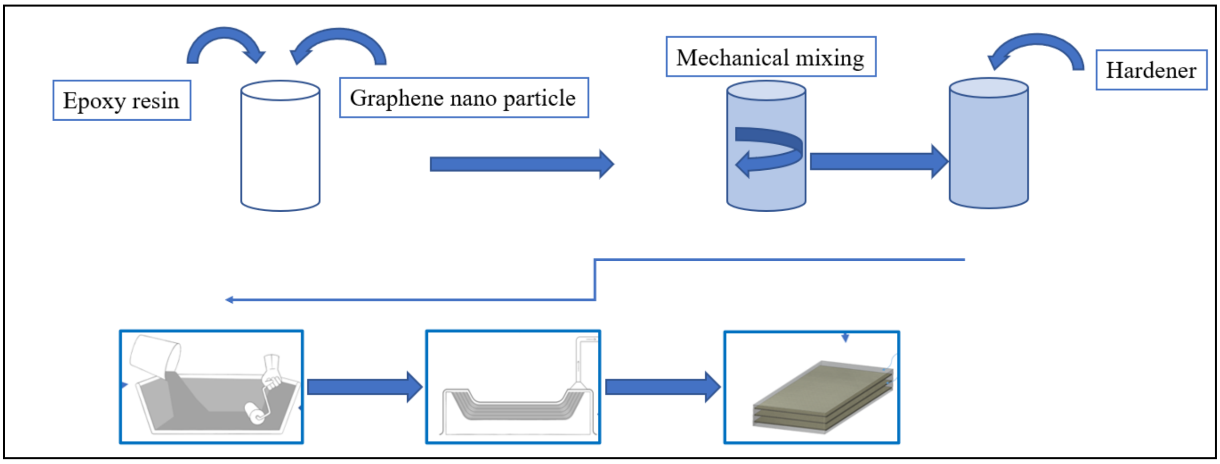


Figure 1. *The schematic view of the production of composites plates*

Stereo microscope was used to observe the microstructures of the produced composites.

A. Three-Point Flexural Test

Flexural tests of the composites produced by adding graphene at different rates and the images and dimensions of which are given in Figure 2 were carried out in Zwick/Roell Z600 brand device at room temperature at 1 mm/min jaw progression speed with 50 mm distance between supports. By using the data obtained after the experiment and the equations given below, Flexural stress (Equation 1), Flexural strain (Equation 2), and Flexural modulus (Equation 3) were calculated, respectively.

$$\sigma L = \frac{3\mathrm{P} \times \mathrm{L}}{2b \times d^2} \tag{1}$$

Here; σL = Flexural stress (MPa), P = Flexural load (N), L = distance between support points (mm), b = Specimens width (mm), d = Specimens Thickness (mm)

$$\varepsilon L = \frac{6\delta \times \mathrm{d}}{L^2} \tag{2}$$

Here; εL = Flexural strain (mm/mm), δ = Deflection of sample (mm), L = distance between support points (mm), d = Specimens Thickness (mm). The constant equivalent of δ is taken as P = 300 N in the equation.

$$EL = \frac{L^3 \times \mathrm{m}}{4\mathrm{b} \times d^3} \tag{3}$$

Here; EL = Flexural elastic modulus (MPa), L = distance between support points (mm), b = Specimens width (mm), d = Specimens thickness (mm), m = tangent angle of the deflection curve.

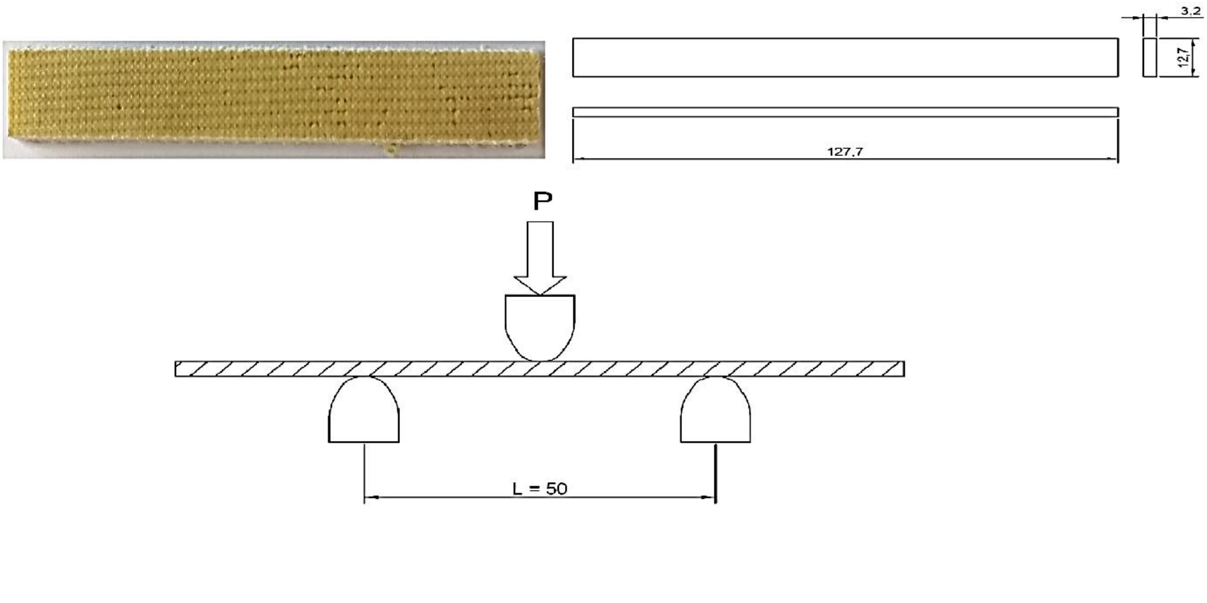


Figure 2. *The images of flexural test samples and dimensions*

A. 1. Tensile Testing

The tensile tests of the composites produced by adding graphene at different rates and the images and dimensions of which are given in Figure 3 were carried out at room temperature at a tensile speed of 2 mm/min in a 100kN capacity MTS Landmark servohydraulic universal tensile device.

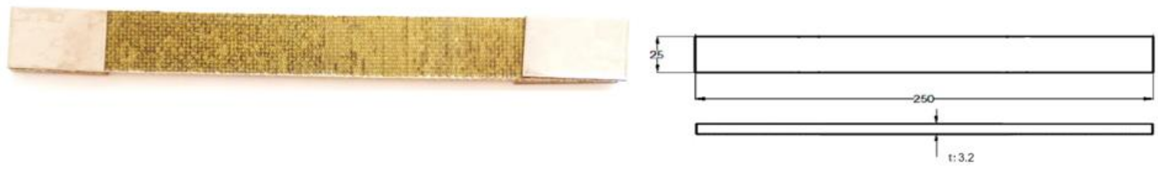


Figure 3. *Tensile test samples and dimensions*

III. RESULTS AND DISCUSSION

B. Microstructure Examination

Figure 4 shows the stereomicroscope images of the composites obtained by adding 0%, 0.25%, 0.5%, 1%, and 2% graphene nanoparticles into aramid fiber woven fabric epoxy, respectively.

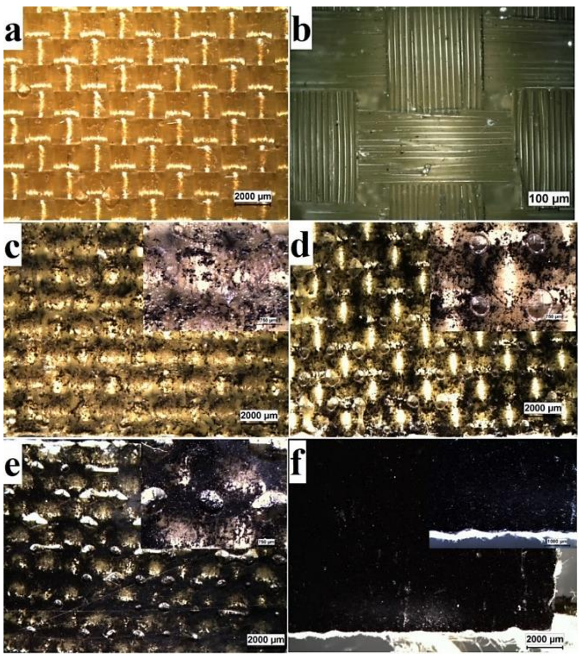


Figure 4. *Stereomicroscope images of the composites with different amount of graphene;* ***(a)*** *epoxy/aramid 0% graphene* ***(b)*** *epoxy/aramid 0% graphene (high magnification)* ***(c)*** *epoxy/aramid*

0.25% graphene ***(d)*** *epoxy/aramid 0.5% graphene* ***(e)*** *epoxy/aramid 1% graphene f) epoxy/aramid 2% graphene*

As can be seen in the microstructure images given above, the aramid fabric is basket type (a = undoped), and the matrix completely covers the aramid fabric. When the microstructure images of the composites produced by adding graphene at different rates to the epoxy aramid composite were examined, it was determined that the added graphene was collected in certain regions of the composite layer and agglomerated in these regions. In the composite to which 2% graphene was added (Figure.2f), the color of the epoxy matrix became completely black and caused it to agglomerate in all regions. The most significant problem seen in nanocomposites is that the added nanoparticles tend to agglomerate due to the high surface energy and the strong Van der Waals bond. The presence of agglomeration causes the supplemented element not to be distributed homogeneously in the structure. This adversely affects the mechanical properties of nanocomposites [19, 20].

B. 1.Three-Point Flexural Test

The load-deformation graphs of the composites obtained by adding 0%, 0.25%, 0.5%, 1%, and 2% graphene nanoparticles to Aramid fiber woven fabric epoxy are given in Figure 5.

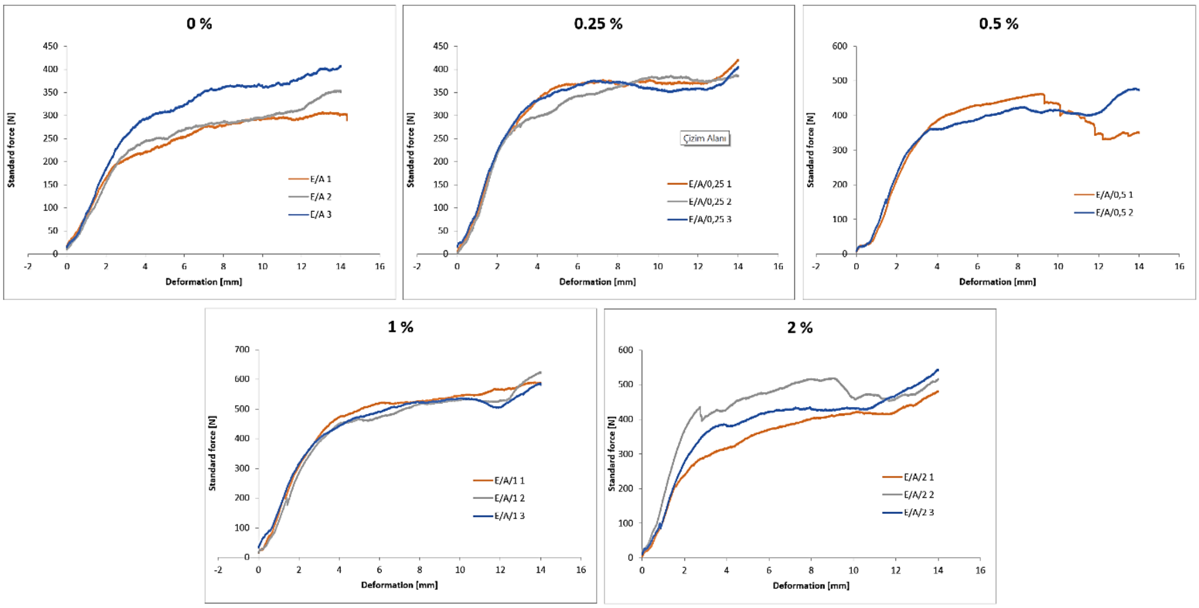


Figure 5. *The load-deformation graphs of the composites with different amount graphene*

The flexural strength, flexural elongation, and flexural modulus values calculated using these values according to equation 1, equation 2, and equation 3 are given in Table 1. The flexural strength and flexural modulus graph created by using these values are given in Figure 6.

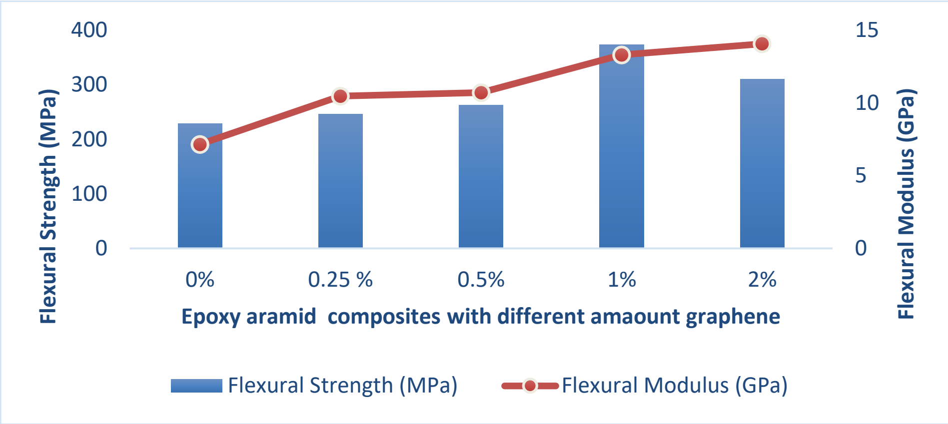


Figure 6. *The flexural strength and flexural modulus graph*

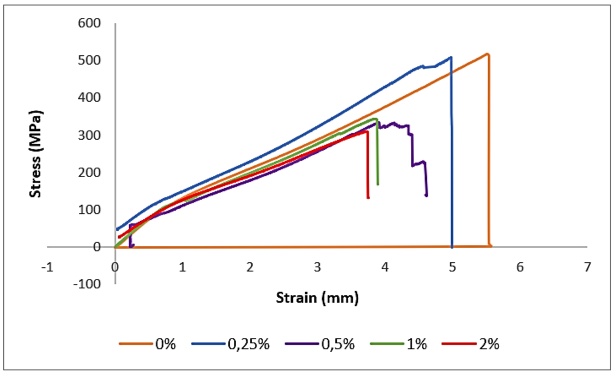
Table 1 *The Flexural properties of produced composites with different amount graphene*

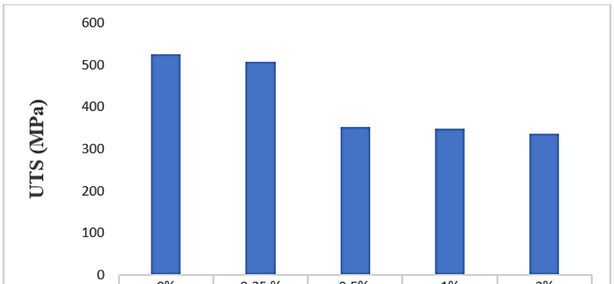
Composite	Flexural Strength (MPa)	Flexural strain(mm/mm)	Flexural Modulus (MPa)
Epoxy/ Aramid	228,47	0,015296	7163,16
Epoxy/Aramid/ 0.25% Graphene	246,04	0,003844	10468,30
Epoxy/Aramid/ 0.5% Graphene	263,12	0,003198	10726,06
Epoxy/Aramid/ 1% Graphene	373,35	0,002409	13317,77
Epoxy/Aramid/ 2% Graphene	310,60	0,002164	14068,11

As can be seen in Figure 6, while the maximum flexural strength of the undoped aramid/epoxy composite was 228 MPa, the flexural strength is 246 MPa in the composite with 0.25% graphene, 263 MPa in the composite with 0.5% graphene, 373 MPa in the composite with 1% graphene, and 310 MPa in the composite with 2% graphene. The highest flexural strength was achieved in the composite with 1% graphene. This rate increased by about 64% compared to the undoped composite. It was determined that the flexural strength increases with the graphene added. The main reason for this increase is that the added graphene increases the interfacial bond between epoxy, which is the matrix, and aramid, which is the reinforcing element. Previous studies emphasized that the nano-sized fillers increased the interfacial bond between the matrix and the fiber, thus facilitating the load transfer from the matrix to the fiber and increasing the strength [9, 19, 20]. The flexural strength of the 2% added composite decreased. As can be seen from the microstructure pictures given in Figure 4, the reason for this decrease is that the added graphene agglomerates in certain regions and reduces the mechanical properties.

B. 2. Tensile Testing

The stress-strain graph and ultimate tensile results of the composites produced by adding graphene nanoparticles at the rates of 0%, 0.25%, 0.5%, 1%, and 2% into the aramid fiber woven fabric epoxy is given in Figure 7.





	0%	0.25 %	0.5%	1%	2%
UTS (MPa)	526,03	507,5	351,7	347,65	335,4

Figure 7 *Stress-Strain and Ultimate tensile results of composites with different amount graphene*

While the highest tensile strength was obtained in the composite without graphene added, this value decreases slightly in the composite with 0.25% graphene added. However, in composites with 0.5% and above graphene added, tensile strength decreases significantly. The added graphene ratio is expected to increase tensile strength. However, since the graphene added during production is concentrated in certain regions and agglomerated in these regions, it reduces the tensile strength. The fact that the specimen is subjected to compressive stress in the load-applied region and tensile stress on the outer surface of the specimen in the bending test while it is exposed to vertical stress in the tensile test is considered the reason for this decrease in tensile strength while there is an increase in flexural strength.

IV. CONCLUSION

The results of this study, in which the mechanical properties of the composites produced by adding graphene nanoparticles at the rates of 0%, 0.25%, 0.5%, 1%, and 2%, into the aramid fiber woven fabric epoxy were examined are summarized. In microstructure examinations, it was observed that as the amount of graphene added to the aramid/epoxy composite increased, graphene was agglomerated in certain regions. It was determined that graphene added to the Aramid/Epoxy composite at different rates affects the flexural properties of the composite. As the amount of added graphene increased, the flexural strength and flexural modulus increased compared to the undoped composite. The highest flexural strength was measured in the composite with 1% graphene added. It was determined that the undoped composite had the highest tensile strength, agglomeration occurred in the structure with the increase in the amount of added graphene, and the tensile strength decreased after this agglomeration.

ACKNOWLEDGEMENTS: The authors are thanks to the financial support for this study from Karabuk University Scientific Research Projects Department (KBÜBAP-21-YL-010)

V. REFERENCES

[1] I. Taraghi, A. Fereidoon, and A. Mohyeddin, "The effect of Mwcnts on the mechanical properties of woven kevlar/epoxy composites," *Steel and Composite Structures,* vol. 17, no. 6, pp. 825-834, 2014. doi:10.12989/scs.2014.17.6.825.

[2] S.A. Mirsalehi, A.-A. Youzbashi, and A. Sazgar, "Enhancement of out-of-plane mechanical properties of carbon fiber reinforced epoxy resin composite by incorporating the multi-walled carbon nanotubes," *SN Applied Sciences,* vol. 3, no. 6, pp. 1-12, 2021. doi:10.1007/s42452-021-04624-2.

[3] T. Topkaya, Y.H. Çelik, and E. Kilickap, "Mechanical properties of fiber/graphene epoxy hybrid composites," *Journal of Mechanical Science and Technology,* vol. 34, no. 11, pp. 4589-4595, 2020. doi:10.1007/s12206-020-1016-4.

[4] T.P. Sathishkumar, S. Satheeshkumar, and J. Naveen, "Glass fiber-reinforced polymer composites–A review," *Journal of Reinforced Plastics and Composites,* vol. 33, no. 13, pp. 1258-1275, 2014. doi:10.1177/0731684414530790.

[5] T.K. Das, P. Ghosh, and N.-C. Das, "Preparation, development, outcomes, and application versatility of carbon fiber-based polymer composites: A review," *Advanced Composites and Hybrid Materials*, vol. 2, no. 2, pp. 214-233, 2019. doi:10.1007/s42114-018-0072-z.

[6] B. Zhang, L. Jia, M. Tian, N. Ning, L. Zhang, and W. Wang, "Surface and interface modification of aramid fiber and its reinforcement for polymer composites: A review," *European Polymer Journal,* vol. 147, p. 110352, 2021. doi:10.1016/j.eurpolymj.2021.110352.

[7] G. Mittal, K.Y. Rhee, V. Mišković-Stanković, and D. Hui, "Reinforcements in multi-scale polymer composites: Processing, properties, and applications," *Composites Part B: Engineering,* vol. 138, pp. 122-139, 2018. doi:10.1016/j.compositesb.2017.11.028.

[8] S. Prashanth, K.M. Subbaya, K. Nithin, and S. Sachhidananda, "Fiber Reinforced Composites–A review," *Journal of Material Science and Engineering,* vol. 6, no. 3, pp. 2-6, 2017. doi:10.4172/2169-0022.1000341.

[9] Y. Wu, B. Tang, K. Liu, X. Zeng, J. Lu, T. Zhang, and X. Shen, "Enhanced flexural properties of aramid fiber/epoxy composites by graphene oxide," *Nanotechnology Reviews,* vol. 8, no. 1, pp. 484-492, 2019. doi:10.1515/ntrev-2019-0043.

[10] S. Kumar, D.S. Gupta, I. Singh, and A. Sharma, "Behavior of kevlar/epoxy composite plates under ballistic impact," *Journal of Reinforced Plastics and Composites,* vol. 29, no. 13, pp. 2048-2064, 2010. doi:10.1177/0731684409343727.

[11] M.E. Islam, T.H. Mahdi, M.V. Hosur, and S. Jeelani, "Characterization of carbon fiber reinforced epoxy composites modified with nanoclay and carbon nanotubes," *Procedia Engineering,* vol. 105, pp. 821-828, 2015. doi:10.1016/j.proeng.2015.05.078.

[12] M.S. Tareq, S. Zainuddin, E. Woodside, and F. Syed, "Investigation of the flexural and thermomechanical properties of nanoclay/graphene reinforced carbon fiber epoxy composites," *Journal of Materials Research*, vol. 34, no. 21, pp. 3678-3687, 2019. doi:10.1557/jmr.2019.302.

[13] A.H.I. Mourad, N. Cherupurakal, F. Hafeez, I. Barsoum, F.A. Genena, M.S. Al Mansoori, and L.-A. Al Marzooqi, "Impact strengthening of laminated kevlar/epoxy composites by nanoparticle reinforcement," *Polymers*, vol. 12, no. 12, p. 2814, 2020. doi:10.3390/polym12122814.

[14] S. Sharma, A.K. Pathak, V.N. Singh, S. Teotia, S.R. Dhakate, and B.P. Singh, "Excellent mechanical properties of long multiwalled carbon nanotube bridged kevlar fabric," *Carbon*, vol. 137, pp. 104-117, 2018. doi:10.1016/j.carbon.2018.05.017.

[15] V.G.S. Veerakumar, B.P. Shanmugavel, and S. Harish, "On the influence of the functionalization of graphene nanoplatelets and glass fiber on the mechanical properties of GFRP composites," *Applied Composite Materials,* vol. 28, no. 4, pp. 1127-1152, 2021. doi:10.1007/s10443-021-09908-9.

[16] A. Namdev, A. Telang, and R. Purohit, "Effect of graphene nano platelets on mechanical and physical properties of carbon fibre/epoxy hybrid composites," *Advanced Materials and Processing Technologies*, vol. 8, pp. 1168-1181, 2022. doi:10.1080/2374068X.2021.1939557.

[17] A. Kumar, K. Sharma, and A.R. Dixit, "Effects of various functional groups in graphene on the tensile and flexural properties of epoxy nanocomposites: A comparative study," *Fullerenes, Nanotubes and Carbon Nanostructures,* vol. 30, no. 11, pp. 1123-1133, 2022. doi:10.1080/1536383X.2022.2077332.

[18] A.F. Ávila, L.D.O. Peixoto, A. S. Neto, J.D. Ávila Junior, and M.G.R. Carvalho, "Bending investigation on carbon fiber/epoxy composites nano-modified by graphene," *Journal of the Brazilian Society of Mechanical Sciences and Engineering,* vol. 34, pp. 269-275, 2012.

[19] M. Alsaadi, B. Younus, A. Erklig, M. Bulut, O. Bozkurt, and B. Sulaiman, "Effect of Graphene Nano-Platelets on Mechanical and Impact Characteristics of Carbon/Kevlar Reinforced Epoxy Hybrid Nanocomposites," *Proceedings of the Institution of Mechanical Engineers, Part C: Journal of Mechanical Engineering Science,* vol. 235, no. 23, pp. 7139-7151, 2012. doi:10.1177/09544062211016883.

[20] S. Fu, Z. Sun, P. Huang, Y. Li, and N. Hu, "Some basic aspects of polymer nanocomposites: A critical review," *Nano Materials Science,* vol. 1, no. 1, pp. 2-30, 2019. doi:10.1016/j.nanoms.02.006.