Bilecik Şeyh Edebali Üniversitesi Fen Bilimleri Dergisi, Cilt: 4, Sayı: 2, 2017
ISSN: 2458-7575 (http://edergi.bilecik.edu.tr/index.php/fbd)

Araştırma Makalesi/Research Article

Investigation of The Mechanical and Thermal Properties of Graphene Oxide Filled Polypropylene Composites

S. Hakan YETKİN[1*], Bilhan KARADENİZ[2], Mustafa GÜLEŞEN[3]

Abstract- **In this study, the effect of graphene oxide (GO) on the mechanical and thermal properties of polypropylene (PP) composites were examined. Twin-screw extruder was used for preparing the GO filled PP polymer granules. Injection molding method was utilized to produce graphene oxide/PP nanocomposites. According to tensile tests, the tensile strength and modulus of elasticity of PP matrix were increased by 42% and 71% with GO reinforcement, respectively. Conversely, the elongation at break and impact strength were decreased by 341% and 56%, respectively. Also, the prepared GO/PP nanocomposite represented higher thermal stability as heat deformation temperature (HDT) and the vicat softening temperature (VSP) were simultaneously increased. The fracture surfaces obtained after tensile test were analyzed by scanning electron microscopy.**

Keywords- **Graphene oxide, Mechanical properties, Thermal Properties**

I. INTRODUCTION

Polypropylene (PP) is one of the most widely used semi-crystalline thermoplastics due to low weight, relatively low cost, excellent heat distortion temperature, and its recyclability [1-2]. However, polypropylenes have certain disadvantages such as low impact toughness and poor adhesion of hydrophilic reactive groups in PP chains [3]. The regarding properties of the pure polypropylenes can be improved by using different additives. In recent years, researchers have focused on nanosized materials reinforced polymers as an alternative to traditional fillers of polymers [4-10]. Polymeric nanocomposites reinforced with carbon-based nano-filler such as carbon nanotube, carbon nanofibers, and fullerenes to graphene exhibit high thermal and electrical conductivities [11, 12]. Among these filler, graphene derived from graphite which consist of a single layer of carbon atoms densely packed in a two-dimensional honeycomb lattice with high mechanical properties (1060GPa in Young's modulus and an ultimate strength of 130GPa) [13]. In addition to these unique mechanical properties, high specific surface area of graphene sheets result in strong interfacial interactions between polymeric matrix and this provides better load transfer between polymeric matrix and filler particles [14, 15].

In this study, the effect of graphene oxide content on the mechanical and thermal properties of polypropylene (PP) nanocomposites were examined. For this purpose, 0.05-0.5 wt.% nanographene oxide was added into PP polymer.

II. EXPERIMENTAL STUDIES

A. Materials

Graphene oxide (TEGO-1) was produced in Nanografen Co in Turkey. PP3374E3 commercial grade polypropylene was used as the matrix material. The density of the polypropylene is 0.91 g/cm^3.

B. Preparation of TEGO-1 filled PP nanocomposites

Figure 1 represents schematically the manufacturing of TEGO-1 filled PP nanocomposites. first, TEGO-1 was dispersed in water by probe sonicator (30min and 35% amplitude) to exfoliate graphene layers. This solution mixed with PP polymer and then was dried at 100 ^{o}C for 2 hours before extrusion process.TEGO-1 filled PP composites were prepared by NRII-75-WernerPfleiderer model twin-screw extruder at 185-210^{o}C. The specimens for mechanical tests were produced using the injection-molding machine. The injection heater temperatures are set as 190-220^{o}C. The mold temperature was fixed at 30°C. By means of this procedure, the prepared samples with measured densities and Shore A values are given in Table 1.

[1*] Sorumlu yazar iletişim: hakan.yetgin@dpu.edu.tr

[2,3] İletişim: bilhan.karadeniz@dpu.edu.tr , mustafa.gulesen@dpu.edu.tr

[1,3] İmalat Mühendisliği Bölümü, Simav Teknoloji Fakültesi, Dumlupınar Üniversitesi, SİMAV-KÜTAHYA*
[2] Yapı İşleri ve Teknik Daire Başkanlığı, Dumlupınar Üniversitesi, SİMAV-KÜTAHYA

C. Characterization

Tensile tests were carried out on a Zwick Roell Z-100 machine in accordance with ASTM D638 standard, at room temperature and at speed of 10 mm/min. Notched isod impact tests were carried out in accordance with TS 1005 standard at room temperature. The impact tests were carried out using 4x10x80mm specimens with Zwick / Roell HIT 5-5P impact tester. The heat distortion temperature (HDT) and Vicat softening point of the samples were evaluated according to ISO 75-2 and ISO 306 respectively by Ceast HDT 3 machine. In HDT test, a constant pressure (0.45 MPa or 1.8 MPa) is applied to the middle of the test sample. The temperature at which the center of the test sample is tilted to 0.254mm (0.01in) is the bending temperature at a pressure of 1.8MPa. In Vicat test, temperature was increased with the rate of 120°C/h until standard indenter with area of 1mm^2 penetrate 1mm into the specimens by appliying 50N. Surface morphologies of graphene oxide flakes were analyzed by FEI-NanoSEM 650 model scanning electron microscopy (SEM).

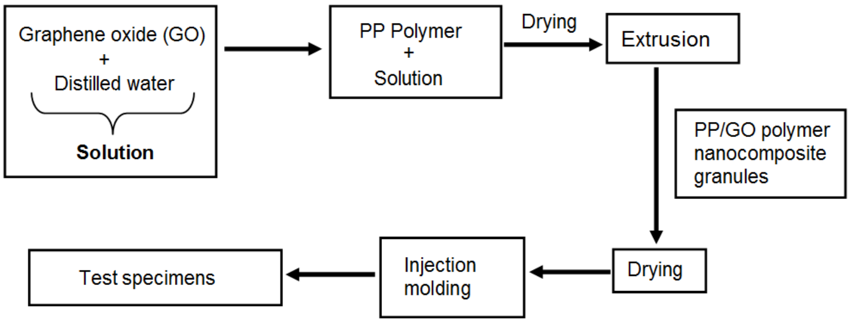

Figure 1. The manufacturing process of PP nanocomposites

Table 1. Produced samples and abbreviations, and result of density and Shore A

No	Formulation	Abbreviation	Density, g/cm^3	Shore D
1	Polypropylene	PP	0,8775	72,25
2	Polypropylene+0.05wt.% Graphene oxide	PP-0.05GO	0,8793	72,29
3	Polypropylene+0.15wt.% Graphene oxide	PP-0.15GO	0,8801	74,21
4	Polypropylene+0.3wt.% Graphene oxide	PP-0.3GO	0,8808	73,83
5	Polypropylene+0.5wt.% Graphene oxide	PP-0.5GO	0,8806	73,63

III. EXPERIMENTAL RESULT

Figure 2 represents SEM images of TEGO-1 flakes before and after mixing process. TEGO-1 has worm-like structure since thermal expansion leads to the decomposition of oxygen functional groups from the surface of graphene layers and layers are separated each other and accordion-like expansion is observed [16]. TEGO-1 has approximately 4% surface oxygen groups in its structure that is obtained from EDX characterization [11].

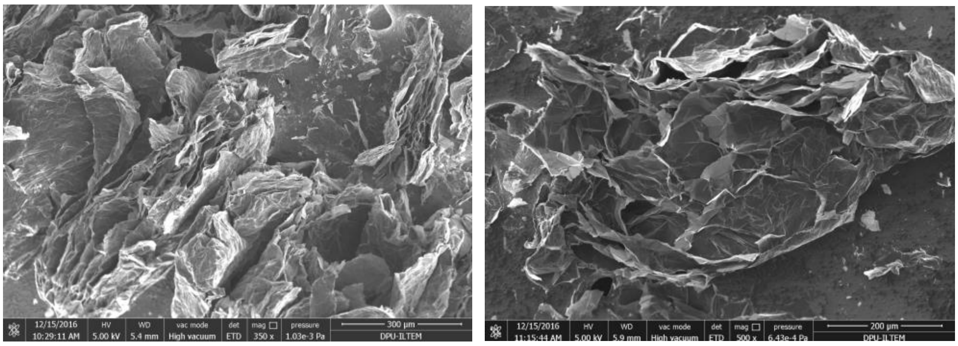

Figure 2. SEM images of graphene oxide before and after solution

Figure 3 and 4 displays the variation of HDT and Vicat temperature of the PP and GO/PP nanocomposites, respectively. HDT and Vicat softening point are indicative of the load carrying capacities of polymeric materials under increasing temperature and pressure. It is clear from these figures that the HDT and Vicat temperature increases with the graphene oxide loading. In the case of 0.5GO filler addition, the increase is about 4.3% for HDT and 9.2% for Vicat temperature. HDT values of PP polymer are 46.2^{o}C, and the increase of HDT is only 2^{o}C PP-0.5GO samples. Vicat is measured about 71.3^{o}C for PP polymer while it is measured as 75.7^{o}C, 76.2^{o}C, 76.8^{o}C and 77.9^{o}C for PP-0.005GO, PP-0.15GO, PP-0.3GO and PP-0.5GO, respectively. This increase in the thermal properties of polymeric composites is a limitation of the polymer chain motility due to the interactions between graphene oxide sheets and polymer chains. Similar results were obtained by [17].

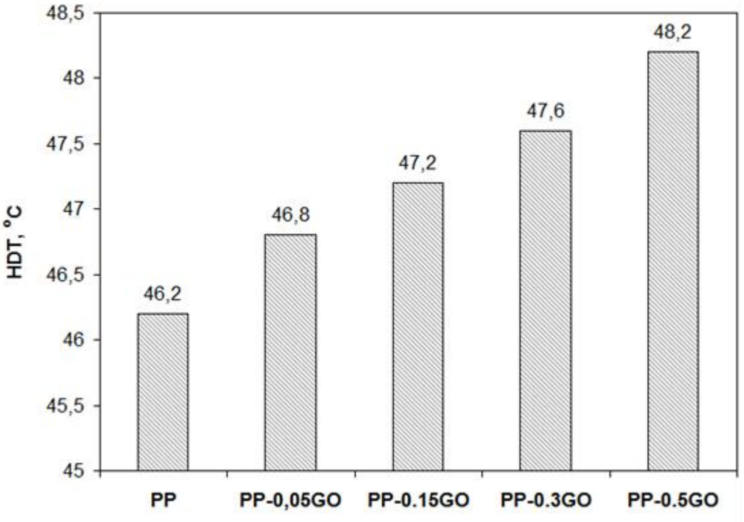

Figure 3. Variation of the HDT values of graphene oxide filled PP

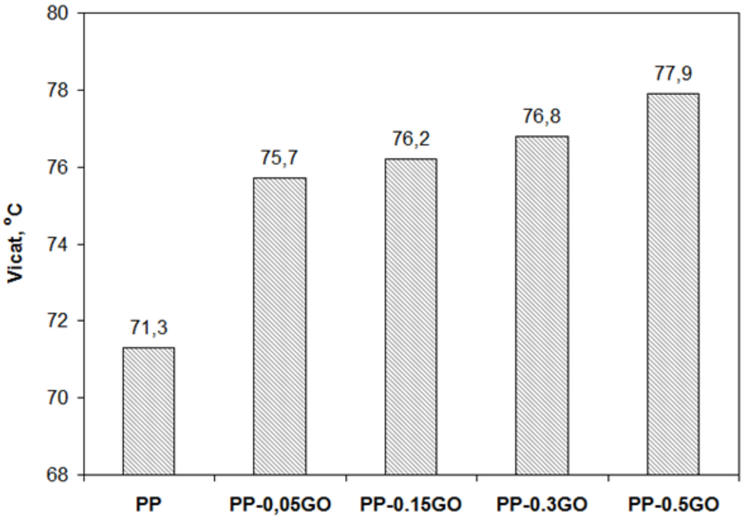

Figure 4. Variation of the Vicat values of graphene oxide filled PP

The mechanical properties of graphene oxide filled polymeric nanocomposites depend on the dispersion of GO sheets in the polymeric matrix and strong interfacial interaction between the GO and the polymer matrix [18]. Figure 5 display the variation of the tensile strength of the PP and PP nanocomposites. It is clear from this figure, tensile strength increases with the increase in the GO filler percentage. For 0.3wt% GO filler addition, the influence is quite high and the increase is about 42%. Above this amount, there is a decrease in the tensile strength. Liang et. al. [19] investigated the GO filled polyvinylalcohol (PVA). They obtained 76% increase in the tensile strength and 62% in the modulus of elasticity at 0.7wt%of GO. Figure 6 shows the variation of modulus of elasticity of PP and GO/PP nanocomposites. The modulus of elasticity increases linearly with the increase of GO loading. The modulus of elasticity of the PP polymer of 1028 MPa reached up to 1767 MPa with an increase by 71% for PP-0.5GO. When inorganic particles are loaded into polymer materials, the movement of molecular chains will be limited due to a number of physical crosslinking points between the filler and the polymeric matrix, leading to improve the stiffness of the composites systems. For a crystalline polymer, the inclusions will play a role of heterogeneous nucleation, leading to increase the crystalline degree or to change the crystal type structure, and to improve the stiffness of the composite this case. When the filler particles uniformly disperse in the polymeric matrix, the stiffness of polymer composites will increase with increasing the number of the inclusions [10].

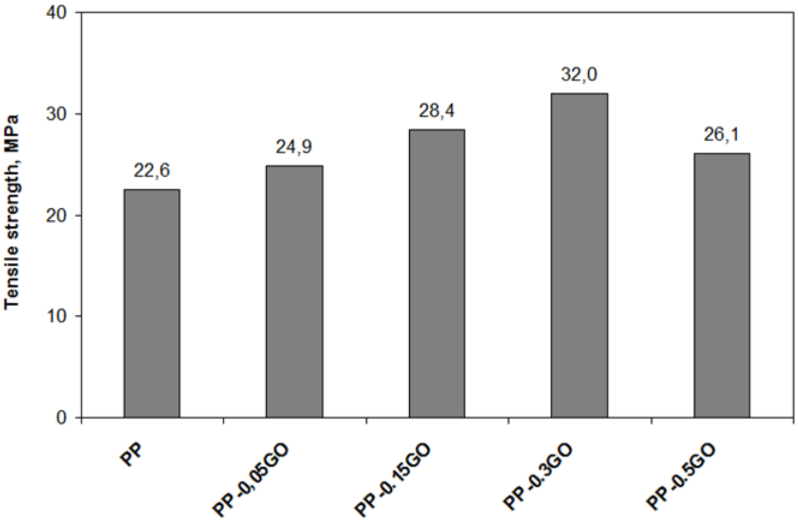

Figure 5. Variation of the tensile strength of PP and PP nanocomposites

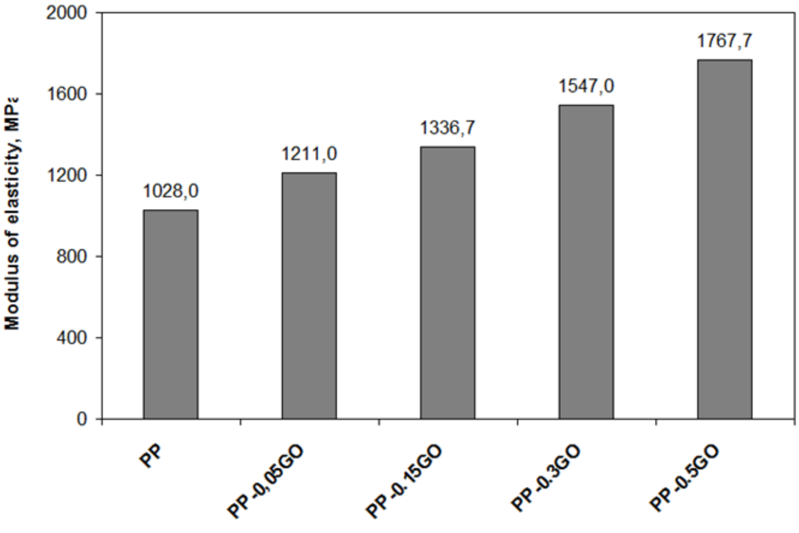

Figure 6. Variation of the modulus of elasticity of PP and PP nanocomposites

The elongation at break is an important parameter for characterization the ductility of materials [10]. Figure 7 illustrate the variation of the elongation at break value for PP and PP nanocomposites. The result show that the elongation at break decreased with increase GO ratio. In the case of 0.05GO, 0.15GO, 0.3GO and 0.5GO filler addition, the decrease is about 90%, 123%, 207% and 341%, respectively. Figure 8 present the variation of impact strength with graphene oxide filler. It is clear from this figure, the impact strength decreases with the addition of GO filler. The graphene oxide added to the PP polymer decreased the izod impact strength by 56%. This is explained by immobilizations of the macromolecular chains by the GO which increase the brittleness of the PP polymer.

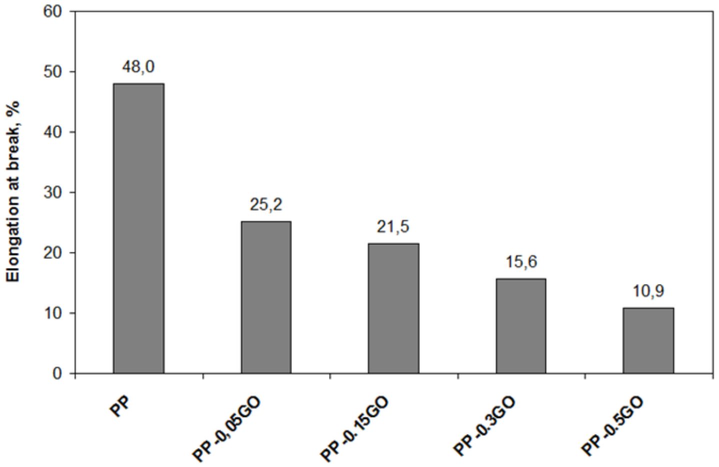

Figure 7. Variation of the elongation at break of PP and PP nanocomposites

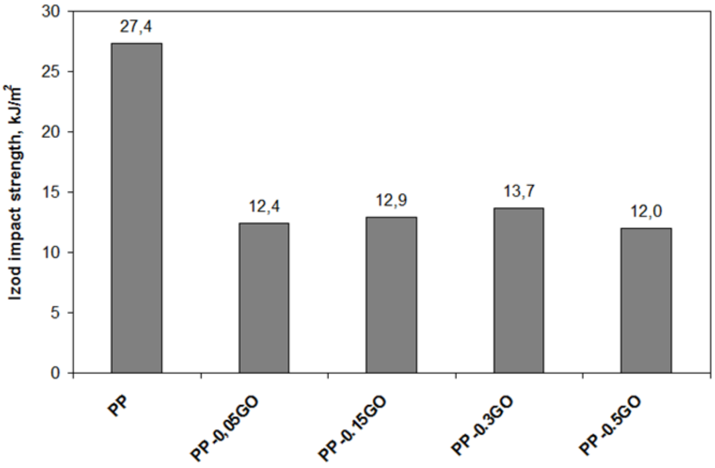

Figure 8. Variation of the izod impact strength of PP and PP nanocomposites

Figure 9 present the variation of bending strength of PP and PP nanocomposites. The result shows that the bending strength increase up to 0.3wt.% GO loading and there is a decrease in the bending strength for higher GO loadings. The bending strength improved from 32MPa for PP polymer to 44MPa at 0.3GO filler concentration. Additional of GO resulted in decrease as observed at 37MPa for 0.5GO. Similar result obtained by Christopher et. al. [17].

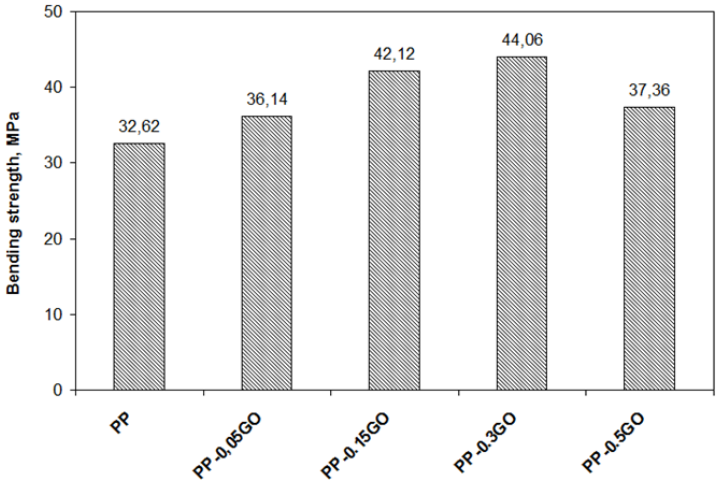

Figure 9. Variation of the bending strength of PP and PP nanocomposites

IV. RESULTS

Graphene oxide addition of PP polymer increased HDT and Vicat values. The graphene oxide addition of the PP polymer increased the tensile strength by 42%. The modulus of elasticity of the PP polymer of 1028 MPa reached 1767 MPa with an increase of 71%. The graphene oxide added to the PP polymer reduced the elongation at break by about 341% compared to the PP polymer. The graphene oxide added to the PP polymer decreased the izod impact strength by 56%. The bending strength of the PP polymer of 32 MPa reached 44 MPa when 0.5% graphene oxide is added.

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