

# Investigation of the Synergistic Effect of Addition the Hybrid Carbon Fiber, Graphene Nanoplatelet and Matrix Modifier to Poly(phenylene sulphide) on Physical Properties

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**Abstract:** This study aims to investigate the effect of graphene nanoplatelet (GNP) and terpolymer addition on the mechanical, adhesive wear and thermal properties of carbon fiber (CF) reinforced poly(phenylene sulphide) (PPS) matrix composites. In the composites the CF content was fixed at 10 wt.% and the terpolymer content at 2 wt.%, while the GNP content was changed to 0.1, 0.3 and 0.5 wt.%. Physical properties of composites were evaluated by using tensile test, adhesive wear test, differential scanning calorimeter analysis and scanning electron microscopy analysis. As a result, it was concluded that the simultaneously addition of both 0.5 wt.% GNP and 2 wt.% terpolymer to CF reinforced composites improved the mechanical and tribological properties of composites. Thus, a solution has been developed to increase the efficiency of use of high performance composite materials produced for use in areas requiring good mechanical and tribological performance such as the aerospace and automotive industries.

**Keywords:** Composites, Carbon fiber, Mechanical properties, Nanoparticles, Thermal properties

## Introduction

Carbon fiber (CF) reinforced poly(phenylene sulphide) (PPS) matrix composites are commonly used for the production of materials which are utilized in the automotive industry and defense/military applications. These materials are also used in the production of electrical and electronic components, sporting vehicles, seats, wings, and tail and body parts of space and air vehicles because of their important properties like low density, high specific strength, modulus and rigidity. Depending on the diversity of their usage areas, these materials are subjected to many tribological and mechanical environmental effects. It is an important requirement that these materials be operated in environments requiring tribological and mechanical performance by improving their properties. Because, that will provide ecological benefit as well as economic benefit by extending the service life of these materials.

In order to achieve these aims, the tribological and mechanical properties of CF reinforced PPS matrix composites should be optimized also by taking into account the properties of the interface between CF and PPS. Because, the ultimate mechanical and tribological properties of composites are widely related to the nature of the interfacial adhesion between fiber and matrix [1-5]. However, the interfacial interaction between PPS and CF is weak. Because PPS exhibits low adhesion and wettability due to the low amount of hydrophilic reactive groups in its structure. In addition, CF exhibits low interaction with polymers due to

its non-polar surface properties. Thus, the matrix modification method can be used to improve the interaction between CF and PPS and promote interfacial adhesion. The matrix modification method aims to make the matrix more reactive by using a physical or chemical method and to generate a chemical bridge between the fiber-matrix interface [6]. In this study, ethylene-acrylic ester-glycidyl methacrylate (GMA) terpolymer was used as matrix modifier due to the reactive tendency between GMA and PPS [7,8].

One of the commonly used methods to promote fiber-matrix interface interaction in composites is the matrix modification method, but the use of carbon-based nanomaterials is also a prominent method for this purpose. In particular, carbon nanotube (CNT) and graphene (GN) are used for this purpose due to their outstanding properties [9-14]. Although GN exhibits higher performance than CNT, it is a new generation material that can be produced at lower production cost than CNT. However, GN's poor surface functionality limits its performance. For this purpose, graphene oxide (GO) structure containing reactive groups on its surface is used. However, there are two important factors that limit the use of GO in the production of high performance composites. The first is the weight of reactive groups on the GO surface. Because it causes the GN sheets to bend and worse the mechanical properties of GN. The other is GO's high production cost for high volume production processes. Therefore, the use of relatively low-cost graphene nanoplatelets (GNPs) has emerged as an alternative way to reinforce polymers and improve fiber-matrix interfacial interaction. GNPs are structured as small stacks of multiple GN sheets containing graphite nanocrystals in a plate-like structure.

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The planar nature of the GNPs and the ultrahigh aspect ratio give them a large surface area in which they can contact the polymer [14-16]. In addition, the weak Van der Waals bonds between the GNP layers facilitate sliding between layers and reduce the coefficient of friction. For this reason, GNPs are promising materials for tribological applications [14,17,18]. However, GNPs also have some disadvantages. Their major disadvantage is that they agglomerate in the composite production process. Because, the surface areas of agglomerated GNPs reduce and they show properties similar to graphite. For this reason, they cannot exhibit their performance sufficiently [15,16]. The hybrid use of GNP with fiber is an option to overcome this disadvantage. Because, the hybrid usage of GNP with fiber increases the polarity of the composite system, improves the homogeneous dispersion of GNP and prevents agglomeration of GNP in the polymeric matrix [13]. In such a case, it is expected that well-dispersed GNP can easily locate at the fiber-matrix interface and promotes interfacial adhesion between the fiber and the matrix.

In the literature, there are many studies that investigate the properties of composites, which are produced by adding GNP to pure polymers and fiber reinforced polymers [14,15,19-31]. Besides, in the literature there are some studies investigate the properties of GNP included PPS matrix composites [32-38]. However, the literature review revealed that there are no studies in the literature examining the effect of GNP addition on physical properties of CF reinforced PPS in the presence of some type of matrix modifier. In order to contribute to this deficiency in the literature, the effect of GNP and terpolymer addition on the mechanical, adhesive wear and thermal properties of CF reinforced PPS matrix composites was investigated in this study.

## Experimental

### Materials

Poly(phenylene sulphide) (Fortron<sup>®</sup>0205P4) with a density of 1.35 g/cm<sup>3</sup> was purchased from Celanese. Commercially available AC1101 CF (DowAksa) with the length of 6 mm was employed in this study. GNP was purchased from Nanografi (Turkey) and used as received. According to the manufacturer's data sheet, average diameter, thickness, purity and surface area values belong to GNP are 1.5 µm, 3 nm, 99.9 % and 800 m<sup>2</sup>/g, respectively. Ethylene-acrylic ester-glycidyl methacrylate based terpolymer (Lotader<sup>®</sup> AX8900) was supplied from Arkema (France).

### Composite Preparation

In this study, composite preparation procedure was performed in two stages. In the first stage; PPS, 10 wt.% CF, GNP (the content was altered as 0.1 wt.%, 0.3 wt.%, 0.5 wt.%) and 2 wt.% Lotader<sup>®</sup> were mixed in laboratory

**Table 1.** Compounding ratios and code names of composites

PPS/CF/GNP/Lotader <sup>®</sup>	PPS (wt.%)	CF (wt.%)	GNP (wt.%)	Lotader <sup>®</sup> (wt.%)
PPS	100	0	0	0
PPS-10CF	90	10	0	0
PPS_0.1GNP	99.9	0	0.1	0
PPS_0.3GNP	99.7	0	0.3	0
PPS_0.5GNP	99.5	0	0.5	0
PPS_10CF_0.1GNP	89.9	10	0.1	0
PPS_10CF_0.3GNP	89.7	10	0.3	0
PPS_10CF_0.5GNP	89.5	10	0.5	0
PPS_10CF_0.1GNP_2L	87.9	10	0.1	2
PPS_10CF_0.3GNP_2L	87.7	10	0.3	2
PPS_10CF_0.5GNP_2L	87.5	10	0.5	2
PPS_10CF_2L	88	10	0	2

scale micro compounder for 3 min. The temperature and the screw speed were maintained at 310 °C and 100 rpm, respectively. In the second stage; prepared compounds subsequently injection molded by using a laboratory scale injection molding machine in order to production of the test samples. Injection pressure, melt temperature and mold temperature were maintained at 10 bars, 310 °C and 25 °C, respectively. The compositions and code names of composites were summarized in Table 1.

## Composite Characterization

### Tensile Test

Tensile strength at yield values of composites were determined by using a tensile testing machine (Instron-4411), at room temperature. The crosshead speed was set to 5 mm/min. and tensile strength values in this study were obtained from the average of the results for tensile tests of at least five samples.

### Adhesive Wear Test

Coefficient of friction (COF) values of specimens were determined by using a pin on-disc tester (Nanovea Tribometer), at room temperature. In the adhesive wear test, trial analyzes have been conducted to pre-determination to test conditions. For this purpose, test sample was positioned on a rotating disc and friction radius was set to 4 mm due to the shape of the analyzed sample and the sample was abraded by using 3 mm radius ceramic ball. However, contact load, sliding distance and speed of rotating disc have been changed in order to obtain easy-to-observe wear trace and at the end of the tries these parameters have been set to 30 N, 120 m and 150 rpm, respectively.

Right after the adhesive wear test, wear traces of samples have been examined by using a light microscope and wear volume of the samples has been calculated by assuming that there has been no significant pin wear and using equation

below [39]:

$$V = \frac{\pi \cdot R \cdot D^3}{6 \cdot r} \quad (1)$$

where  $V$  is the wear volume ( $\text{mm}^3$ );  $R$  is the friction radius (mm);  $D$  is the wear trace width (mm); and  $r$  is the ball radius (mm). Then the wear rate  $k$  was calculated according to the equation below [39]:

$$k = \frac{V}{L \cdot X} \quad (2)$$

where  $V$  is the wear volume ( $\text{mm}^3$ ),  $L$  is the load (N),  $X$  is the sliding distance (m) and  $k$  is the wear rate ( $\text{mm}^3/\text{N}\cdot\text{m}$ ).

#### Differential Scanning Calorimetry Analysis

Glass transition temperature ( $T_g$ ), cold crystallization temperature ( $T_c$ ), melting temperature ( $T_m$ ), cold crystallization enthalpy ( $\Delta H_c$ ) and heat of fusion ( $\Delta H_m$ ) values of composites were determined by using a differential scanning calorimetry (DSC) analyzer (TA Instruments-Q200). The analyze was performed in a temperature range from 25 °C to 300 °C at a heating rate of 5 °C/min. The relative degree of crystallinity ( $X_{c,rel}$ ) values of the composites were calculated by using following expression:

$$(X_c)_{rel} = \frac{\Delta H_m - \Delta H_c}{(1 - \omega_{fp}) \cdot \Delta H_m^0} \quad (3)$$

In this equation,  $\Delta H_m$  and  $\Delta H_c$  express the heat of fusion and cold crystallization enthalpy of each sample, respectively. Additionally,  $\omega_{fp}$  expresses the weight ratio of reinforcement materials and  $\Delta H_m^0$  expresses the heat of fusion of the neat PPS (used as  $\Delta H_m - \Delta H_c$  value for neat PPS in this study).

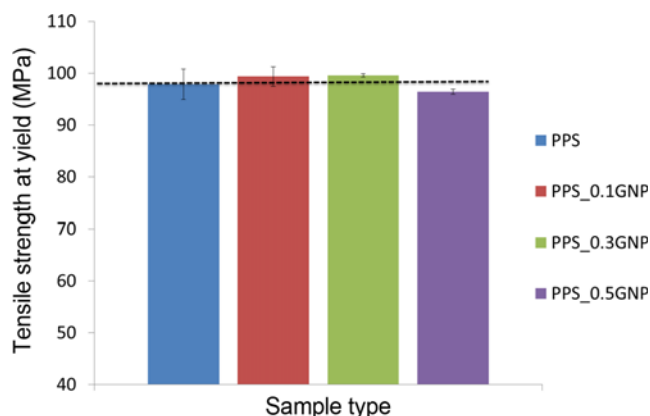
#### Scanning Electron Microscopy Analysis

Morphological investigations of the wear trace, wear debris and tensile fracture surfaces of the composites were performed by using a bench top scanning electron microscope (JEOL-JCM-6000). The worn surfaces of the composites were coated with gold by sputter coating method before analyses.

## Results and Discussion

### Tensile Test

In this study, the reasons for using GNP were both to reinforce the PPS matrix and to improve the interfacial interaction between CF and PPS matrix. In addition, the GMA-based terpolymer was used as a matrix modifier to promote the interfacial interaction between the reinforcement materials (GNP and CF) and the PPS matrix by increasing the polarity of the composite system. With this perspective, the tensile test results were given in three steps. In the first step, the effects of only GNP addition to the PPS matrix on tensile strength at yield were evaluated, in order to make a comparison with later steps. In the second step, the effects of both GNP and CF simultaneously addition to PPS matrix



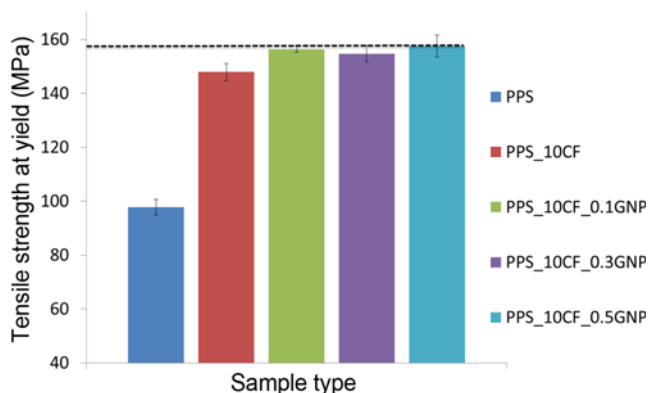
**Figure 1.** Tensile strength at yield values of only GNP included composites.

**Table 2.** Tensile strength at yield values of samples

PPS/CF/GNP/Lotader <sup>®</sup>	Tensile strength at yield (MPa)
PPS	97.81 (±2.93)
PPS_0.1GNP	99.35 (±1.87)
PPS_0.3GNP	99.59 (±0.32)
PPS_0.5GNP	96.45 (±0.50)
PPS-10CF	147.87 (±3.22)
PPS_10CF_0.1GNP	156.40 (±1.19)
PPS_10CF_0.3GNP	154.67 (±3.01)
PPS_10CF_0.5GNP	157.56 (±3.17)
PPS_10CF_2L	147.73 (±0.47)
PPS_10CF_0.1GNP_2L	154.55 (±1.31)
PPS_10CF_0.3GNP_2L	158.00 (±3.2)
PPS_10CF_0.5GNP_2L	160.98 (±2.07)

composites on tensile strength at yield were evaluated to find out whether GNP alters the interfacial interaction between CF and PPS matrix. In the last stage, the effects of the simultaneous addition of GNP, CF and Lotader<sup>®</sup> to the PPS matrix composites on tensile strength at yield were evaluated in order to reveal whether Lotader<sup>®</sup> further alters the interfacial interaction between the reinforcing materials and the PPS matrix. According to the above explanation, tensile strength at yield values of composites containing only GNP were given in Figure 1 and Table 2.

As can be seen from Figure 1 and Table 2, the addition of 0.1 and 0.3 wt.% GNP to neat PPS slightly increased the tensile strength at yield value, while the addition of 0.5 wt.% GNP decreased this value. As mentioned before, agglomeration is one of the most obvious drawbacks of GNP. The main reasons for this situation, which prevents GNP from interfacial adhesion with the polymeric matrix, are both the restacking tendency between GNP layers and the chemical inertness of GNP. In addition, the type of polymer used has a

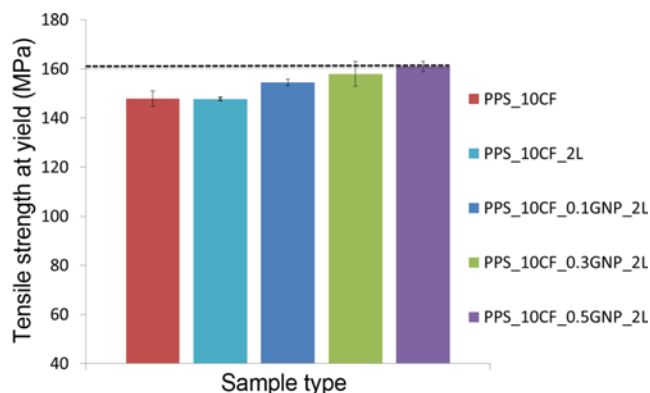


**Figure 2.** Tensile strength at yield values of GNP and CF included hybrid composites.

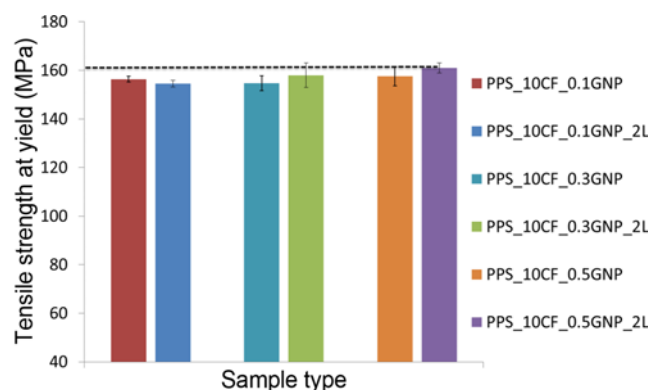
significant effect on the GNP's tendency to agglomeration [40]. Based on this information, it can be said that GNP cannot exhibit its performance when adding 0.1 and 0.3 wt.% to the PPS matrix due to the increased agglomeration and reduced surface area. Moreover, with the addition of 0.5 wt.% GNP to the PPS matrix, the agglomeration of GNP further increases and accordingly tensile strength at yield value decreases. As a result, it can be concluded that adding only GNP to the neat PPS matrix does not significantly contribute to the tensile strength at yield value of PPS (for a content of 0.1, 0.3 and 0.5 wt.% GNP).

The tensile strength at yield values of both GNP and CF reinforced PPS matrix hybrid composites were given in Figure 2 and Table 2. The graph and table show that the addition of only CF to neat PPS increased tensile strength at yield value, as expected. In addition, the graph and table show that the simultaneously addition of both GNP and CF to neat PPS matrix increased the tensile strength at yield value. Such that, the highest increase rate of 6.5 % was obtained in composites containing 0.5 wt.% GNP.

There are basic requirements that must be met in order to obtain an efficient reinforcement performance from nano-sized particles such as GNP. These requirements are good dispersion, high aspect ratio, efficient interfacial stress transfer, and good alignment. Dispersion is the most important of these requirements and is directly related to the others [41,42]. If a good dispersion is obtained, the problem of agglomeration is eliminated and the high aspect ratio and surface area of GNP are used more efficiently. Thus, the interfacial interaction between GNP-polymer and GNP-CF improves. Additionally, well-dispersed GNPs can easily locate at the fiber-matrix interface and improves fiber-matrix interaction, as well. According to these mechanisms, if Figure 2 and Table 2 are re-evaluated, it can be said that adding only GNP does not affect the tensile strength at yield value of neat PPS; however, it can also be said that the using both GNP and CF simultaneously improves the dispersion of GNP in the PPS matrix. This is because the well-dispersed



**Figure 3.** Tensile strength at yield values of GNP, CF and Lotader<sup>®</sup> included hybrid composites.



**Figure 4.** Effect of Lotader<sup>®</sup> addition on the tensile strength at yield values of GNP and CF included hybrid composites.

GNPs position at the interface between CF and PPS matrix and they improve fiber-matrix interaction. Thus, the improved fiber-matrix interaction enabled a more efficient interfacial stress transfer from PPS to CF; and as a result, the tensile strength at yield value of composites improved.

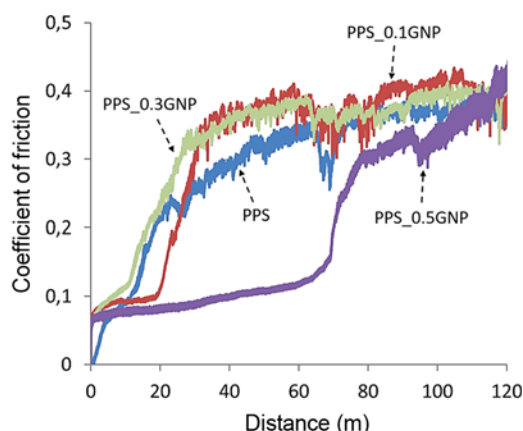
Tensile strength at yield values of hybrid composites, simultaneously containing GNP, CF and Lotader<sup>®</sup>, were given in Figure 3, Figure 4 and Table 2. The values show that only Lotader<sup>®</sup> addition to only CF reinforced PPS matrix composites did not change the tensile strength value. This result is indicative of the reaction tendency between PPS, CF and Lotader<sup>®</sup>. Because, the tensile strength value of neat Lotader<sup>®</sup> is 4 MPa and this value is much lower than the tensile strength value of only CF reinforced PPS matrix composites. According to rule of mixture, if there is no chemical interaction between Lotader<sup>®</sup> and composite, the addition of Lotader<sup>®</sup> to the composite reduces the tensile strength at yield value of composite. Therefore, in this study, the absence of a decrease in the tensile strength value of the composite with the addition of Lotader<sup>®</sup>, is an indicator of mentioned interfacial interactions for our study (for this weight ratio of Lotader<sup>®</sup>).

Additionally, it can be observed from Figure 3, Figure 4 and Table 2 that the tensile strength values of both GNP and CF containing hybrid composites increased with the insertion of 2 wt.% Lotader<sup>®</sup> (for 0.3 and 0.5 weight ratios of GNP). Such that, the highest rate of increase was obtained in composites containing 0.5 wt.% GNP as 8.8 %. It can be concluded that this result may originate from the reaction between the following structures: (i) glycidyl methacrylate groups on the Lotader<sup>®</sup> structure; (ii) hydroxyl, carboxyl, amine groups in the phenoxy based sizing material on the CF surface; (iii) oxygen included functional groups on the GNP surface [7,43]. By means of the reactions between these reactive groups in the structure of the composite constituents, the interfacial interaction between reinforcement materials and PPS has increased. Thus, a more efficient stress transfer from the PPS matrix to the reinforcement materials has been provided. In addition, the homogeneous dispersion of GNPs in the composite further improved and some of the well-dispersed GNPs positioned on CF surface and formed an interphase between CF and PPS. This case has led to obtain a better interfacial interaction between CF and PPS matrix. Accordingly, the improved interfacial interaction resulted in more efficient stress transfer from PPS to CF; and it improved tensile strength at yield [14].

### Adhesive Wear Test

In this study, tribological properties of samples have been investigated in three steps. In the first step, the effects of only GNP addition to the PPS matrix on tribological properties were evaluated, in order to make a comparison with later steps. In the second step, the effects of both GNP and CF simultaneously addition to PPS matrix composites on tribological properties were evaluated to find out whether GNP alters the interfacial interaction between CF and PPS matrix. In the last step, the effects of the simultaneously addition of GNP, CF and Lotader<sup>®</sup> to the PPS matrix composites on tribological properties were evaluated in order to reveal whether Lotader<sup>®</sup> further alters the interfacial interaction between the reinforcing materials and the PPS matrix.

Coefficient of friction curves and average wear rate values for composites containing only GNP were given in Figure 5 and Table 3. This figure and table show that the addition of 0.1 and 0.3 wt.% GNP to neat PPS did not affect the average wear rate value and COF. However, the addition of 0.5 wt.% GNP to neat PPS significantly reduced the average wear rate value and COF, especially for the first 70 meters of sliding distance. When this result is evaluated together with the tensile test results, it can be concluded that reinforcement and lubrication mechanisms occur differently for GNP reinforced PPS matrix composites. Such that, for the same weight ratios of GNP, while the decrease in tensile strength can be associated with the agglomeration of GNPs; the improvement in tribological properties can be attributed to



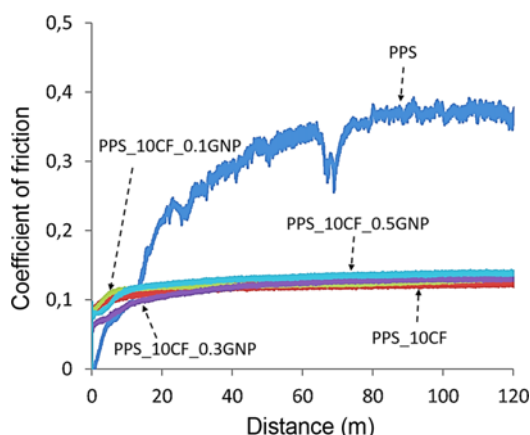
**Figure 5.** Coefficient of friction curves of only GNP included composites.

**Table 3.** Average wear rate values of samples

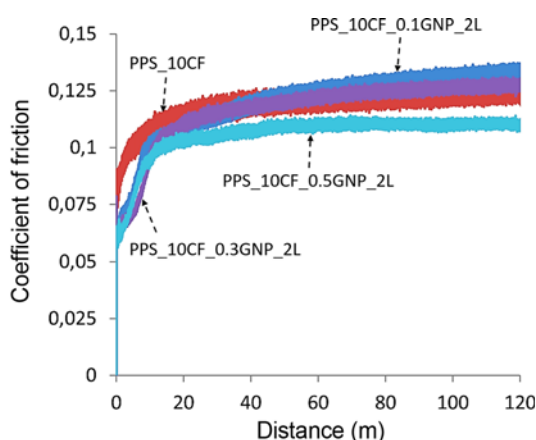
PPS/CF/GNP/Lotader <sup>®</sup>	Average wear rate (mm <sup>3</sup> /N.m.10 <sup>-5</sup> )
PPS	15.027 (±1.335)
PPS-10CF	6.115±1.374
PPS_0.1GNP	15.907±0.553
PPS_0.3GNP	16.813±1.047
PPS_0.5GNP	4.135±0.550
PPS_10CF_0.1GNP	5.638±1.484
PPS_10CF_0.3GNP	5.465±0.540
PPS_10CF_0.5GNP	5.699±0.168
PPS_10CF_0.1GNP_2L	3.615±0.450
PPS_10CF_0.3GNP_2L	5.553±0.009
PPS_10CF_0.5GNP_2L	4.983±0.291

the platelet shape of GNP and the interlayer sliding of these platelets along the friction direction. Because, as mentioned before, there are weak Van der Waals bonds between the two-dimensional layers of GNPs, and the presence of these bonds allows easy sliding of the GNP layers during the wear process [17,44,45].

COF curves and average wear rate values belong to both GNP and CF reinforced PPS matrix hybrid composites were given in the Figure 6 and Table 3. This figure and table show that the addition of only CF to neat PPS considerably decreased the average wear rate value and the COF. This is because, the carbon fibers behave like a solid phase in the polymeric matrix and reduce the contact area between the wear surface and abradant. This reduces the abrasion of the wear surface of the polymer and results in a decrease in the average wear rate value and COF. On the other hand, Figure 6 and Table 3 also show that average wear rate values and COF were not significantly affected by the simultaneously addition of both GNP and CF to neat PPS matrix. As can be seen from the Figure 6 and the Table 3, hybrid reinforced



**Figure 6.** Coefficient of friction curves of GNP and CF included hybrid composites.



**Figure 7.** Coefficient of friction curves of GNP and CF included hybrid composites in the presence of Lotader®.

composites show similar COF tendency as only CF reinforced composites. This may be due to the more dominant

lubricating effect of CF compared to GNP, (for these GNP weight ratios).

COF curves and average wear rate values of hybrid composites (simultaneously containing GNP, CF and Lotader®) were given in Figure 7 and Table 3. This figure and table indicate that with the simultaneously addition of both 0.5 wt.% GNP and 2 wt.% Lotader® to CF reinforced PPS matrix composites, the COF and the average wear rate value decreased. It can be said that this result is due to the reactions between the reactive groups in the structures of the composite constituents, as mentioned earlier [7,43]. Because, by means of the reactions between these reactive groups in the structure of the composite constituents, the interfacial interaction between reinforcement materials and PPS increased. Thus, a more homogeneously dispersion of GNPs and a better orientation of GNP layers, in the PPS matrix, has been provided [39,40]. The more homogenous dispersion of GNPs in the PPS matrix and the better orientation of the GNP layers enabled the GNP layers to slide on each other and the lubricating effect of the GNP to improve. Consequently, the average wear rate value and the COF decreased for composites, simultaneously containing both 0.5 wt.% GNP and 2 wt.% Lotader® [44].

#### Differential Scanning Calorimetry Analysis

DSC analysis was performed and  $T_g$ ,  $T_c$ ,  $T_m$ ,  $\Delta H_c$ ,  $\Delta H_m$  and  $(X_c)_{rel}$  values of composites were given in Table 4, collectively. As can be seen from Table 4, the addition of CF to neat PPS did not significantly alter the  $T_g$  and  $T_m$  values, while the  $T_c$  value of PPS decreased with the addition of CF. In addition, an increase in the  $(X_c)_{rel}$  value of PPS was obtained with the addition of CF. As is known, adding fiber to a neat polymer, at an optimum fiber weight or volume ratio, generally affects the crystallinity degree of the polymer by increasing the amount of nucleation sites and the crystal growth rate. Besides, it lowers the cold crystallization temperature promoting crystallization of the polymer matrix.

**Table 4.** DSC analysis results of samples

PPS/CF/GNP/Lotader®	$T_g$ (°C)	$T_c$ (°C)	$T_m$ (°C)	$\Delta H_c$ (J/g)	$\Delta H_m$ (J/g)	$(X_c)_{rel}$
PPS	87.74	111.19	287.68	8.61	40.77	1 (Ref.)
PPS-10CF	86.49	107.96	285.77	3.31	34.71	1.08
PPS_0.1GNP	88.19	109.43	286.53	2.08	38.33	1.12
PPS_0.3GNP	88.77	109.50	285.73	1.78	38.59	1.14
PPS_0.5GNP	88.25	109.97	287.77	1.71	39.49	1.18
PPS_10CF_0.1GNP	85.91	110.25	285.77	5.59	38.72	1.14
PPS_10CF_0.3GNP	86.06	112.90	285.08	6.66	37.86	1.08
PPS_10CF_0.5GNP	86.25	113.83	286.08	5.45	35.70	1.05
PPS_10CF_0.1GNP_2L	86.06	114.57	285.58	17.82	35.14	0.61
PPS_10CF_0.3GNP_2L	86.92	113.58	285.69	15.15	34.96	0.70
PPS_10CF_0.5GNP_2L	86.4	111.77	285.34	7.51	34.58	0.96



When the data in Table 4 is evaluated according to the statements above, it can be said that the weight ratio of CF in composites (10 wt.%), in this study, is at the optimum level. Therefore, the results in Table 4 can be interpreted as CF increased the amount of nucleation sites and the crystal growth rate in the PPS matrix and reduced the cold crystallization temperature.

It can be observed from Table 4 that the addition of only GNP to neat PPS did not significantly alter the  $T_g$  and  $T_m$  values. However, the  $T_c$  of neat PPS decreased with the addition of GNP, while the  $(X_c)_{rel}$  value increased significantly with the addition of GNP. Additionally, the increasing GNP content in composites increased the  $(X_c)_{rel}$  value. This result is originated from the promoting effect of GNP on the crystallization of PPS matrix.

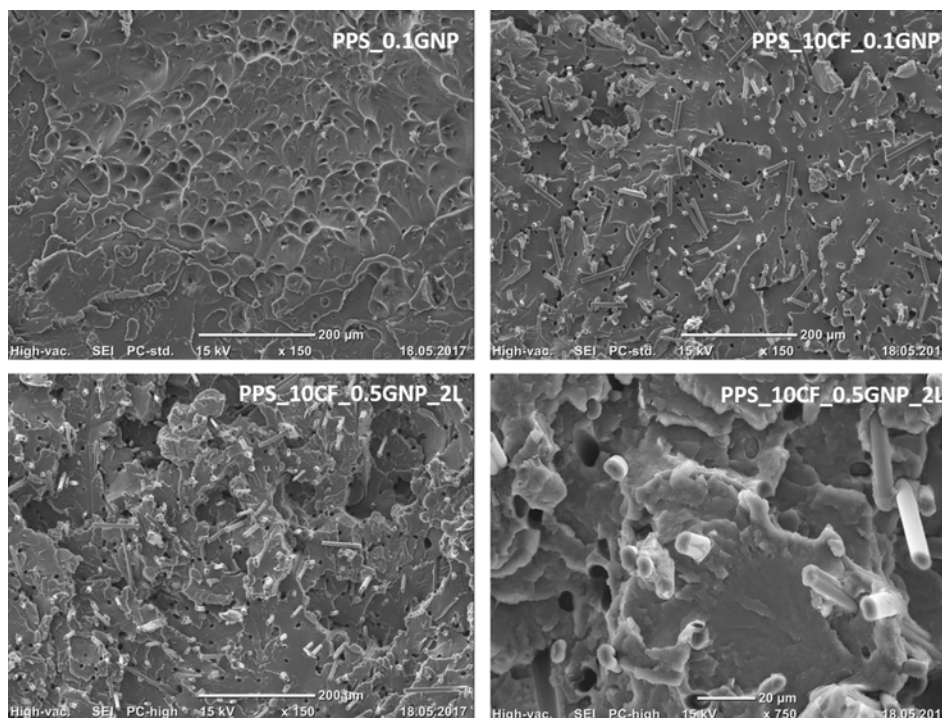
It can be observed from Table 4 that the simultaneously addition of both GNP and CF to PPS did not significantly alter the  $T_g$  and  $T_m$  values compared to  $T_g$  and  $T_m$  values of only CF reinforced PPS matrix composites. Besides, in hybrid composites,  $T_c$  decreased and  $(X_c)_{rel}$  value increased in the presence of 0.1 wt.% GNP, compared to only CF reinforced PPS matrix composites. However, with increasing GNP content in hybrid composites (0.3 and 0.5 wt.% GNP),  $T_c$  increased and  $(X_c)_{rel}$  value decreased. This result is due to the restriction effect of the simultaneous use of CF and GNP on the crystallization behavior of PPS. This restrictive effect takes place according to the following mechanism: the movement of the fibers is inhibited due to the improved fiber-matrix interaction, so the polymer chains around the

fibers cannot be easily oriented, and therefore the crystal growing process is limited. For these reasons, relative  $X_c$  value of composite decreases and  $T_c$  increases because there is more need to heat in order to mobilize the hindered polymer chains.

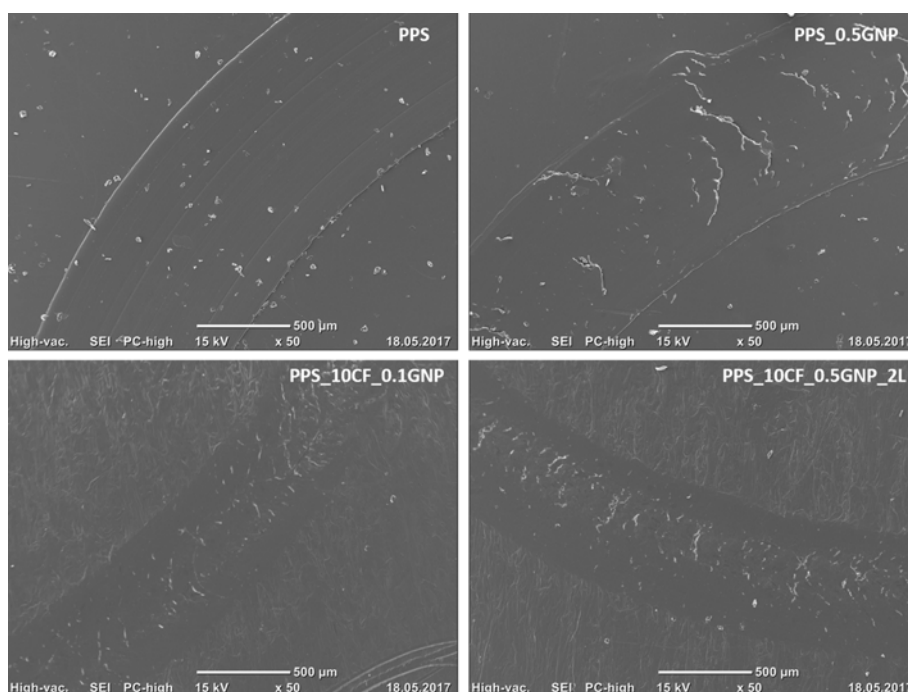
Table 4 shows that the presence of Lotader<sup>®</sup> in hybrid composites did not significantly alter the  $T_g$  and  $T_m$  values. On the other hand, in the presence of Lotader<sup>®</sup>, the  $T_c$  of hybrid composites increased and the  $(X_c)_{rel}$  value decreased significantly. It can be said that the increase in  $T_c$  and significant decrease in  $(X_c)_{rel}$  value, in the presence of Lotader<sup>®</sup>, is due to the improved fiber-matrix interaction and the inhibitory effect of Lotader<sup>®</sup> on the crystallization of PPS.

### Scanning Electron Microscopy Analysis

The fracture surfaces of the samples that were tensile tested were examined by SEM analysis to estimate interfacial adhesion between CF, GNP and PPS, in the presence of Lotader<sup>®</sup>. SEM micrographs were given in Figure 8. It can be seen from Figure 8 that in composites containing only GNP, the GNPs were dispersed in the PPS matrix, in the form of agglomerates. On the other hand, in the case of simultaneously usage of both GNP and CF, GNP agglomerates are not observed. This is due to the improved homogeneous dispersion of GNP when simultaneously use with CF. Figure 8 also shows that in hybrid composites containing Lotader<sup>®</sup>, carbon fibers are embedded in the matrix and most fibers' surface are coated with PPS matrix. According to SEM observations, it can be concluded that there is good wetting



**Figure 8.** SEM micrographs of tensile fracture surfaces of composites.



**Figure 9.** SEM micrographs the worn surfaces of composites.

between CF and PPS matrix in hybrid composites containing Lotader<sup>®</sup>. This good wetting can be shown as a proof of strong fiber-matrix adhesion and good interfacial properties.

Wear traces of composites, which were applied adhesive wear test, were examined by SEM analysis. Micrographs of composites were given in Figure 9. As can be seen from Figure 9, the wear trace of the neat PPS is smooth, while the wear trace has become rough and wavelike with the addition of CF and/or GNP. This is due to the fact that with the addition of CF to PPS, the abrader comes into contact with CF instead of PPS and thus fiber abrasion occurs. Similarly, with the addition of GNP to PPS, the abrader comes into contact with GNP instead of PPS, and inter-layer sliding in multi-layer GNPs takes place. For these reasons, it can be said that the PPS matrix was reinforced by CF and GNP and protected against wear, and accordingly the appearance of the wear trace changed.

### Conclusion

Tensile test results revealed that the addition of only GNP to neat PPS did not increase the tensile strength value (for the used weight percentages of GNP). The tensile test results also showed that the simultaneously addition of both GNP and CF to PPS matrix increased the tensile strength value. In addition, test results revealed that the tensile strength value of hybrid composites increased, in the presence of Lotader<sup>®</sup>. The highest improvement was achieved in composites containing 0.5 wt.% GNP in the presence of Lotader<sup>®</sup>.

The adhesive wear test results revealed that the addition of 0.1 and 0.3 wt.% GNP did not affect the COF of neat PPS, but the addition of 0.5 wt.% GNP significantly reduced COF. Besides, the simultaneously addition of both GNP and CF did not significantly affect COF and hybrid reinforced composites exhibited similar COF tendency as only CF reinforced composites. On the other hand, COF decreased with the simultaneous addition of 0.5 wt.% GNP and CF in the presence of Lotader<sup>®</sup>, compared to the COF of only CF-reinforced composites.

According to the results of the DSC analysis, it was found that while the  $T_c$  value of neat PPS decreased with the addition of only GNP, the  $(X_c)_{rel}$  value increased significantly with the addition of only GNP. Besides, in hybrid composites,  $T_c$  decreased and  $(X_c)_{rel}$  value increased in the presence of 0.1 wt.% GNP, compared to only CF reinforced PPS matrix composites. However, with increasing GNP content in hybrid composites (0.3 and 0.5 wt.% GNP),  $T_c$  increased and  $(X_c)_{rel}$  value decreased. Moreover, in the presence of Lotader<sup>®</sup>, the  $T_c$  of hybrid composites increased and the  $(X_c)_{rel}$  value decreased significantly.

SEM micrographs revealed that in composites containing only GNP, GNPs were dispersed in agglomerate form within the PPS matrix. However, GNP agglomerates were no longer observed in the presence of Lotader<sup>®</sup> and when GNP and CF were used simultaneously.

From the results listed above, it can be concluded that the simultaneously addition of both 0.5 wt.% GNP and 2 wt.% Lotader<sup>®</sup> to CF reinforced composites increases the tensile



strength and COF property of the composites. Thus, the performance of these composites increases, the service life is extended and the usage area is expanded.

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