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Published in: Journal of Applied Polymer Science

*DOI:* 10.1002/app.49821

Published: 10/02/2021

Document Version Peer-reviewed accepted author manuscript, also known as Final accepted manuscript or Post-print

Published under the following license: Unspecified

Please cite the original version:

Tarhini, A., Tehrani-Bagha, A., Kazan, M., & Grady, B. (2021). The effect of graphene flake size on the properties of graphene-based polymer composite films. *Journal of Applied Polymer Science*, *138*(6), Article 49821. https://doi.org/10.1002/app.49821

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# The Effect of Graphene Flake Size on the Properties of Graphenebased Polymer Composite Films

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## ABSTRACT

In this work, the role of graphene flake size on the properties of poly(vinylidene fluoride-cohexafluoropropylene) (PVDF-HFP) composites was studied. Graphene flakes were added to PVDF-HFP using a solution mixing and molding process. By increasing graphene particle size and its concentration in the composites, higher electrical conductivity, in-plane thermal conductivity, and elastic modulus were achieved. Maximum tensile strength was obtained for the composites with average graphene flake size of 2, 5, and 7  $\mu$ m at graphene concentrations of 10 wt%, 5 wt%, and 20 wt%, respectively. Thick flexible composite films (0.2-0.4 mm) with ultrahigh in-plane electrical conductivity (~4500 S/m), in-plane thermal conductivity (~26 W/m/K), and tensile strength (~50 MPa) were obtained for the samples containing the graphene flakes with a larger average particle size of 7  $\mu$ m. To our knowledge, the first two values are larger than any other values reported in the literature for PVDF-based composites.

**Keywords:** Graphene composites, electrical conductivity, thermal properties, mechanical properties.

## **INTRODUCTION**

Graphene, which is a one-atom-thick with  $sp^2$  bonded and densely packed carbon atoms in a honeycomb structure, is a well-known 2-D material. Graphene has a very large specific surface area of 2630 m<sup>2</sup>/g and impressive properties such as high mechanical strength and modulus, coupled with very high thermal and electrical conductivity values that enable its use as a filler in various composites<sup>1-4</sup>. For a free-standing monolayer of graphene, Young's modulus and the intrinsic strength can reach up to 1 TPa and 130 GPa, respectively<sup>1</sup>. These values make graphene 100 times stronger than a steel sheet with the same thickness<sup>5</sup>. A single layer of graphene has an in-plane thermal conductivity of ~5000 W/m/K and electrical conductivity of ~6000 S/cm, both of which are considerably higher than most conductive metals<sup>6–8</sup>. Also, graphene has an extremely high charge carrier mobility (200,000 cm<sup>2</sup>/V/s) which makes it a very promising material for optoelectronic applications<sup>4,9–11</sup>. In contrast, most polymers are electrical insulators and they have very low thermal conductivity values in the range of 0.1- 0.5 W/m/K. Therefore, many studies have examined the effect of adding graphene or graphene-based fillers to polymers to enhance the electrical, thermal, and mechanical properties of the polymer matrix as there is a growing demand for such conductive polymer composites in many new applications (e.g., batteries, electronics, solar cells, and sensors)<sup>12-14</sup>.

Over the last decade, many researchers have incorporated graphene and graphene-based fillers in polymeric matrices to improve physical and thermal properties<sup>1,15</sup>. Table 1 summarizes some of the most successful attempts using functionalized graphene sheet-nanodiamond<sup>16</sup>, reduced graphene oxide<sup>17–21</sup>, graphene flakes<sup>22</sup>, and graphite nanoplatelets<sup>23</sup>. Fan et al. succeeded in improving the electrical conductivity of PVDF reaching 10<sup>-3</sup> S/m using solution mixing followed by casting and compression molding with in-situ reduction for 3.5 vol% of graphene oxide<sup>19</sup>. Li et al. achieved a high electrical conductivity of 0.05 S/m for PVDF composite with 4 wt % of graphite nano-platelets using solution mixing, casting, and hot-pressing<sup>23</sup>. Jung et al. tried to tackle the problem of alignment of graphene flakes in a specific direction as graphene fillers tend to agglomerate due to Van der Waals force of attraction between these fillers, so they fabricated graphene-based PVDF composites using melt compression in an L-shaped tube and succeeded in improving the in-plane thermal and electrical conductivity of the polymer to reach 10 W/m/K and 10 S/m for 20 vol% of graphene flakes in PVDF. To the best of our knowledge, the highest

electrical conductivity value ever achieved was ~3000 S/m by Kumar et al. where they used solution mixing, casting, followed with reduction for large-area graphene oxide (rLGO) with hydriodic acid in PVDF-HFP<sup>21</sup>. They also achieved good mechanical strength of ~54 MPa and high in-plane thermal conductivity of 19 W/m/K for 27.2 wt% of rLGO in PVDF-HFP. The increase in thermal conductivity, electrical conductivity, and mechanical strength was attributed to the good interfacial interaction between PVDF-HFP and graphene-based filler, the high aspect ratio, the proper orientation of graphene-based filler along the composite film direction, and the use of large-area graphene-oxide sheets that have smaller inter-sheet resistance compared to smallarea graphene-oxide sheets<sup>24</sup>. Despite a large number of papers on graphene-based polymer composites, few have comprehensively studied the effect of graphene flake size on the electrical, thermal, and mechanical properties of the polymer composite<sup>1,25</sup>. This paper contains a detailed characterization of polymer properties for three different types of pure graphene with various average flake sizes (7, 5, and 2 µm for G1, G2, and G3, respectively). These graphene flakes were incorporated in PVDF-HFP using simple solution mixing and molding process. The electrical conductivity, thermal conductivity, and mechanical properties of the composite films were measured.

**Table 1:** Electrical, thermal, and mechanical properties for different PVDF and PVDF-based

 composites reported in the literature

PVDF and PVDF-based Composites with graphene-based filler <sup>a</sup>	Filler content	Electrical Conductivity (S/m)	Thermal conductivity (W/m/K)	Mechanical Strength (MPa)	Reference
Solution mixing of GO in PVDF followed by reducing GO and hot pressing at 200 °C	1 vol %	0.64	N.A.	N.A.	17
Mixing GO solution with PVDF then performing casting and compression molding with in-situ GO reduction	3.5 vol %	~10 <sup>-3</sup>	N.A.	N.A.	19

Melt compression of GNF solution in PVDF using an L-shaped tube	20 vol %	30	10	N.A.	22
Mixing GNP solution in PVDF followed by casting and hot pressing	4 wt %	~0.05	N.A.	N.A.	23
Mixing GO in PVDF then performing GO reduction using solar radiation	7 wt %	10	N.A.	N.A.	20
Casting LGO in PVDF-HFP then undergoing a chemical reduction process	27.2 wt %	3000	19.5	54	21
Mixing FGS/NDs with PVDF solution using ultrasonic dispersion followed by hot pressing	45 wt %	7.1×10 <sup>-5</sup>	0.66	N.A.	16

<sup>a</sup> GO: Graphene Oxide; PVDF: Polyvinylidene fluoride; GNF: Graphene nanoflake; GNP: Graphite nanoplatelet; LGO: Large-area Graphene oxide; HFP: Hexafluoropropylene; FGS/NDs: Functionalized Graphene sheets with nanodiamonds filler; N.A.: Not Applicable / Investigated

## **EXPERIMENTAL SECTION**

1. Materials: three different types of pure graphene were purchased: graphene flakes (G1) was purchased from Graphene 3D Lab Inc. (USA) with a purity of 98.5% and specific surface area less than 40 m<sup>2</sup>/g; graphene nano-platelets (G2) Grade M with average specific surface area 120-150 m<sup>2</sup>/g and graphene nano-platelets (G3) Grade C, with an average surface area of 750 m<sup>2</sup>/g were purchased from XGSciences Inc. (USA). The PVDF-HFP copolymer with an average molecular weight of 455,000 g/mol and N,N-dimethylformamide (DMF) with 99.8% purity were purchased from Sigma. A cubical silicon mold with multiple cavities each with a length of 3.5 cm was purchased from Silikomart (Italy).

2. Graphene-based composite films preparation: Graphene powder was added to DMF then the mixture was bath sonicated at 35°C for 30 min. The polymer solution and the graphene dispersion were mixed at high temperature ( $\sim$ 70°C) according to recipes provided in Table 2. The mixture was stirred for 15 min using a magnetic stirrer with an apparent speed of 500-1000 rpm and poured into a cubical silicon mold. The mold was then inserted into an oven at 90°C for 24 hours to fully evaporate DMF <sup>26</sup>.

<b>Table 2:</b> Preparation recipes of graphene-based polymer composite films for three different
types of graphene (G1, G2, and G3)

Graphene type		Graphene	Polymeric	Graphene
Siuphene type	Sample name↓	dispersion	solution	content
		[5 g/L]	[20 g/L]	(wt %)
G1	PVDF-HFP-G1-1	1 ml	25 ml	0.99%
G1	PVDF-HFP-G1-5	5 ml	25 ml	4.8%
G1	PVDF-HFP-G1-10	11 ml	25 ml	9.9%
G1	PVDF-HFP-G1-20	15 ml	15 ml	20%
G2	PVDF-HFP-G2-1	1 ml	25 ml	0.99%
G2	PVDF-HFP-G2-5	5 ml	25 ml	4.8%
G2	PVDF-HFP-G2-10	11 ml	25 ml	9.9%
G2	PVDF-HFP-G2-20	15 ml	15 ml	20%
G3	PVDF-HFP-G3-1	1 ml	25 ml	0.99%
G3	PVDF-HFP-G3-5	5 ml	25 ml	4.8%

G3	PVDF-HFP-G3-10	11 ml	25 ml	9.9%
G3	PVDF-HFP-G3-20	15 ml	15 ml	20%

3. GNF/PVDF-HFP film characterization techniques: The surface morphology was measured at an accelerating voltage of 20 KV by MIRA 3 LMU, a scanning electron microscopy from Tescan (Czech Republic) at different magnifications. The X-ray powder diffraction patterns of each graphene flake were obtained using a D8 ADVANCE from Bruker (Germany) with the angle  $2\theta$ varying from 10° to 60° at room temperature. A Cary 630 attenuated total reflectance-Fourier transform infrared spectroscopy (ATR-FTIR) from Agilent Technologies (USA) was used for collecting data for a spectral range between 600–3600 cm<sup>-1</sup>. Temperature- and power-dependent Raman measurements were performed in the backscattering configuration using a 532-nm laser line from an Ar-ion laser to deduce the thermal conductivities of three samples from each composite. This technique was explained and used by the authors in a previous study and it was explained in detail in the supporting information (SI)<sup>27</sup>. An 850-mm focal length spectrometer with 2400 groves/mm holographic grating was used in the measurements to obtain precise relation between the laser power and Raman shift. Elemental analysis for the graphene type was performed using FlashEA 1112 from ThermoFisher Scientific (US). The process includes generating (for a few seconds) very high temperatures (~1800°C), to allow the conversion of the sample to elemental gases that were determined and measured. The electrical conductivity for each film was measured using a four-point probe system from Ossila (UK), see more detail in the SI. The mechanical properties of the composites were tested using a universal test machine Hounsfield HK-100 (China) with a load cell of 1 KN. Rectangular composites (length: 30 mm, width: 15 mm; thickness: 0.2-0.5 mm) were placed between the clamps of the machine with a gauge length of  $\sim 15$ mm and then extended at a tensile strain rate of 2 mm/min until fracture. The ultimate tensile strength and Young's modulus were obtained from the stress-strain curve <sup>21</sup>.

#### **RESULTS AND DISCUSSION**

As shown in Figure 1, for an individual flake, the average of three different lengths (L1, L2, and L3) was calculated to determine the flake size for that flake, and this data was collected from more than 50 samples. The average particle size of each graphene type was obtained to be  $7 \pm 0.35 \mu m$ ,  $5 \pm 0.25 \mu m$ , and  $2 \pm 0.1 \mu m$  for G1, G2, G3, respectively. ATR-FTIR spectra of the three types

of graphene (G1, G2, and G3) are shown in Figure 2a. All three types had the same functional groups with two representative peaks for graphene at 1547 cm<sup>-1</sup> and 3651 cm<sup>-1</sup> representing the skeletal vibrations of graphene backbone and –OH stretching vibrations, respectively <sup>28</sup>.



**Figure 1**: Statistical analysis of average flake size for the three different types of Graphene (G1, G2, and G3) as a function of the number of flakes taken into account. The average flake size (FS) converges to the asymptotic average values of  $7 \pm 0.35 \,\mu\text{m}$ ,  $5 \pm 0.25 \,\mu\text{m}$ , and  $2 \pm 0.1 \,\mu\text{m}$  for G1, G2, G3, respectively.

Figure 2b shows the XRD patterns of the graphene flakes. A sharp peak at  $2\theta=26.1^{\circ}$  corresponding to the crystalline structure of graphene was observed noticeably for G1 and G2

corresponding to the (002) plane. This peak was relatively weak and broad for the G3 type indicating the low crystallinity of G3. The disappearance of this peak could mean that the distance between graphene sheets is irregular due to a high number of defects or that the number of graphene sheets per flake is very small. Raman spectra from the three types of graphene are consistent with the former explanation. In the Raman spectrum for graphene, the G band occurs due to the in-plane vibrations of sp2 bonded carbon atoms whereas the D band occurs due to the out of plane vibrations sp3 which is resulted by the presence of structural defects<sup>25</sup>. For G3, the intensity of the D peak was higher than the intensity of the G peak, which indicates the poor quality of G3 in comparison to G1 or G2, as shown in Figure 2e. The ratio of the intensity of D peak to G peak, which indicates the number of defects in graphene is the highest ( $I_D/I_G=1.08$ ) for G3, which implies that more sp3 defects are available in G3 than in G1 and G2 ( $I_D/I_G=0.74$  and 0.83 respectively). Further, elemental analysis of the graphene flakes also showed that G3 had more impurities, and the concentration of nitrogen in the sample was noticeably higher than that in G1 and G2 (Figure 2f)<sup>6</sup>.





**Figure 2:** (a) ATR-FTIR analysis for G1, G2, and G3 (b) X-ray diffraction patterns for G1, G3, and G3 (c) Raman signal for G1 (d) Raman signal for G2 (e) Raman signal for G3 (f) Table showing the elemental analysis done on G1, G2, and G3.

Graphene-based PVDF-HFP composites were fabricated using a solution-based method where DMF was used to dissolve each graphene type and the polymer matrix PVDF-HFP, as described in the experimental section. SEM micrographs, presented in Figure 3, showed the uniform distribution of graphene flakes on the surface of the composite films, which have an average thickness of 0.2-0.4 mm. For G1, many flakes were seen overlapping at 5 wt %, 10 wt %, and 20 wt %, respectively. Similar results were obtained for G2 composites in Figure 3(e-h). As for G3 composites, G3 flakes appear to be rounded as shown in Figure 3(i-l). For all composites, more graphene flakes appeared on the top surface with increasing wt % of graphene in these composites. With regards to the cross-sectional view, a stack of graphene flakes appeared to be on top of each other for PVDF-HFP-G1-20 in Figure 4d. Similar results were seen for PVDF-HFP-G2-20 in Figure 4e. However, G3 flakes had a small rounded shape and appeared to be more isotropic than G1 and G2 as seen for PVDF-HFP-G3-20 in Figure 4f.



**Figure 3:** SEM micrographs of Graphene-based polymeric composites for different Graphene types different magnifications (a) PVDF-HFP-G1-1 (b) PVDF-HFP-G1-5 (c) PVDF-HFP-G1-10 (d) PVDF-HFP-G1-20 (e) PVDF-HFP-G2-1 (f) PVDF-HFP-G2-5 (g) PVDF-HFP-G2-10 (h) PVDF-HFP-G2-20 (i) PVDF-HFP-G3-1 (j) PVDF-HFP-G3-5 (k) PVDF-HFP-G3-10 (l) PVDF-HFP-G3-20 [colored figure].

The polymer composites showed very high values of electrical conductivity (Figure 4) and showed an enhanced electrical conductivity in the order of flake size: G1>G2>G3. Comparing to

the values in Table 1, a record electrical conductivity of ~4445 S/m was reported for PVDF-HFP-G1-20. PVDF-HFP is an insulator with a very low electrical conductivity of  $10^{-14}$  (S/m), and the aforementioned electrical conductivity corresponds to a significant improvement making these composites comparable to conductive metals such as stainless steel that has an electrical conductivity of ~ 2×10<sup>6</sup> S/m<sup>21</sup>.

Similar to metallic fillers in polymeric composites, the shape and size of graphene flakes have a significant influence on the electrical conductivity of obtained composites <sup>29</sup>. Lower conductivity is attributed to inferior quality and smaller size of G3 flakes. The former reduces the inherent conductivity of the flakes while the latter means more inter-particle contacts are present in a conductive path resulting in a higher scattering rate of electrons at boundaries. G1 based polymer composites showed the highest electrical conductivity because large G1 flakes have larger density and smaller inter-sheet contact resistance. The electrical conductivity increased further after adding 5 wt% and 10 wt% of graphene but with a slower rate due to typical percolation transition behavior for electrical conductivity in conductive polymer composites <sup>21,30</sup>. Consistent with SEM images in Figure 4(d-f), large planar flakes (G1 and G2) can be aligned in a plane much better than small round-shape (isotropic) graphene flakes (G3).



**Figure 4:** In-plane electrical conductivity as a function of graphene content (wt.%) for three different graphene types (a,b), and a picture showing a light bulb under 9V battery where PVDF-HFP-G1-20 film was used as an electrical connection (c). SEM micrographs for the cross-section of graphene-based polymer composites for different graphene types: (d) PVDF-HFP-G1-20 (e) PVDF-HFP-G2-20 (f) PVDF-HFP-G3-20. [colored figures]



**Figure 5:** The Raman signals of Graphene-based composites (a) PVDF-HFP-G1-20 (b) PVDF-HFP-G2-20 (c) PVDF-HFP-G3-20 at 5 mW and 3 mW laser power (d) G-peak wavenumber versus Temperature for Graphene at different Temperatures (e) Enhancement of in-plane thermal conductivity as a function of Graphene content for three Graphene types G1, G2, and G3. [colored figures]

G1-based polymer composites showed the highest value for the in-plane thermal conductivity, which was also confirmed and reported before using a laser flash method  $^{27,30}$ . An ultra-high value of ~26 W/m/K was achieved, which again is the highest measured values for in-plane thermal conductivity for PVDF composites. For the same graphene wt%, the in-plane thermal conductivity value was lower for G2 and G3, respectively indicating that the average flake size also has a direct effect on the in-plane thermal conductivity values of the composites. With larger graphene flakes, the density of filler/matrix interfaces is smaller, and the interfacial heat resistance (Kapitza resistance) will have less of an effect on the thermal conductivity<sup>31</sup>. G1 composites were more aligned and stacked in an overlapping structure than G2 composites, as shown in Figures 4(d-e). Also, just as with electrical conductivity, the high defect concentration as confirmed by elemental analysis in Figure 2f for G3 decreased the in-plane thermal conductivity of the flakes themselves obtained due to an increased rate of phonon-defect scattering mechanism  $^{32}$ .



**Figure 6:** Mechanical properties as a function of Graphene content (wt %) for three different Graphene types (a) G1 (b) G2 (c) G3 (d) Two photos showing the flexibility of PVDF-HFP-G1-20. [colored figures]

Typical stress-strain curves for the composites are shown in Figure 6 with the values for the tensile strength and Young's modulus as a function of graphene content to the right. Figure 6a shows that increasing the concentration of the graphene flakes increases Young's modulus and the tensile strength; the tensile strength increased to  $\sim 50$  MPa for PVDF-HFP-G1-20. For this sample, the large graphene flakes, with the uniform dispersion of the flakes as shown in Figure 3d, with the smaller interlayer distance and cross-sectional area with a denser structure as shown in Figure 4d, and the orientation of G1 in the stretch direction enhanced the tensile and modulus properties of the composite film compared to other composites. For graphene contents higher than 20 wt %, the mechanical properties of the G1 composites are expected to decrease and this observation was presented by the authors in a previous paper<sup>30</sup>. The tensile strength for G1 composites was higher than the tensile strength for G2 and G3 composites with the same graphene loading. For example, for 5 wt % G1, the ultimate strength was ~26 MPa, whereas it was 24 MPa and 14.5 MPa for G2 and G3, respectively. These results showed that the size of the graphene flakes can significantly affect the tensile strength of the composites as large flakes tend to have a more compact and aligned structure for the composite than smaller flakes <sup>24</sup>. Other researchers have suggested that there is usually an effective critical length for these graphene flakes for efficient reinforcement because of wrinkles and other defects<sup>1,33</sup>. Of course, the significant enhancement in tensile strength for G1 composites is also a result of both proper dispersion and adhesion with PVDF-HFP <sup>30</sup>. Inferior response was seen for small flakes in G2 and G3 composites. In G2 and G3 composites, the tensile strength was the highest at 5 wt% and 10 wt%, respectively. This lower response in mechanical properties for these composites may be due to the poor dispersion of small size flakes which may agglomerate during incorporation to the polymer matrix PVDF-HFP<sup>34</sup>. Chunks of graphene (like G3 flakes as seen in Figure 4f) can act as stress-concentration points causing these composites to fail especially with high graphene loading (wt %) where graphene flakes tend to agglomerate due to the strong Van der Waals force of attraction between graphene particles <sup>16</sup>.

### CONCLUSION

A series of PVDF-HFP graphene-based composites using three graphene types with respective average flake size (7  $\mu$ m, 5  $\mu$ m, and 2  $\mu$ m) were prepared using a solution mixing and molding process. Due to vigorous mixing and slow evaporation process, the graphene flakes aligned in

the plane of the film as with more alignment for the larger, more planar flakes. In-plane electrical and thermal conductivity values improved significantly with increasing the content of graphene (Figure S1 in the supporting information). Large-area graphene flakes increase the in-plane electrical and thermal conductivity values more because of a reduction in carriers scattering caused by inter-particle resistance. Lower conductivity is attributed to inferior quality and smaller size of G3 flakes. The former reduces the inherent conductivity of the flakes while the latter means more inter-particle contacts are present in a conductive path resulting in a higher scattering rate of electrons and phonons at boundaries. The mechanical properties also increased with the addition of graphene depending on the graphene flake sizes: G1>G2>G3. However, for small graphene flakes (G2 and G3) the tensile strength dropped above certain graphene content due to voids and aggregates of graphene filler that likely act as stress-concentration points.

#### ACKNOWLEDGMENTS

Authors declare no conflict of interest and would like to acknowledge the financial supports from the American University of Beirut and the Lebanese National Council for Scientific Research.

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