



# Sustainable Approach: Utilizing Plastic Waste-Derived rGO for Multifunctional Thermoplastic Nanocomposites

Rajshree Vijayvargiya, Alok K Sahu\* and Ambika Joshi

Central Institute of Petrochemical Engineering & Technology (CIPET), Raipur, CG, India

Received: 04.05.2024 Accepted: 22.06.2024 Published: 30.06.2024

\*aloksahu@cipet.gov.in



## ABSTRACT

Presently, the escalating global demand for plastic has led to an alarming rise in waste plastics, posing a significant threat to the environment. Materials like PP, PE, and polystyrene (PS) are identified as key precursors for producing carbon nanomaterials such as CNTs, carbon nanofibers, and graphene Nano sheets (GNs) derived from plastic waste. This study investigates the impact of reduced Graphene Oxide (rGO), derived from plastics waste, on the various properties of polyolefin nanocomposites. This research involved preparing high-density polyethylene (HDPE) reinforced with rGO nanocomposites, loading a very low% of rGO varying as 0.025wt%, 0.050 wt%, and 0.075 wt% by using Co-rotating twin-screw extruder and the mechanical, thermal, electrical and morphological properties were assessed using UTM, TGA, Dielectric breakdown tester, and SEM respectively and also compared with the virgin HDPE material. The findings demonstrated that the significant enhancements in tensile strength, flexural strength and modulus were observed at a 0.075% rGO loading, surpassing other compositions. This improvement likely resulted from enhanced stress transfer from the polymer matrix to the nanofiller. TGA analysis revealed that all compositions of HDPE/rGO nanocomposites exhibited higher thermal stability compared to their virgin counterparts. It is due to the rigidity in polymer chains movement because of dispersion of rGO, shows restriction in thermal vibrations of the C–C bond of the HDPE matrix. However, a slight decrease in dielectric strength as the % of rGO increased, due to the induced polarization effect and intrinsic high dielectric constant of rGO. Despite this, these nanocomposites, with their exceptional flexural and tensile properties even at low levels of rGO incorporation, offer cost-effective prospects in plastic industries.

**Keywords:** Plastics waste; rGO; Nanocomposites; Mechanical properties.

## 1. INTRODUCTION

The current scenario of plastics waste has become a challenging issue, leading to various environmental problems. Fortunately, the high carbon content within plastics offers a new avenue for improved waste management by creating value-added products. Recently, several researchers have explored the synthesis of carbon nanomaterials from waste plastics, including carbon nanotubes, graphene nanosheets, carbon spheres, and carbon nanofibers (Wu *et al.* 2014; Zhuo *et al.* 2014; Gong *et al.* 2014b; Gong *et al.* 2014a; Sharma *et al.* 2014). While various methods exist for synthesizing graphene, such as chemical techniques, chemical vapor deposition, and electrochemical exfoliation techniques (Madurani *et al.* 2020), they tend to be time-consuming and result in high production costs. Consequently, using rGO produced through these methods becomes challenging for low-cost industrial applications. In the present study, we used Reduced Graphene Oxide (rGO) which is derived from plastic waste through a two-step pyrolysis process (Garg *et al.* 2022). This method is cost-effective, making the current form of rGO suitable for a variety of real-life applications. Not only does it help to reduce the density of solid plastics waste in the

environment, but it also presents a practical way to convert waste to wealth, particularly for high-end concrete applications.

Polyethylene (PE) stands out as one of the most commonly utilized thermoplastics, albeit with a mechanical strength relatively lower than that of other thermoplastics. Despite its lower mechanical strength, polyethylene is recognized for its high ductility and remarkable impact strength. The incorporation of graphene or its derivatives has been identified as a means to enhance the mechanical properties of resulting nanocomposites (Trivedi *et al.* 2022). Mittal *et al.* (2016) employed the melt mixing method to fabricate a PE/graphene nano-composite. Three polyethylene samples (PE1, PE2, and PE3) characterized by different degrees of crystallinity (61%, 56%, and 52%) were utilized in creating nano-composites with graphene as the nano-filler. The study revealed that the degree of crystallinity in PE1 was improved by incorporating 2 to 4 wt% graphene. Furthermore, the PE1 composite with 4 wt% graphene exhibited enhanced tensile modulus. The peak stress and elongation also experienced improvement with the addition of graphene to the PE matrix. In another study, (Hari *et al.* 2021) developed LDPE/GO nano-

composites using the melt mixing method. Various samples were prepared with different loadings of GO, ranging from 0.5 to 3 wt%. The results indicated that the incorporation of GO in the LDPE matrix contributed to increased tensile strength, modulus of elasticity, and storage modulus. Conversely, the tan delta peak was found to decrease.

In the present investigation, initially an attempt was made to create nanocomposites using High Density Polyethylene (HDPE) reinforced with varying weight percentages of reduced graphene oxide (rGO), obtained from plastic waste, through a melt blending process. The dispersion of rGO in Polyolefins (HDPE) was achieved by using a co-rotating twin-screw extruder. Test specimen for testing is prepared by an Injection Moulding machine. The impact of rGO on the mechanical, thermal, electrical and morphological properties of HDPE was investigated using a Universal Testing Machine (UTM), Impact Tester, TGA, Dielectric tester and Scanning Electron Microscopy (SEM). The obtained results were then compared with those of the virgin HDPE material.

## 2. EXPERIMENTAL

### 2.1 Materials and Methods

High Density Polyethylene (HDPE) was procured from Indian Oil Corporation Limited, P80 Extrusion grade, having density of  $0.900 \text{ gm/cm}^3$  at room temperature. rGO derived from Plastics waste, was supplied by Shivi Graphene. The specifications of the supplied rGO are as follows:

- Colour: Black
- Number of layers: 2-5
- Thickness: 1-2.1 nm according to AFM
- Pore diameter: 2.154 nm

### 2.2 Preparation of HDPE/rGO Nanocomposites by melt blending

Initially, virgin HDPE was mixed with various weight ratios (0.025%, 0.050%, & 0.075%) of rGO after that homogeneous HDPE/rGO mixture was then introduced into a high-performance co-rotating intermeshing twin-screw extruder (model MG20, manufactured by SAN Engineering) for melt extrusion. The extrusion process maintained a temperature range of  $200 \text{ }^\circ\text{C}$  to  $220 \text{ }^\circ\text{C}$  with a screw speed of 370 rpm. The resulting HDPE/rGO extrudates were granulated and utilized for sample preparation. Test specimens for analyzing mechanical properties were created using an injection molding machine (Model – STD-90, manufactured by S & T Engineering, India). The barrel temperature was maintained at  $190 \text{ }^\circ\text{C}$ ,  $200 \text{ }^\circ\text{C}$ ,  $215 \text{ }^\circ\text{C}$ , and  $220 \text{ }^\circ\text{C}$  from hopper to nozzle, with a cycle time of 38 seconds. A simple schematic representation of the nanocomposite preparation process is illustrated in Fig. 1

(a & b). Before testing, the test specimens underwent initial conditioning at  $23 \pm 2 \text{ }^\circ\text{C}$  and  $50 \pm 5\% \text{ RH}$  for 24 hours.

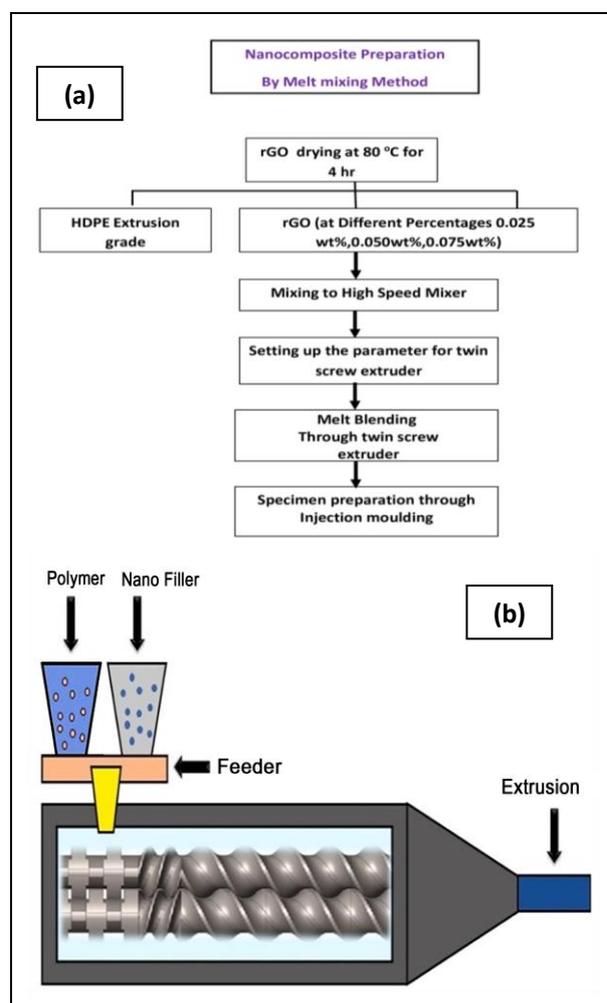


Fig. 1: Schematic representation of the preparation process of Nanocomposites

### 2.3 Testing of mechanical Properties

Mechanical properties such as tensile strength, flexural strength and impact strength of prepared HDPE/rGO nanocomposites are evaluated. Tensile strength is determined with the help of Tinius Olsen Universal testing machine model 25ST at room temperature with a gauge length of 50 mm and crosshead speed of 5 mm/min. Tensile test are evaluated according to the standard ASTM D638 using dumb-bell shaped specimens. Flexural properties are evaluated according to ASTM D790 using Tinius Olsen universal testing machine model 25ST. The dimensions of the specimen were 127 mm in length, 12.7 mm in width and 3mm thickness at room temperature. Impact properties are evaluated according to ASTM D-256 using an Impact tester machine (Presto Izod Tester). The dimensions of the specimens are  $64 \times 12.7 \times 3.2 \text{ mm}$  for izod at room temperature. The tests of mechanical properties and

physical properties were conducted five times and the mean of these results is reported in the present work.

## 2.4 Thermo Gravimetric Analysis (TGA)

The thermal stability and degradation behavior of PP/rGO and HDPE/rGO nanocomposites are studied with the help of Perkin-Elmer Pyres TGA. The TGA measurements have been conducted with a constant heating rate of 10 °C/min under nitrogen atmosphere from 50 to 800 °C.

## 2.5 Testing of Electrical Properties

Dielectric strength has been evaluated according to ASTM D-149 using a Di-electric breakdown Tester (Arcon Enterprises). The dimensions of the specimens are 100 X 100 mm with 3 mm thickness for testing at room temperature.

## 2.6 Phase Morphology

The surface morphology of the tensile fractured surface was performed on SEM (JEOL JSM 6490LV) with an accelerating voltage of 10 kV. The tensile fractured surface of the nanocomposite was gold coated with the help of a gold sputtering unit to enhance the emission of secondary electrons and avoid the charging effect.

## 3. RESULTS AND DISCUSSION

### 3.1 Tensile Strength and Tensile Modulus

Tensile properties HDPE/rGO nanocomposites of different compositions are given in Table 1 & Fig. 2 respectively. It is evident from the results that the maximum improvement (i.e., 11%) in tensile strength is observed at 0.075 wt% rGO loading as compared to other 0.025 wt%, 0.050 wt% rGO filled HDPE nanocomposites. This improvement in tensile strength may be due to stress transfer from polymer matrix to nanofiller. It is an established fact that the effective stress transfer between the polymer matrix and nanofiller entirely depends upon the interfacial interaction between the polymer and nanofillers as well as on the dispersion of nanofiller over the entire polymer matrix (Mishra *et al.* 2023).

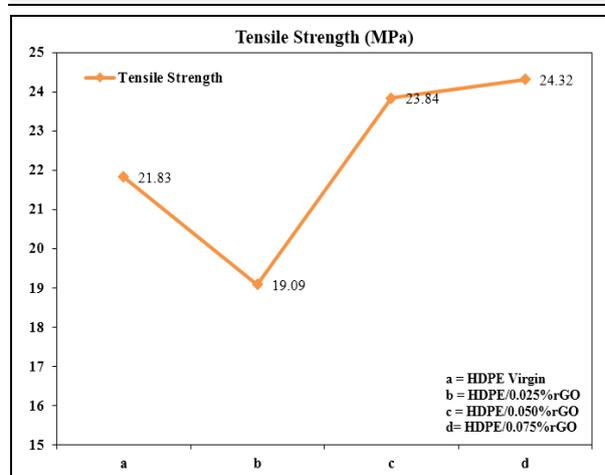
### 3.2 Flexural Properties

Like tensile properties, flexural properties of rGO filled HDPE nanocomposites were investigated. The flexural strength denotes the maximum stress that the material has experienced before yield or fracture. The HDPE/0.075% rGO nanocomposites illustrate the highest increase in flexural properties of the nanocomposites as shown in Table 2 & Fig. 3. The flexural strength and flexural modulus are recorded as

119 and 4156 MPa which show 91% and 152% increments respectively as compared to that of virgin HDPE. This improvement in flexural strength and modulus may be due to stress transfer from polymer matrix to nanofiller.

**Table 1. Tensile properties of HDPE virgin and different composition of HDPE/rGO**

Sample code	Tensile Strength (MPa)	Increase %	Tensile Modulus (MPa)
HDPE virgin	21.83		320.43
HDPE/0.025% rGO	19.09	-0.095	269.66
HDPE/0.050% rGO	23.84	-0.092	285.68
HDPE/0.075% rGO	24.32	11	310.85



**Fig. 2: Comparative results of Tensile Strength of different composition of HDPE/rGO**

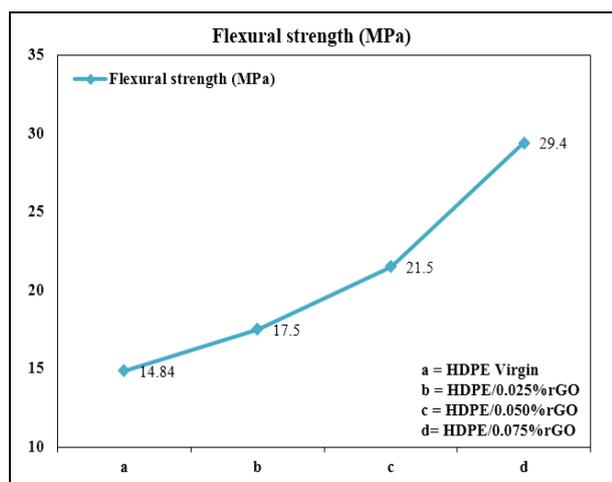
**Table 2 Flexural properties and Impact strength of HDPE virgin and different composition of HDPE/rGO**

Sample code	Flexural strength (MPa)	Increase %	Flexural modulus (MPa)	Increase %	Izod Impact (Notched) (J/M)
HDPE virgin	14.84		812.5		247.08
HDPE/0.025% rGO	17.5	17	973.5	20	241.0
HDPE/0.050% rGO	21.5	44	1515.0	86	240.43
HDPE/0.075% rGO	29.4	91	2050.0	152	239.75

### 3.3 Impact strength

The impact test shows how much energy can be absorbed by a polymer before fracture. If the amount of energy is low, it can be concluded that the material is

more fragile and has a high sensitivity to impact forces. On the other hand, if the amount of energy absorbed is high, the material is soft and deformable and can bear higher impact force (high toughness). Table 2 also illustrates the effect of rGO on the impact strength of nanocomposites. The impact strength slightly declined with an increasing amount of rGO up to 0.075 wt%, which is recorded as 239.75 J/m for and 56 J/m for 0.075 wt% rGO. However, the impact strength decreases at higher rGO loading (0.075% wt%) due to the agglomeration of nanofillers.



**Fig. 3: Comparative results of Flexural Strength of different composition of HDPE/rGO**

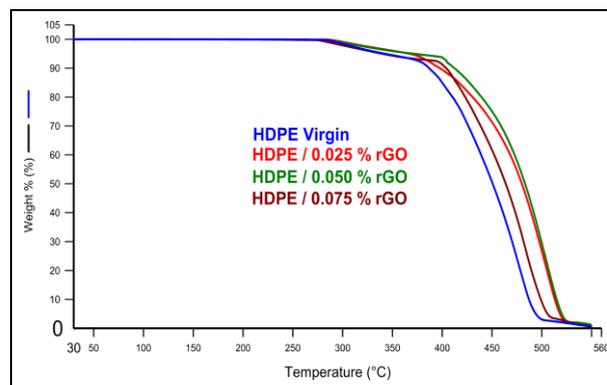
**Table 3. TGA results of different composition of HDPE/rGO**

Sample code	TGA Analysis residue at 520 °C (%)
HDPE virgin	0.09
HDPE/0.025%rGO	0.33
HDPE /0.050%rGO	0.49
HDPE /0.075%rGO	0.44

### 3.4 Thermal Properties Analysis

To investigate the effect of rGO on the thermal properties HDPE/rGO nanocomposites has been analyzed by TGA and the results are given in Table 3 and Fig. 4. It has been observed that HDPE/rGO nanocomposites of all compositions demonstrated higher thermal stability as compared to its virgin counter-part. It is observed from the results that maximum residue 0.49% at 521 °C is observed for 0.050 wt% rGO contents. The 5% degradation of HDPE/0.050 wt% rGO nanocomposite was recorded at 400 °C, while for virgin HDPE, the 5% degradation was observed at 300 °C. The incorporation of rGO provides a path in reducing the chain flexibility and inter chain slippage of the polymer matrix by imposing large numbers of restricted sites which results in reducing the thermal vibration of C-C bond. Earlier investigators (Mishra *et al.* 2018; Paraskar *et al.* 2020; Mishra *et al.* 2023) have also advocated about the barrier properties of the nanofiller that may be

responsible for improved thermal stability of the developed nanocomposites.



**Fig. 4: Thermograph of virgin HDPE and HDPE/rGO different compositions**

### 3.5 Electrical Properties Analysis

The effect of rGO on the electrical properties HDPE/rGO nanocomposites have been analyzed by Dielectric breakdown tester and the results are given in Table 4. Dielectric strength is measured as the maximum voltage required to producing a dielectric breakdown through a material. Higher dielectric strength corresponds to better insulation properties of thermoplastic materials. But due to the induced polarization effect and intrinsic high dielectric constant of reduced graphene oxide (He *et al.* 2018) there is slight decrease in dielectric strength from 8.69 kV/mm to 7.85 kV/mm by increasing the concentration of rGO in the HDPE/rGO Nanocomposites.

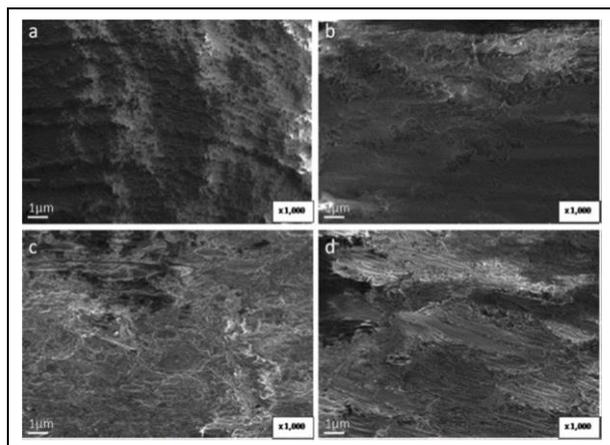
**Table 4. Electrical Properties of HDPE virgin and different composition of HDPE/rGO nanocomposites**

Sample code	Dielectric Strength (kV/mm)
HDPE virgin	8.69
HDPE/0.025%rGO	8.49
HDPE /0.050%rGO	7.67
HDPE /0.075%rGO	7.85

### 3.6 Phase Morphology

It has also been evident from the SEM images in Fig. 5 (a,b,c,d) that a uniform surface is observed in the case of 0.050 wt% loading of rGO (Fig. 5c) while greater addition (0.075 wt%) of rGO leads to a bigger average domain sizes and uneven surface (Fig. 5d) due to agglomeration of nanostructures (Mishra *et al.* 2023).

The smallest average domain sizes of 0.050% prove the uniform dispersion of nanofillers in the matrix, which results in improved interaction of interfaces of the polymers (Mishra *et al.* 2023). The improved interaction of the interface of the polymer optimizes the load transferability from the polymer matrix to nanofillers.



**Fig. 5: Scanning electron micrographs of a tensile fractured surface of a Virgin HDPE; b HDPE/0.025%; c HDPE/0.050; and d HDPE/0.075% nanocomposites**

#### 4. CONCLUSION

The study on HDPE/rGO nanocomposites revealed notable improvements in tensile and flexural strength, particularly at a 0.075% rGO loading, surpassing other compositions. Enhanced stress transfer from the polymer matrix to the nanofiller likely contributed to this improvement. TGA analysis indicated increased thermal stability in all HDPE/rGO nanocomposites compared to virgin counterparts, attributed to improved interfacial interaction and restricted thermal vibrations. Despite a slight decrease in dielectric strength with higher rGO concentration, these nanocomposites exhibit exceptional properties, making them cost-effective for plastic pipe industries and PE water tank manufacturing. Plastics waste derived Reduced Graphene oxide (rGO)-based HDPE nanocomposites have garnered significant attention in Plastics industries by offering several important advantages:

- Enhanced flexural and tensile Properties.
- Lightweight
- Corrosion Resistance
- Sustainability and Environmental Benefits

#### 5. FUTURE ASPECTS

Previous research showed that graphene-based nanocomposites are employed as additives in coatings and paints. So, in future studies, rGO which is derived from plastics waste will mix in metallic paint by solvent mixing method to enhance corrosion resistance, wear resistance, and barrier properties of metals. In future we will also prepare the rGO-based ABS nanocomposites for automotive industries for lightweight and strong materials. Their high strength-to-weight ratio and exceptional mechanical properties contribute to fuel

efficiency, reduced emissions, and improved structural integrity.

#### FUNDING

This research received no specific grant from any funding agency in the public, commercial, or not-for-profit sectors.

#### CONFLICTS OF INTEREST

The authors declare that there is no conflict of interest.

#### COPYRIGHT

This article is an open-access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (<http://creativecommons.org/licenses/by/4.0/>).



#### REFERENCES

- Garg, K. K., Pandey, S., Kumar, A., Rana, A., Sahoo, N. G., Singh, R. K., Graphene nanosheets derived from waste plastic for cost-effective thermoelectric applications, *Results Mater.* 13, 100260 (2022). <https://doi.org/10.1016/j.rinma.2022.100260>
- Gong, J., Liu, J., Jiang, Z., Feng, J., Chen, X., Wang, L., Mijowska, E., Wen, X., Tang, T., Striking influence of chain structure of polyethylene on the formation of cup-stacked carbon nanotubes/carbon nanofibers under the combined catalysis of CuBr and NiO, *Appl. Catal. B Environ.* 147, 592–601 (2014a). <https://doi.org/10.1016/j.apcatb.2013.09.044>
- Gong, J., Liu, J., Wen, X., Jiang, Z., Chen, X., Mijowska, E., Tang, T., Upcycling Waste Polypropylene into Graphene Flakes on Organically Modified Montmorillonite, *Ind. Eng. Chem. Res.* 53(11), 4173–4181 (2014b). <https://doi.org/10.1021/ie4043246>
- Hari, B. ., Mahesh Kumar, K. ., Krishnamurthy, K., Sathish Kumar, P., Gobinath, V. K., Sachinbala, R., Rajasekar, R., Influence of graphene oxide on the morphological and mechanical behaviour of compatibilized low density polyethylene nanocomposites, *Mater. Today Proc.* 39, 1487–1493 (2021). <https://doi.org/10.1016/j.matpr.2020.05.378>
- He, Z., Yu, X., Yang, J., Zhang, N., Huang, T., Wang, Y., Zhou, Z., Largely enhanced dielectric properties of poly(vinylidene fluoride) composites achieved by adding polypyrrole-decorated graphene oxide, *Compos. Part A Appl. Sci. Manuf.* 104, 89–100 (2018). <https://doi.org/10.1016/j.compositesa.2017.10.029>

- Madurani, K. A., Suprpto, S., Machrita, N. I., Bahar, S. L., Illiya, W., Kurniawan, F., Progress in Graphene Synthesis and its Application: History, Challenge and the Future Outlook for Research and Industry, *ECS J. Solid State Sci. Technol.* 9(9), 093013 (2020).  
<https://doi.org/10.1149/2162-8777/abbb6f>
- Mishra, R. M., Ahamad, A., Vijayvargiya, R., Pandey, K. N., Mishra, S., Rai, J. S. P., Effect of graphene on various properties of binary blend of polyetherimide/siliconerubber reinforced with halloysite nanotubes, *Polym. Bull.* 80(5), 5469–5479 (2023).  
<https://doi.org/10.1007/s00289-022-04291-2>
- Mishra, R. M., Rai, J. S. P., Preparation and properties of nanocomposites based on polyetherimide (PEI)/silicone rubber reinforced with halloysite nanotubes, *Indian J. Pure Appl. Phys.* 56(10), 787–791 (2018).  
<https://doi.org/10.56042/ijpap.v56i10.18499>
- Mittal, V., Kim, S., Neuhofer, S., Paulik, C., Polyethylene/graphene nanocomposites: effect of molecular weight on mechanical, thermal, rheological and morphological properties, *Colloid Polym. Sci.* 294(4), 691–704 (2016).  
<https://doi.org/10.1007/s00396-015-3827-x>
- Paraskar, P., Bari, P., Mishra, S., Influence of amine functionalized graphene oxide on mechanical and thermal properties of epoxy matrix composites, *Iran. Polym. J.* 29(1), 47–55 (2020).  
<https://doi.org/10.1007/s13726-019-00772-w>
- Sharma, S., Kalita, G., Hirano, R., Shinde, S. M., Papon, R., Ohtani, H., Tanemura, M., Synthesis of graphene crystals from solid waste plastic by chemical vapor deposition, *Carbon N. Y.* 72, 66–73 (2014).  
<https://doi.org/10.1016/j.carbon.2014.01.051>
- Trivedi, D. N., Rachchh, N. V., Graphene and its application in thermoplastic polymers as nano-filler-A review, *Polymer (Guildf)*. 240, 124486 (2022).  
<https://doi.org/10.1016/j.polymer.2021.124486>
- Wu, C., Nahil, M. A., Miskolczi, N., Huang, J., Williams, P. T., Processing Real-World Waste Plastics by Pyrolysis-Reforming for Hydrogen and High-Value Carbon Nanotubes, *Environ. Sci. Technol.* 48(1), 819–826 (2014).  
<https://doi.org/10.1021/es402488b>
- Zhuo, C., Levendis, Y. A., Upcycling waste plastics into carbon nanomaterials: A review, *J Appl Polym Sci.*  
<https://doi.org/10.1002/app.39931>