Research Article

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

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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 polypropylene, polyethylene, and polystyrene are identified as key precursors for producing carbon nanomaterials such as carbon nanotubes, carbon nanofibers, and graphene nano sheets 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 0.025 wt%, 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 0.075% rGO loading, surpassing other compositions. This improvement likely resulted from enhanced stress transfer from the polymer matrix to the nanofiller. The 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 chain movement because of dispersion of rGO, leading to 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; Reduced graphene oxide; 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 reduced graphene oxide (rGO) produced through these methods becomes challenging for low-cost industrial applications. In the present study, we used rGO 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 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 nano-composites (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 rGO, obtained from plastic waste, through a melt blending process. The dispersion of rGO in HDPE was achieved by using a co-rotating twin-screw extruder. Test specimen was 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 having a density of 0.900 gm/cm^3 at room temperature was procured from Indian Oil Corporation Limited, P80 Extrusion grade. rGO derived from plastic 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 and the 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 °C to 220 °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 fabricated using an injection molding machine (Model – STD-90, manufactured by S & T Engineering, India). The barrel temperature was maintained at 190 °C, 200 °C, 215 °C, and 220 °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 ± 2 °C and 50 ± 5% relative humidity for 24 hours.

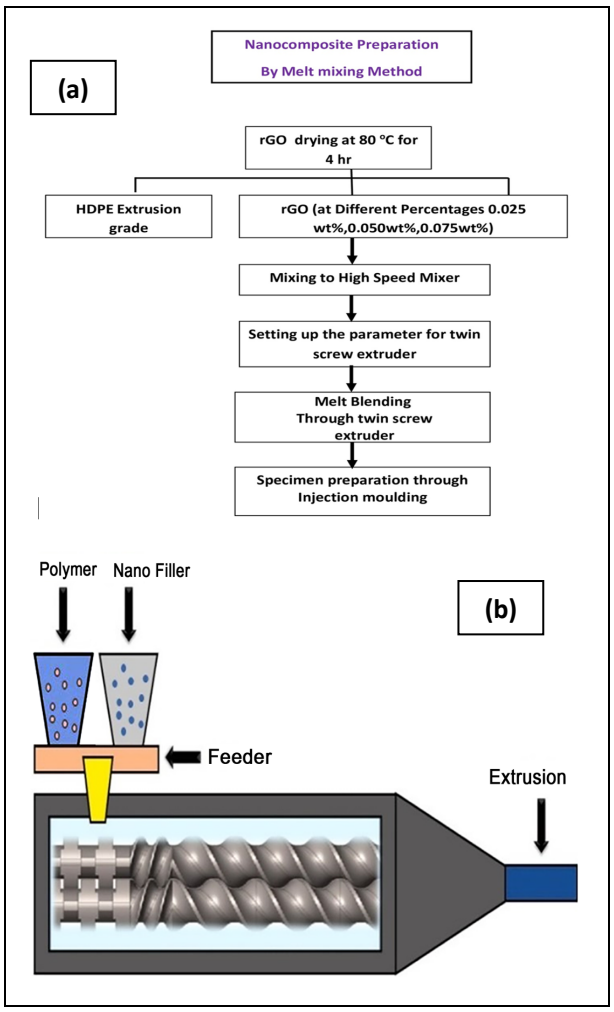


Fig. 1: Schematic representation of the preparation process of Nanocomposites

2.3 Testing of Mechanical Properties

The mechanical properties such as tensile strength, flexural strength and impact strength of prepared HDPE/rGO nanocomposites were evaluated. Tensile strength was 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 was evaluated according to the standard ASTM D638 using dumb-bell shaped specimens. Flexural properties were 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 3 mm thickness at room temperature. Impact properties were evaluated according to ASTM D-256 using an impact tester machine (Presto Izod Tester).The dimensions of the specimens were 64 × 12.7 × 3.2 mm for izod at room temperature. The tests for evaluating mechanical

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

2.4 Thermo Gravimetric Analysis (TGA)

The thermal stability and degradation behavior of PP/rGO and HDPE/rGO nanocomposites were studied with the help of a 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

The dielectric strength was evaluated according to ASTM D-149 using a Di-electric breakdown Tester (Arcon Enterprises). The dimensions of the specimens were 100 × 100 mm with 3 mm thickness.

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

The tensile properties of 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 nanocomposite illustrated the highest increase in flexural properties of the nanocomposites as shown in Table 2 & Fig. 3. The flexural strength and flexural modulus were 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

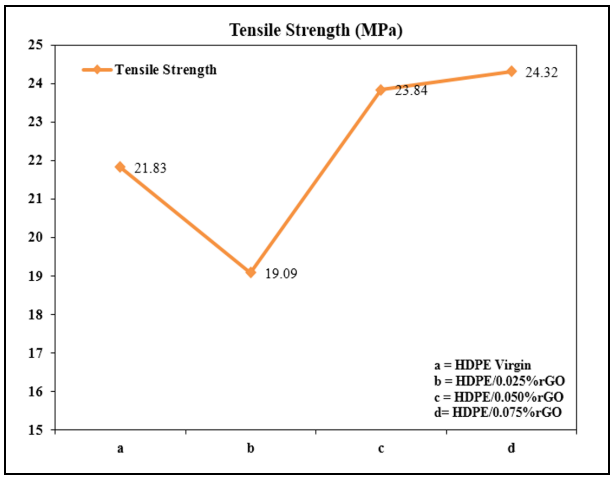


Fig. 2: Comparative results of tensile strength of different composition of HDPE/rGO

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.

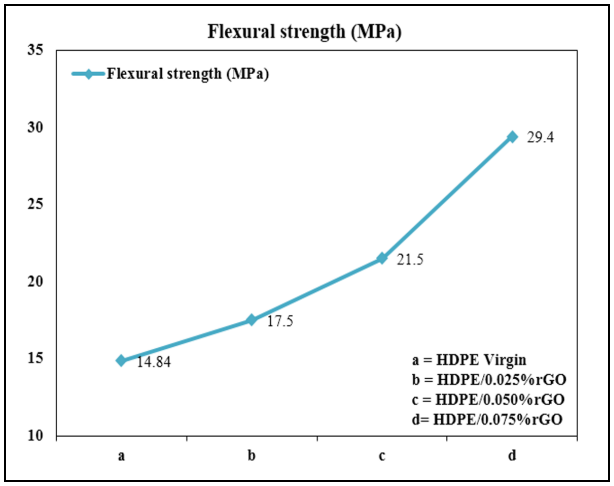


Fig. 3: Comparative results of flexural strength of different composition of HDPE/rGO

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 were 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 counterpart. It is observed from the results that maximum residue of 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 works (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.

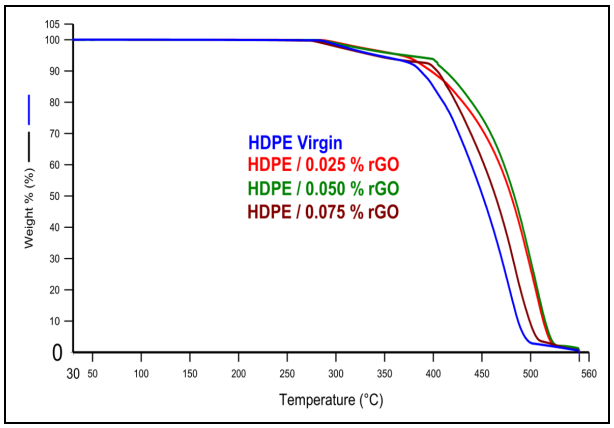


Fig. 4: Thermograph of virgin HDPE and HDPE/rGO different compositions

3.5 Electrical Properties

The effect of rGO on the electrical properties HDPE/rGO nanocomposites 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 is 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 increased addition (0.075 wt%) of rGO leads to a high 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.

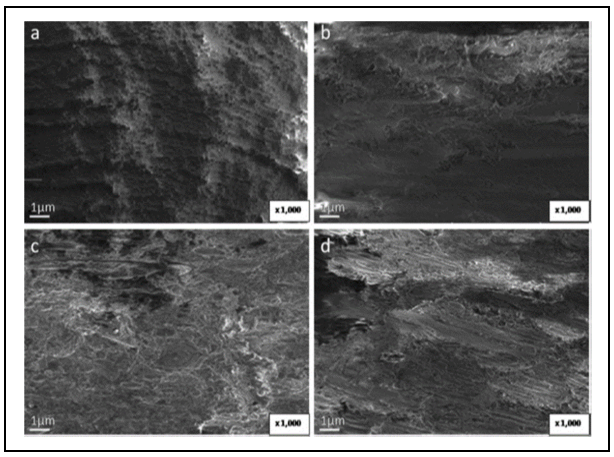


Fig. 5: Scanning electron micrographs of a tensile fractured surface of a Virgin HDPE; b HDPE/0.025%; c HDPE/0.050; and d HDPE/0.075% nanocomposites

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. The 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.

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CONFLICTS OF INTEREST

The authors declare that there is no conflict of interest.

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