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Mechanical Properties and Morphology of Graphene Oxide Reinforced EPDM/HDPE Composites

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The mechanical properties of Graphene Oxide reinforced Ethylene Propylene Diene Terpolymer rubber (EPDM) / High Density Poly Ethylene (HDPE) composites were studied. Blends of Ethylene Propylene Diene Terpolymer rubber (EPDM) with High Density Poly Ethylene (HDPE) were obtained by solution intercalation. Composites of EPDM/HDPE blends with Graphene Oxide at varying concentrations (1,3,5,7,10phr) were prepared to investigate the effects of Graphene Oxide on the mechanical behavior of EPDM/HDPE blends. The structures of these blends were characterized using Fourier-transform infrared spectroscopy (FTIR). The mechanical properties of the blend, such as tensile strength, tensile strain at break, percentage of elongation, and modulus, were determined. The EPDM/HDPE/GO composites exhibited good mechanical properties. The EPDM/HDPE/GO composite containing 7 phr Graphene Oxide showed a high tensile strength of 15.23 MPa. The EPDM/HDPE/GO composites containing a small amount of Graphene Oxide exhibited better mechanical properties than their corresponding blends without Graphene Oxide. The EPDM/HDPE/GO composite containing 7 phr graphene oxide showed the highest tensile strength and elongation at break.

Keywords: EPDM/HDPE, Graphene oxide, mechanical properties, nanocomposite.

Introduction

Polymer nanocomposites based on graphene oxide were known as progressive technologies which were prepared by using nano graphene [1,2]. This nanoplatelets has extensive applications in electronic devices such as sensors, actuators and other devices [3,4]. Graphene nanoplatelets have multi-layer structures which conducted to particular behaviors. Beside suitable properties, these nanoplatelets have high tendency to aggregation due to having strong Van der Waals interactions between layers, therefore dispersion of nanoplatelets into polymer matrix is very difficult [5,6]. In the literature, many researches have been carried out about dispersion of nanographene oxide platelets in polymeric matrix.

Ethylene propylene diene monomer (EPDM) is an elastomer which is characterized by a wide range of applications. High density polyethylene (HDPE) is a commodity thermoplastic

polymer and is widely used in different applications due to its outstanding features such as regular chain structure, combination of low cost and low energy demand for processing, excellent bio compatibility, and good mechanical properties [7,8]. Properties such as mechanical, thermal, processing etc of HDPE can be further improved by the addition of organic or inorganic particles into the polymer matrix [7-11].

The well-known method for modifying the properties of HDPE and EPDM is blending technique; it is economically viable and versatile way to produce new materials and to overcome the deficiency in some material characteristics [12]. However, most of polymer blends form heterogeneous systems that affect their physico-optical properties [13].

Recently polymer blending is considered one of the most economical techniques that plays a special role to improve the physical and mechanical properties of the new finished product [12,14]. As a result of blending, the physical properties can be altered to produce useful materials with a wide range of applications and possess a combination of physical and rheological properties that are unavailable in a single polymer [14-16]. This type of blending is considered as a convenient route for obtaining materials with improved properties and low-cost performance [17-19].

In this study, the presence of GO on properties of elastomeric blend nanocomposites based on EPDM/HDPE are investigated. As regards, there is very little information about EPDM/HDPE/GO nanocomposites, the role of GO on physical characteristics of EPDM/HDPE blend is investigated.

Experimental Procedure

Materials

The EPDM rubber, a product of DOW, was obtained in the form of pellets. HDPE was sourced from ONGC Petro Additions Limited (O-Pal), India. Graphene oxide (GO) was obtained from Adnano Technologies, Karnataka, India, with a purity of 99%, particle size less than 10 μm, and thickness ranging from 0.8 to 2 nm. The cross-linking agent, 98% pure dicumyl peroxide crystals, was purchased from Sigma-Aldrich, manufactured by Aldrich, Japan. Analytical reagent (AR) grade toluene with 99.5% purity was obtained from NICE Chemicals, Cochin.

Preparation of EPDM/HDPE/GO composites

In the present study, investigations were conducted on various composite systems prepared with different quantities of Graphene Oxide in EPDM/HDPE blend. EPDM rubber (100 parts) and HDPE (10phr) were dissolved in toluene, and varying amounts of ultrasonicated Graphene Oxide (1,3,5,7 and 10phr) were added to prepare composite via the solution intercalation process using a small-scale internal mixer to attain hot melt. The blending process was carried out with a rotor speed of 300 rpm at an operating temperature of 60^{o}C for 5 hours. To influence the curing behavior of EPDM/HDPE/GO nanocomposites, a calculated amount of di cumyl peroxide (3phr) was mixed in the prepared nanocomposites before sheeting and the composition was immediately transferred into glass moulds. The films were then cooled to room temperature. The moulded rubber sheets were left for at least 24 hours to obtain sheets of suitable thickness and then separated from the mould. Finally, cured nanocomposite sheets were cut into standard shapes to carrying out mechanical and other tests for their characterization. The composition of EPDM, HDPE and GO for preparing EPDM/HDPE/GO nanocomposites in this study is summarized in Table 1.

Table 1: Formulations of GO, EPDM/GO and EPDM/HDPE/GO nanocomposites.

Sample codes	Components of EPDM/HDPE-GO nanocomposites (phr)		
	EPDM	HDPE	GO
E	100	0	0
E10H	100	10	0
E10H1GO	100	10	1
E10H3GO	100	10	3
E10H5GO	100	10	5
E10H7GO	100	10	7
E10H10GO	100	10	10

phr (parts per hundred parts of rubber)

Characterization

Infrared spectra were recorded using a SHIMADZU IR Tracer-100 FTIR Spectrophotometer equipped with a Total Attenuated Reflectance (ATR) accessory in a humidity-free atmosphere over the range of 400-4500 cm^{-1}. Mechanical properties were measured using an Instron UTM 3345 instrument, and the test method used was adopted from ASTM D882. All samples were prepared as thin films of 0.3mm thickness.

Results and discussion

FTIR analysis:

The FTIR spectrum of EPDM/HDPE/ Graphene Oxide composite (Fig. 1) shows expected absorption bands corresponding to the EPDM backbone and additional absorption bands.

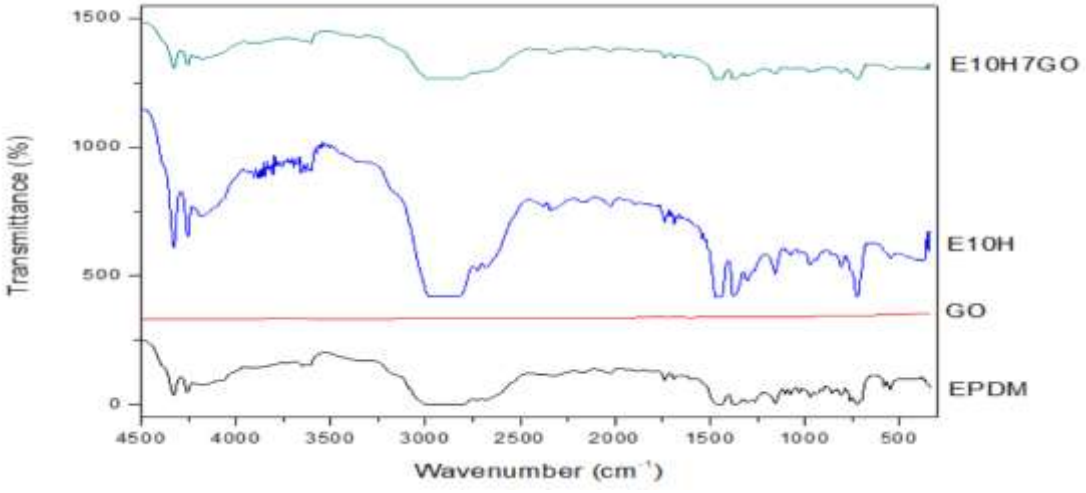


Fig.1 IR spectra of rubber, nanoparticle and nanocomposites

The five characteristic absorption bands of EPDM rubber appeared at 722 cm^{-1} corresponding–(CH_2)– stretching and at 1374 cm^{-1} due to–C–CH_3 stretching. The peak at 2924 cm^{-1} is assigned to asymmetric stretching vibration of methylene in the saturated hydrocarbon backbone, while the peak at 2853 cm^{-1} is assigned to symmetric stretching vibration of methylene [20]. The peaks at 1464 cm^{-1} and 1377 cm^{-1} are assigned to -CH_2- scissoring vibration and symmetric CH stretching vibration of methyl, respectively [21, 22]. One can see the CH antisymmetric and symmetric stretching bands in the region of 3000–2800 cm^{-1}, the asymmetric bending vibration over 1550–1400 cm^{-1}, and CH_2 rocking vibration bands at 750–650 cm^{-1}[23].

FTIR spectrum of the EPDM/HDPE blend containing E10H shows two new absorption peaks appear at 902 cm^{-1} and 933 cm^{-1}, which are likely attributed to the bending vibrations of C–H bonds in alkanes, particularly the CH_2–CH_2 bending mode. A relative increase in the intensity of the band near 1373 cm^{-1} is observed for the EPDM/HDPE blend [24]. Additionally, minor shifts in the bands at 1604 cm^{-1}, 1867 cm^{-1}, and 2322 cm^{-1} may be attributed to the incorporation of HDPE into the EPDM matrix [25].

In the FT-IR spectra of graphene oxide (GO), a broad band around 3400 cm^{-1} is assigned to O–H stretching (from hydroxyls and adsorbed water) on the GO surface. Strong peaks near 1620–1650 cm^{-1} correspond to skeletal C=C vibrations of graphitic domains. Additional absorptions in the 1200–1050 cm^{-1} range (C–O alkoxy groups) further confirm the high oxygen-content of GO [26]. In the EPDM/HDPE/GO nanocomposite (E10H1GO), all polymer bands are retained, confirming that the base hydrocarbon structure is largely unaltered. The intensities of these GO-specific bands are reduced and somewhat broadened compared to neat GO, consistent with GO being well-dispersed in the polymer matrix. The slight broadening of the O–H band and possible downshift of the C=O band suggest hydrogen bonding between GO's oxygen groups and polymer chains (e.g. CH groups) or moisture [27]. Importantly, no new IR peaks appear in E10H1GO, indicating that no covalent bonding (such as chemical grafting of GO to EPDM/HDPE) has occurred during solution mixing. These IR results indicate good dispersion of GO by physical interaction.

Mechanical properties:

Tensile testing machine measured the mechanical properties of the sample. The mechanical properties showed significant improvement. The mechanical testing was performed on the EPDM/HDPE/Graphene Oxide composite under different concentration of Graphene Oxide and variation in tensile strength is given in the below table:

Table 2: Tensile Properties of EPDM/HDPE/GO Composites

EPDM/HDPE/ Graphene Oxide composite (phr)	Tensile Strength [MPa]	Tensile strain at break [%]	Modulus (Automatic) [MPa]	Percentage of elongation (%)
E	4.33	7.50	0.87	856
E10H	11.512	11.184	3.286	1344
E10H1GO	12.84	11.52	3.56	1282
E10H3GO	13.95	11.3	4.14	1234

E10H5GO	14.77	10.97	4.82	1095
E10H7GO	**15.23**	**11.46**	**5.38**	**1230**
E10H10GO	13.50	10.62	4.72	1056

The above table illustrates the mechanical properties of the EPDM/HDPE/Graphene Oxide composite films. The tensile strength of the blends increases as the Graphene Oxide content increases. The tensile strength of E10H7GO is high when compared to other formulations i.e., 15.23 MPa. Tensile Modulus shows a maximum value for E10H7GO. The best elongation is seen in the E10H1GO sample. This improvement can be attributed to the stable filler network achieved using GO with HDPE in the rubber matrix and the strong interaction between GO with the rubber. For the E10H1GO sample the highest percentage of elongation obtained i.e., 1282%. However, incorporating Graphene Oxide into a composite formulation generally enhances its tensile strength compared to formulations without Graphene Oxide.

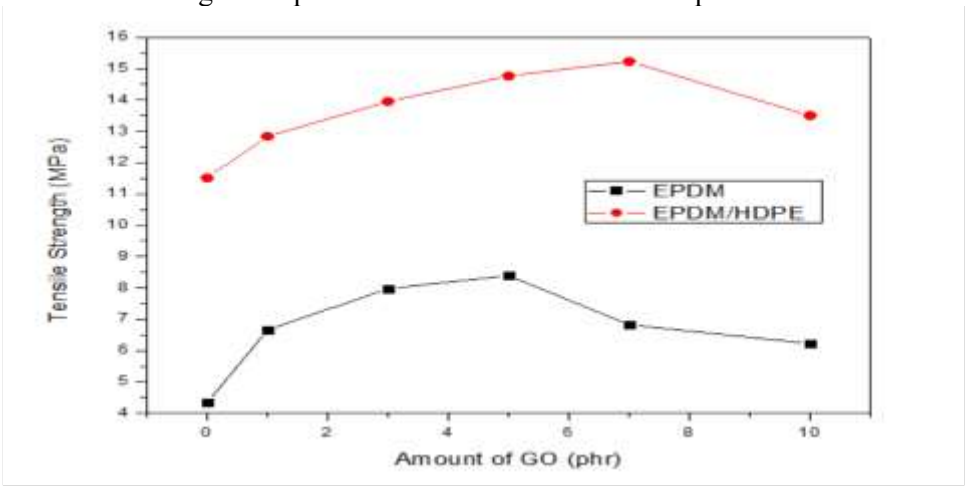


Figure 2: Tensile strength of GO-filled EPDM and EPDM/HDPE Blends

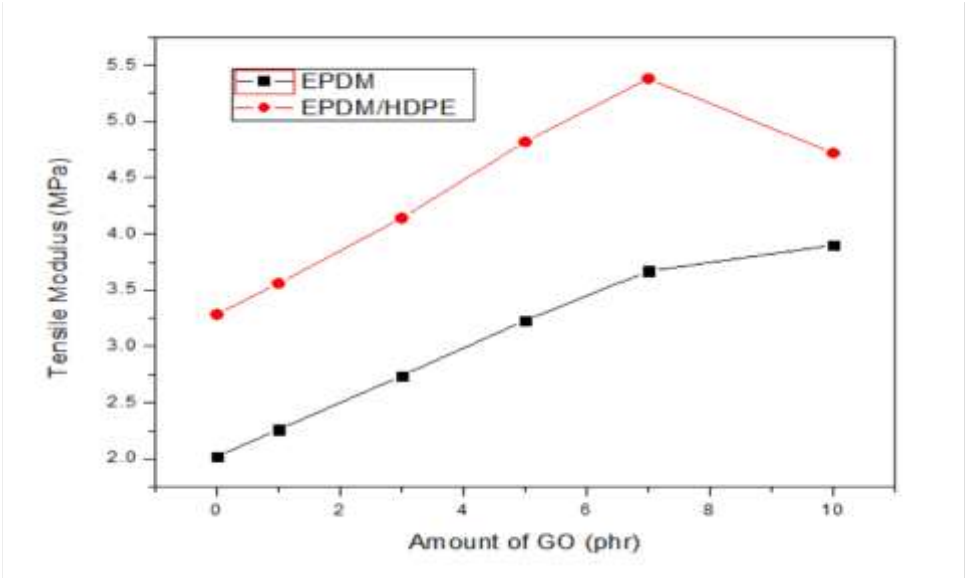


Figure 3: Tensile Modulus of GO-filled EPDM and EPDM/HDPE Blends

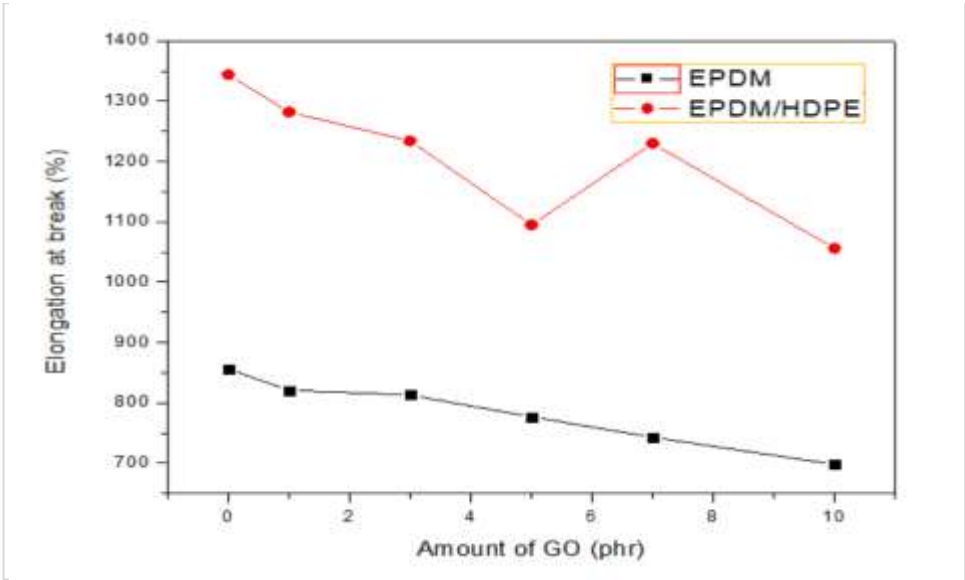


Figure 4: Elongation at break of GO-filled EPDM and EPDM/HDPE Blends

Adding Graphene Oxide to EPDM/HDPE blends results in composites with improved mechanical properties, including increased tensile strength, stiffness, and stress crack resistance because Graphene Oxide effectively transfers the applied loads to the polymer matrix. The tensile strength of EPDM/HDPE/GO nanocomposites decreases at higher graphene oxide loadings mainly due to agglomeration of GO nanosheets, poor dispersion, increased stress concentration, and inefficient stress transfer between the polymer matrix and the filler [27,28].

Morphological observation:

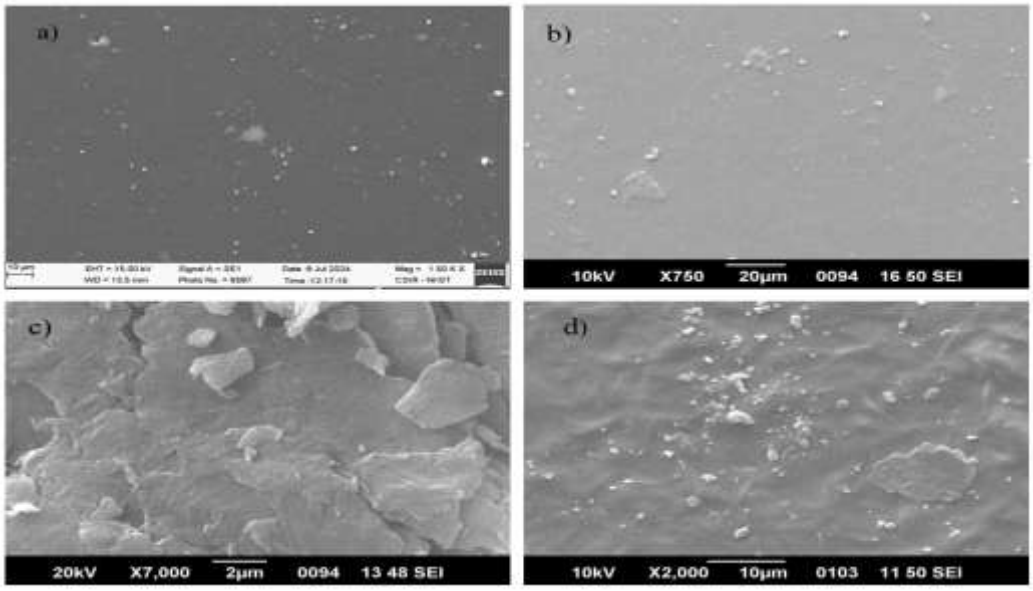


Figure. 5: SEM micrographs of (a) neat EPDM, (b) E10H Blend, (c) GO Nanofiller, (d) E10H7GO Nanocomposite.

The SEM image of the neat EPDM sample (Fig. 5a) reveals a relatively smooth fracture surface, indicating a brittle fracture mode**.** This corresponds with its low tensile strength (4.33 MPa) and limited strain at break (7.5%), as observed in mechanical testing. In contrast, the SEM micrographs of 10 phr EPDM/HDPE blends (Fig. 5b) exhibit significantly rougher fracture surfaces, indicative of tough-type fracture behavior. The rougher surface morphology in these blends is attributed to enhanced interfacial adhesion between the EPDM and HDPE phases, which promotes better stress transfer and energy dissipation during deformation. This behavior also reflects the inherently higher toughness of HDPE**,** contributing to the overall mechanical reinforcement [29].

The SEM micrograph of pure graphene oxide (Fig. 5c) reveals irregular layered nanosheets with a wrinkled and folded morphology. The rough surface and crumpled appearance confirm the successful oxidation and exfoliation of graphite into graphene oxide [30]. The large surface area and abundant oxygen-containing functional groups of GO are expected to improve interfacial adhesion with EPDM/HDPE matrices, thereby enhancing the mechanical and thermal performance of the prepared nanocomposites. In EPDM/HDPE/GO nanocomposites (Fig. 5d) the relatively smooth fracture morphology and the absence of significant interfacial gaps indicate satisfactory compatibility between the polymer phases and good interfacial adhesion between graphene oxide and the polymer matrix [31]. Although a few localized GO agglomerates are visible, most of the filler is distributed uniformly, suggesting effective dispersion during processing.

Conclusions

This research highlights the incorporation of Graphene Oxide into EPDM/HDPE blend significantly enhances the mechanical properties of the EPDM/HDPE/Graphene Oxide

composites. Specifically, the addition of 7 phr Graphene Oxide yields a high-quality film with improved modulus, tensile strength, and tensile strain at break. Mechanical property measurements reveal that the HDPE/EPDM/Graphene Oxide composite exhibits enhanced tensile strength, modulus, and elongation at break compared to EPDM/HDPE blend. The Graphene Oxide reinforcement plays a crucial role in improving these properties, demonstrating an excellent synergistic effect. These findings indicate that EPDM/HDPE/Graphene Oxide composite possess greatly improved mechanical properties, making them suitable for various applications, including the automotive industry, energy management and other sectors.

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