Biointerface Research in Applied Chemistry
Platinum Open Access Journal (ISSN: 2069-5837)

Article
Volume 12, Issue 4, 2022, 5709 - 5715
https://doi.org/10.33263/BRIAC124.57095715

Rheological Properties of HDPE based Thermoplastic Polymeric Nanocomposite Reinforced with Multidimensional Carbon-based Nanofillers

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Received: 30.08.2021; Revised: 10.10.2021; Accepted: 14.10.2021; Published: 21.10.2021

Abstract: The present investigation focused on the evaluation of rheological properties HDPE reinforced with equal weight percentage (i.e., 0.1 wt. %) of Nano-diamond (0D), Carbon nanotubes (1D), and Graphite Nano-platelets (2D) multidimensional nanofillers. The results like storage modulus, loss modulus, Tan delta, and complex viscosity results expounded from the rheological test with a frequency sweep from 10^{-1} to 10^{2} rad/s. The highest storage modulus was perceived by 0.1 CNT-based composites, i.e., 18408 Pa, which decreased to 19, 52, and 85 % for 0.1 GNP, 0.1 ND, and pure, respectively. A similar trend was observed for loss modulus and damping factor results. The shear-thinning behavior observed in viscosity results and the addition of ND nanofillers improve the viscosity to a large amount. The potential applications of the composites include polymer gears, landing mats, cams, and various functional elements.

Keywords: polymer matrix composite; rheological properties; ND; CNT; GNP.

1. Introduction

Thermoplastic polymeric composites are a class of lightweight, low-cost, easily formable polymeric composite materials, making them ideal for a wide range of applications, including automobile, aerospace, nautical, and sports industries [1, 2]. High-density polyethylene (HDPE) is a class of engineering materials that comes under thermoplastic polymer is the key choice due to its high mechanical strength and superior thermal properties [3, 4]. However, it has drawbacks like poor rheological properties [5, 6]. The addition of a suitable nano-filler to the matrix to form an HDPE polymeric nanocomposite will improve the above lacuna to a great extent [7, 8]. Following are some important pieces of literature discussed in line with the topic espoused.

Falaki *et al*. [9] reported on the rheological properties of HDPE and PA-6 microfibrillar reinforced composites (MFCs). The assessment shows that the addition of microfibrils enhanced the storage modulus and complex viscosity to a great amount. Azizi *et al*. [10] investigated the rheological properties of low-density polyethylene (LDPE)/ethylene vinyl acetate (EVA) reinforced with variable wt. % of graphene (i.e., 5 to 20 wt. %). The results exhibited a stiff rise in storage modulus and viscosity value at 20 wt. % of graphene content in

HDPE/EVA composite. The effect of multi-walled carbon nanotube (MWCNT) on the rheological properties of polypropylene was carried out by Yetgin [11]. In this study, the MWCNT content was varied from 0 to 2 wt. % and observed that storage modulus and loss modulus significantly increased with an increase in MWCNT content due to the formation of an interconnected network-like structure, which restricted the mobility of the polymer matrix chain. Biswas *et al*. [12] studied rheological properties HDPE composite reinforced with teak wood flour (10-40 wt. %) and Maize Starch (MS) (5-20 wt. %). The complex viscosity results showed a decrease in trend with an increase in frequency representing shear thinning behavior. However, the storage and loss modulus was increased with frequency. Chen *et al*. [13] investigated the rheological properties of polypropylene (PP)-reduced graphene oxide (RGO) nanocomposite. It was observed that the addition of RGO in PP matrix enhanced viscosity results by about 77% compared to neat PP. The reason for this was due to stronger interaction among PP/RGO matrix by means of the reduction extent upsurges, which in turn restricts the movements of the PP chain. Rheological properties of HDPE matrix filled with 1 to 20 wt. % of aluminium powder carried out by Mysiukiewicz *et al*. [14]. The results suggested that the addition of 20 wt. % of Al powder increases the complex viscosity due to well-dispersed rigid particles in the HDPE matrix, and all the samples showed shear thinning behavior. Al-Baghdadi *et al*. [15] reported the rheological properties of polyethylene/nanotube (PE-NT) composite. The composite was prepared using an injection molding technique with 0 to 3 wt. % of NT ratio in the composite sample. Escocio *et al*. [16] investigated the rheological properties of HDPE composite filled with 10 to 40 wt. % of spouge gourd (SG). It was observed that the addition of 40 wt. % SG content showed an increase in viscosity as the higher filler content act as a barrier to the flow of the polymeric chain. Orji *et al*. [17] evaluated rheological properties of recycled high-density polyethylene (rHDPE) composite reinforced with rice hull particle size varied from 0.5 to 1 mm. It was noted shear thinning behavior for all the samples, and among all the samples, the highest viscosity was observed for rHDPE/<0.5 mm rice hull. The effect of soapstone waste on the rheological properties of HDPE was demonstrated by de Sousa et. [18]. In this study, the composites were fabricated by varying the concentration of the filler from 10 to 30 wt.%, and it was noted that more pseudoplastic behavior was pronounced when 30 wt.% filler was added to the composite.

The literature survey expounded that the addition of nanofillers into the HDPE polymer matrix extraordinarily enhances the rheological properties. The present investigation focused on rheological properties HDPE reinforced with equal weight percentage (i.e., 0.1 wt. %) of Carbon nanotubes (CNT), Nano-diamond, and Graphite Nano-platelets (GNP) nanofillers. The ND, CNT, and GNP are 0, 1, 2-dimensional nanostructures, respectively, and the authors are keen to investigate the individual interaction of multidimensional nano-filler into HDPE matrix. As none of the researchers studied the rheological properties of HDPE matrix reinforced with distinct dimension nanofillers at a time, the present investigation is novel. The choice of ND is due to excellent hardness [19, 20], CNT is due to exceptional modulus and tensile strength [21, 22], and GNP is due to enhanced surface energy and lubricity [23, 24]. The authors are profound to examine the properties of carbon-based multidimensional nanofillers on rheological properties of HDPE-based composites.

2. Materials and Methods

2.1 Materials.

HDPE pellets were procured from IOCL (Indian Oil Corporation Limited), India. The detailed specification HDPE can be referred to Sahu *et al*. [25]. The surface modification of the fillers, i.e., ND, CNT, and GNP, and preparation of composite can be referred to by Badgayan *et al*. [26] and Sahu *et al*. [27]. The morphology of ND, CNT, and GNP after surface modification is represented in Figure 1a-c using a Transmission electron microscope (TEM). The specimens are obtained as per ASTM D 3364–94, and the sample notation is illustrated in Table 1.

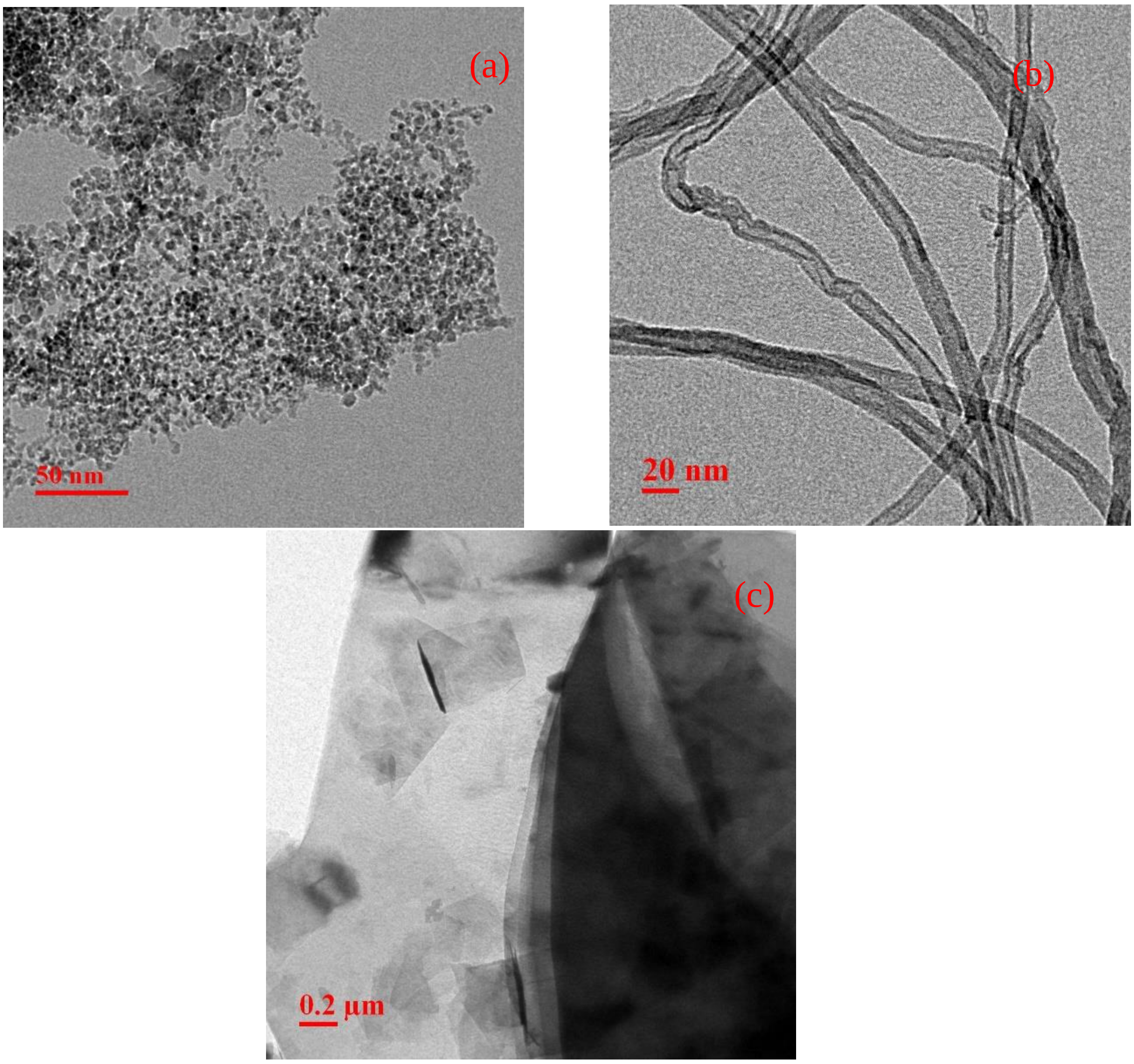

Figure 1. TEM morphology of **a**) ND; **b**) CNT; **c**) GNP.

Table 1. Notation of samples.

Sl. No.	Sample	HDPE (Wt. %)	ND (Wt. %)	GNP (Wt. %)	CNT (Wt. %)
1	Pure	100	-	-	-
2	NC1	99.9	0.1	-	-
3	NC2	99.9	-	0.1	-
4	NC3	99.9	-	-	0.1

2.2. Rheological properties.

The rheological properties of pure HDPE and its composite were carried out with a Rheostress RS 1 (Thermo Electron, Germany). The test was carried out by frequency sweep mode from 10^{-1} to 10^{2} rad/s at a fixed temperature of 90°C using a parallel plate of 40 mm arrangement. The storage modulus, loss modulus, tan delta, and complex viscosity results were evaluated from the test.

3. Results and Discussion

The viscoelastic behavior of the virgin polymer and its composite is measured using rheological testing. Figure 2a shows the storage modulus vs. frequency for all the samples. The storage modulus results show an increase in trend with an increase in frequency for the entire sample, indicating elastic behavior dominance. However, more pronounced results were seen for the NC3 composite. At 10^{-1} rad/s, the storage modulus is 5 Pa, which decreased by 2, 38, and 62 % for NC2, NC1, and pure, respectively. Similarly, at 10^{2} rad/s, the storage modulus for NC3 composite is 18408 Pa, which decreased by 19, 52, and 85 % for NC2, NC1, and pure, respectively.

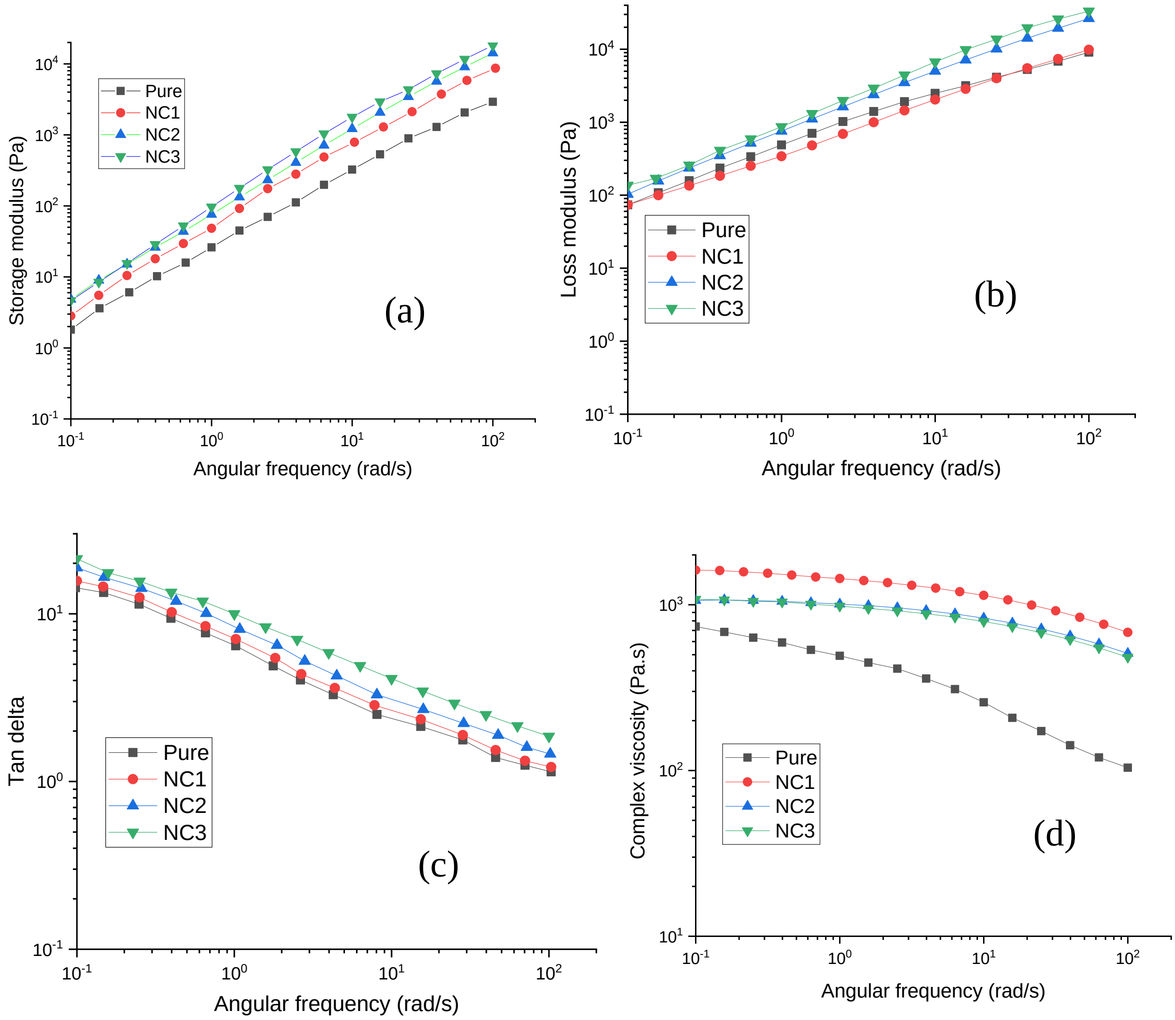

Figure 2. a) Storage modulus vs. frequency; **b**) Loss modulus vs. frequency; **c**) Tan delta vs. frequency; **d**) Complex viscosity vs. frequency.

The reason for higher storage modulus at low and higher frequency section owing to two distinct reasons; firstly the good interfacial adhesion between CNT and HDPE matrix [25], secondly, tubular nature of CNT, which deforms temporarily when the load is applied and recovers when the load is removed, thereby acclaiming higher storage modulus.

Similarly, Figure 2b shows the loss modulus vs. frequency for the entire sample. It is observed that the loss modulus also shows a similar trend, i.e., increase in trend with increase in frequency. The highest loss modulus was shown for NC3 composite, i.e., 143 Pa at 10^{-1} rad/s frequency, which increased to 33112 Pa at 10^{2} rad/s. The possible reason for the higher loss modulus for NC3 composite is the formation of mesophase between particle to particle and particle to polymer interaction, causing a large loss modulus for CNT composite [26]. Tan delta denotes the ratio of loss to storage modulus as a useful parameter to measure damping. Figure 2c shows the tan delta results for all the tested samples, which measures the damping ability. The highest tan delta is depicted by NC3 composite, i.e., 21.7 at 10^{-1} rad/s, which decreased by 14.4, 26.9, and 33.6 % for NC1, NC2, and pure, respectively. A similar trend is observed at a higher frequency as well. The reason for lower damping for NC3 composite can be understood from the higher value of loss modulus results compared to storage modulus. Figure 2d shows the complex viscosity as a function of angular viscosity, which measures the overall resistance to flow. It has shown shear thinning behavior with frequency for all the samples.

The highest complex viscosity is shown by NC1, i.e., 1631 Pa.s at 10^{-1} rad/s followed by NC2, NC3, and pure, i.e., 32, 34, 55 %, respectively. At 10^{2} rad/s, the highest resistance was shown by NC3, i.e., 682 Pa.s, which decreased up to 25, 29, and 85% for NC2, NC1, and pure, respectively. The rationale for the highest viscosity offered by NC1 composite is due to the nanodiamond structure and its strong van der Walls forces, and consequently, it possesses high viscosity [27].

4. Conclusions

The objective of the present investigation is to evaluate the influence of multidimensional carbon-based nanofillers, i.e., 0D (ND), 1D (CNT), and 2D (GNP), on rheological properties of HDPE based nanocomposite. The loading of nanofillers is maintained at 1 wt. % for all the nanocomposite. It was observed that the addition of CNT improvised the storage and loss modulus due to the tubular nature of CNT. The complex viscosity results depicted that the addition of ND nanofillers improves the viscosity to a large amount. The potential applications of the composites include polymer gears, landing mats, cams, and various functional elements.

Funding

None.

Acknowledgments

The authors are thankful to Prof. S. Kanagaraj, IIT Guwahati, for the facilities provided in Materials Lab.

Conflicts of Interest

None.

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