



## Elastomer Toughened Poly (butylene terephthalate) Nanocomposites: Morphology and Impact Strength

Ranjana Sharma, Himani Joshi, Purnima Jain\*

School of Applied Sciences, Netaji Subhas Institute of Technology, University of Delhi, Dwarka, New Delhi

### ABSTRACT

Elastomer toughened nanocomposites consisting of Poly(butylene terephthalate) (PBT), glycidyl methacrylate grafted ultra low density polyethylene (ULDPE-g-GMA), and organoclay (Cloisite 30B) were prepared by melt blending using microcompounder followed by injection molding. ULDPE-g-GMA was used as an impact modifier. The content of ULDPE-g-GMA was kept constant at 2wt% while OMMT at 3wt% and 5wt% has been used. The obtained nanocomposites were analyzed by XRD, polarizing optical microscopy (POM) and izod impact test. We have investigated the effect of organoclay inclusion on the impact strength and morphology of PBT molecules in the nanocomposites.

**Key Words:** Nanocomposites; PBT; XRD; POM; Izod impact test

### INTRODUCTION

Polymer nanocomposite is a new class of composite materials derived from nanoparticles and it is a two phase material where one of the phases is in the nanometer range in at least one dimension being less than 100 nm [1-5]. Nanocomposites based on organic polymers and inorganic clay minerals consisting of layered aluminosilicate have received considerable attention in recent years. They are categorized as polymer/clay nanocomposites. It is a novel technique to develop reinforced polymers. The dispersion of ultra-thin (1nm) ultra high surface area clay layers within a polymer matrix at low loading show extraordinary improved physiochemical properties such as higher strength and modulus, better dimensional stability and thermal stabilities, higher heat distortion temperature, chemical stability, gas barrier properties and flame retardancy, compared with virgin polymers [6-10]. In general, the improvements in the properties may be due to the following factors: (a) high aspect ratio and large surface area; (b) dispersion of the clay in polymer matrix; (c) ionic bond between organic polymer and inorganic clay [11].

Polymer-clay nanocomposites are mostly synthesized by using three methods: solution intercalation, in situ polymerization intercalation and melt intercalation [12-14]. Among them melt intercalation is the most appealing and convenient method because of its versatility, its compatibility with current polymer processing techniques and its environmentally benign character [15,16]. Depending on the degree of polymer penetration into silicate layers, two idealized polymer-clay structures are possible: exfoliated and intercalated [17]. Until now many polymer-clay nanocomposites have been synthesized through incorporating clay in various polymer matrices such as polystyrene, polyamide, PET, polyurethane, polyimide, polypropylene, epoxy resin, silicone rubber, PBT, poly(ethylene oxide) and so on [8,12,15-18].

Nanocomposites based on semicrystalline polymer and layered silicate have been actively investigated by several researchers. Semicrystalline polybutylene terephthalate (PBT) is the most representative and commercially available engineering thermoplastic with many valuable properties, including good abrasion, chemical resistance, thermal stability, high rate of crystallization and excellent processing properties. PBT is widely used in applications such as insulators for electrical engineering, electronics solution and as connectors in automobiles. However, pure PBT has

poor mechanical and heat distortion temperature. So, to overcome the drawbacks of pure PBT, impact modifier with clay has been used to improve properties significantly [17, 19-22].

Li et al [15] prepared PBT/clay nanocomposites via melt intercalation via montmorillonite (MMT), and studied the intercalation and exfoliation behavior of PBT nanocomposites. A further study on the effect of blending sequence on the properties and microstructure of PBT/EVA-g-MAH/Clay ternary nanocomposites was again conducted by Li [17]. Xiao et al [10] reported the preparation and characterization of PBT/clay nanocomposites from thermally stable organically modified MMT and concluded that the melting temperature, crystallization rate and crystallinity of PBT were improved by dispersion of organoclay. Acierno et al [23] studied the effect of different organoclays on nanocomposites, the relationship between processing conditions, nanoscale morphology, and properties of PBT nanocomposites, with the aim of evaluating their possible application in automotive and aerospace fields.

Present work is devoted to study the effect of OMMT (cloisite 30B) on the impact strength and morphology of elastomer toughened PBT nanocomposites with clay loadings of 3 and 5wt %. Characterization of samples was carried out by using HRXRD and polarized optical microscopy equipped with hot stage and izod impact test.

## MATERIALS AND METHODS

### 2.1. Materials:

PBT (T06 200) was obtained from DSM Engineering Plastics (Pune, India). Organoclay was supplied by Southern Clay Product Inc. under the trade name Cloisite 30B, modified with methy, tallow, bis-2-hydroxy ethyl ammonium. The impact modifier (IM) used was ULDPE-g-GMA (trade name GE-344) supplied by Pluss Polymers, India, which contain 2% grafted GMA.

### 2.2. Sample Preparation:

The PBT pellets and OMMT were dried under vacuum oven at 80 °C for 10 hr before use where impact modifier (IM) was used as received. Melt compounding of PBT/IM/OMMT was done in a microcompounder Xplore 15ml, DSM (Netherlands) with organoclay loadings of 3 and 5 wt% with base polymer at 240 °C. The screw speed was set at 50 rpm. As a base of comparison, the neat PBT was also passed through micro compounder at the same conditions. The samples are identified in Table 1.

Table 1. Sample identification and composition

Sample	Composition (wt %)		
	PBT	ULDPE-g-GMA (IM)	OMMT
A	100	0	0
B	98	2	0
C1	95	2	3
C2	93	2	5

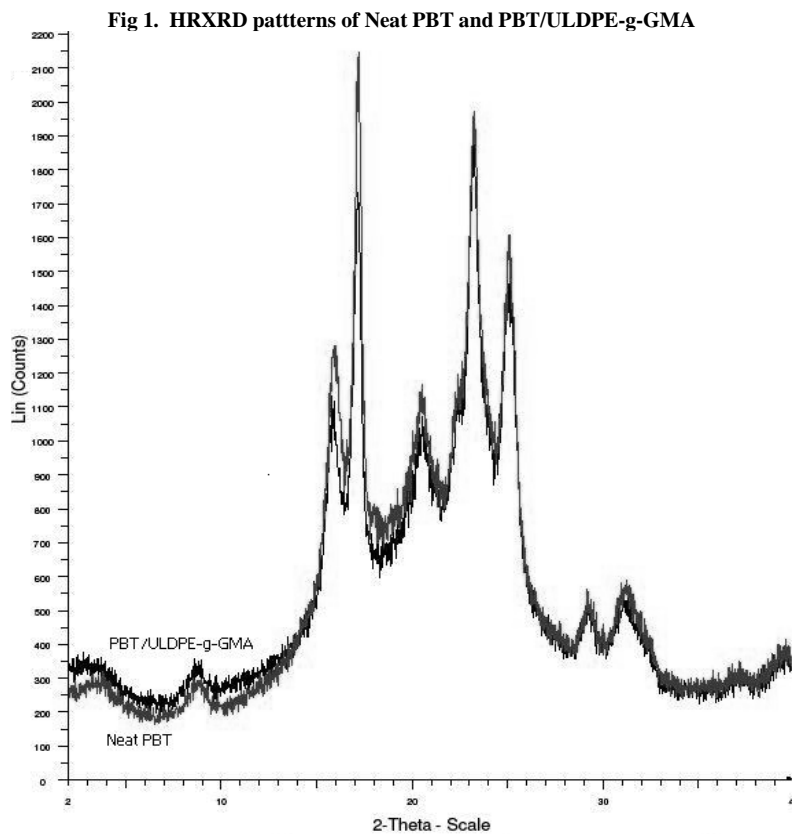
### 2.3. Characterization:

The dispersibility of the silicate layers (OMMT) in the PBT was evaluated using HRXRD (High Resolution X-ray Diffractometry). The experiments were performed at room temperature on D8 Discover, Bruker X-ray Diffractometer (40 kV at a current of 15 mA) with Cu K $\alpha$  ( $\lambda = 1.5406 \text{ \AA}$ ) irradiation at the rate of 5°/min in the range of 2 – 40°. A polarizing optical microscope was used to observe spherulite formation in neat PBT and elastomer toughened PBT/clay nanocomposites. A polarizing optical microscope (POM) observation was performed using Zeiss Axioscope microscope using 10 times magnification. POM experiments were carried out by heating sample (pellets) to 250 °C and holding them for 3 min for complete melting. The crystallization was observed during cooling process. Nanocomposites melt was cooled to 200 °C and kept at this temperature for 30 min. Notched izod impact tests were performed at room temperature with a Tinius Olsen, USA impact tester according to ASTM D256 and taken as measurement of the impact toughness. The results reported here are average of three successive tests.

## RESULTS AND DISCUSSION

### 3.1. HRXRD characterization:

XRD is most commonly used to elucidate the structure of nanocomposites whether it is intercalated or exfoliated. It allows the precise measurement of silicate layer spacing and intercalation behavior [24-25]. The dispersion of organoclay in the polymer matrix was observed by high resolution X-ray diffraction (HRXRD). For pure PBT, the  $\alpha$  form structure is the most extensively studied structure and characterized by eight distinct crystalline peaks. Figure 1 shows the HRXRD curves of neat PBT and elastomer toughened PBT. Both of them exhibit eight distinguishable peaks. It indicates that the presence of ULDPE-g-GMA does not affect crystalline structure of PBT.



**Fig 2. HRXRD patterns of Cloisite 30B (a) and PBT/ULDPE-g-GMA/OMMT nanocomposites with 3wt% (b) and 5wt% (c) OMMT**

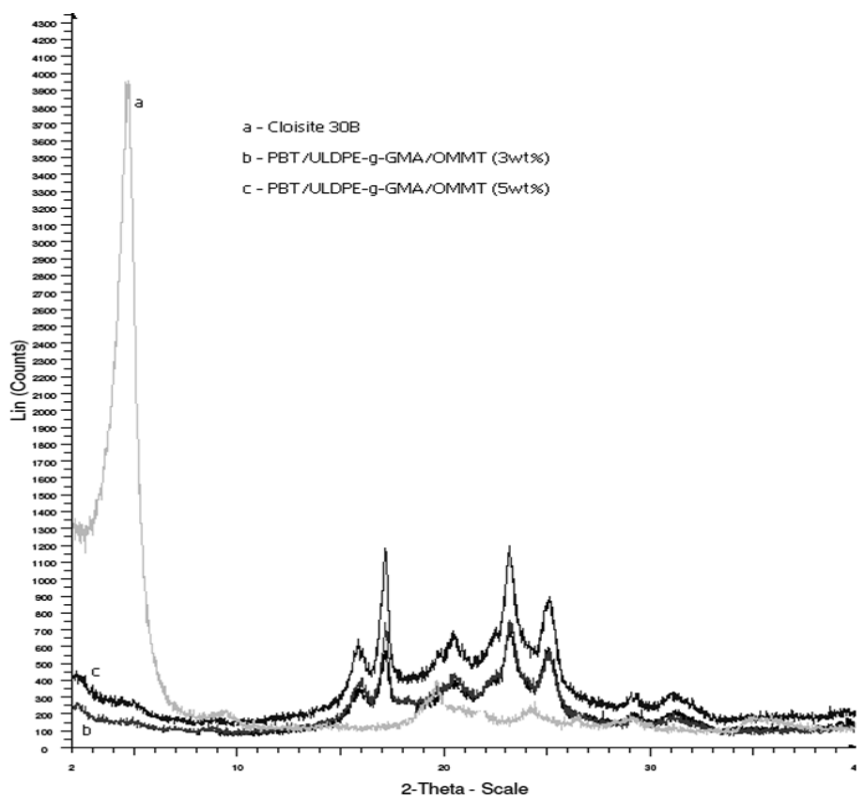
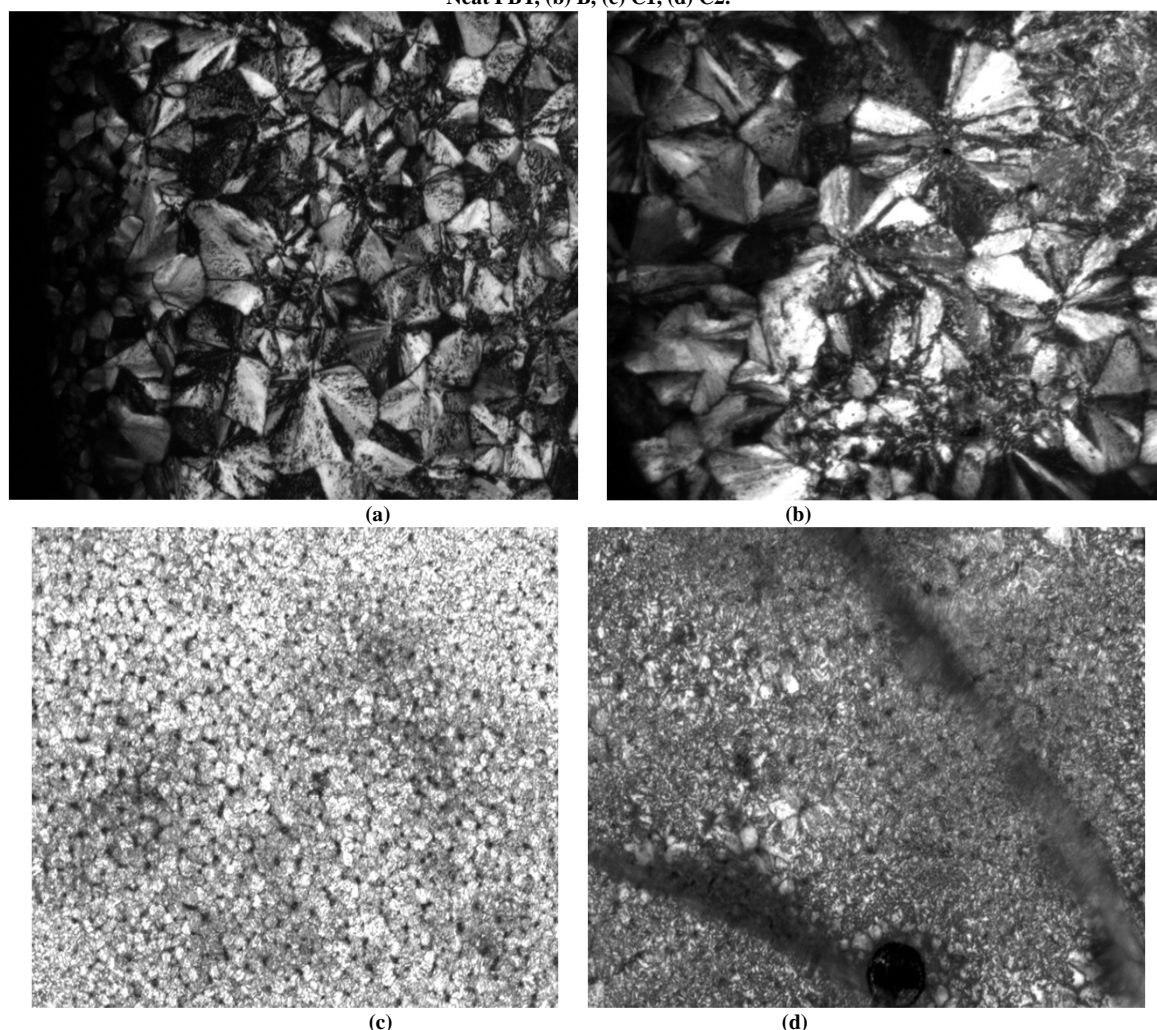


Fig. 3. POM pictures showing spherulite growth. All micrographs were taken under reflection mode with 10 times magnification where (a) Neat PBT, (b) B, (c) C1, (d) C2.



HRXRD patterns PBT/ULDPE-g-GMA/OMMT ternary nanocomposites as well Cloisite 30B are shown in figure 2. The modified clay exhibits a single sharp peak at  $2\theta = 4.73$  ( $18.9\text{\AA}$  basal space) in the range of  $1^\circ$ -  $10^\circ$ . Ternary nanocomposites with 3wt% and 5wt% of OMMT (fig.2, b,c) exhibits weak peak at lower  $2\theta$  angle as compared to Cloisite 30B, implying that the organoclay is partially exfoliated in the PBT matrix. A shift in the first clay peak indicates the formation of an intercalated structure, whereas disappearance of the peak or reduction of its intensity indicates an exfoliated structure [26]. Some researchers also observed featureless XRD patterns even for partially exfoliated nanocomposites [24, 27]. Peak intensity is found less as compared to Cloisite 30B, Neat PBT and of PBT/ULDPE-g-GMA, indicating the decrease in the degree of coherent layer stacking of clay.

### 3.2. Crystalline Morphology observed by POM:

Polarizing optical microscopy provides a basis for the study of the matrix crystalline morphology of the nanocomposites. Polarizing optical micrographs of neat PBT, PBT/IM (B) and PBT/IM/OMMT (C1,C2) nanocomposites during an isothermal crystallization process at  $200^\circ\text{C}$  for 30 min are shown in figure 3. Figure 3 shows that neat PBT have distinct spherulitic structures. The formation of spherulite is uniform throughout the matrix. The presence of 2wt% ULDPE-g-GMA (fig.3, b) in PBT matrix does not show significant effect on the spherulitic morphology of neat PBT. Composition C1 and C2 (fig.3, c,d) shows spherulite morphology of elastomer toughened PBT nanocomposites with 3 and 5wt% clay. From the micrographs it is clear that the PBT nanocomposites consists of higher amount of crystallites with much smaller size than that of neat PBT. C2 shows irregularly shaped crystallites. It shows that the incorporation of organoclay into polymers could lead to changes in their crystallization behavior. This reduction in crystallite size is due to the nucleation effect of clay. This implies that clay layers act as a heterogenous nucleating agent as it increases the nucleation density, promote the crystallization of polymer [28-30]. The degree of crystallinity and growth rate of spherulites are mainly depends on the nucleation and growth process of spherulites [30].

### 3.3. Effect of organoclay and ULDPE-g-GMA on the impact strength:

Table 2, depict the impact strength of the PBT/ULDPE-g-GMA/OMMT ternary nanocomposites as well as of neat PBT and PBT/ULDPE-g-GMA blend. The results from our study revealed that the incorporation of 2wt% of ULDP-g-GMA (Impact modifier) has substantially improved impact strength of neat PBT matrix. The impact strength increased from 54.1 J/m to 100.4 J/m, which is around an 85.6% increase in impact strength. The effectiveness of ULDPE-g-GMA elastomer in toughening of PBT is due to the high compatibility of a PBT/ULDPE-g-GMA blend. It may be seen that the impact strength of the ternary nanocomposite is lower than that of elastomer toughened PBT. It is also noted that the impact strength of the ternary nanocomposites is in between that of the neat PBT and PBT/ULDPE-g-GMA blend. This is may be due to the morphology of PBT/ULDPE-g-GMA blends in the presence of organoclay, and the dispersion of organoclay in the polymer matrix. It may also be due to the less compatibility of ULDPE-g-GMA and organoclay. This result is in favor of the literature. Li et al. [31] showed that the impact strength of the ternary nanocomposites is in between the that of the PBT and PBT/EVA-g-GMA blend.

**Table 2. Impact strength of PBT nanocomposites**

Compositions	Impact Strength (J/m)
Neat PBT	54.1
B	100.4
C1	78.1
C2	76.0

### CONCLUSION

XRD analysis shows that partial exfoliated rubber toughened PBT nanocomposites have been formed by melt intercalation. PBT polymer remains in  $\alpha$  crystal form after the incorporation of impact modifier. Examination with polarized optical microscopy shows that the clay contributes to the reduction in crystallites size of PBT in nanocomposites. Hence, clay layers can act as nucleating agent, thus greatly increase the nuclei number. Addition of impact modifier improved the impact strength of neat PBT. Hence, increases the toughness of PBT. Incorporation of small amount of OMMT slightly decrease the impact strength of elastomer toughened PBT nanocomposites. This improvement in the characteristics of PBT is especially beneficial to applications like automotive and electronic applications in which clay provided the insulation and lowers the weight of material at the same time along with the increased toughness obtained using impact modifiers.

### Acknowledgments

The authors would like to thank LARPM, CIPET, Bhubaneswar for their generous support of the facility for compounding. We are grateful to USIC, University of Delhi for HRXRD studies. The authors also would like to thank DSM Engineering Plastics and Pluss Polymers for kindly supplying the PBT and Impact modifier.

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