

**STUDIES ON POLYBUTYLENE TEREPHTHALATE/
POLYOLEFIN ALLOYS AND THEIR SHORT GLASS
FIBRE REINFORCED COMPOSITES**

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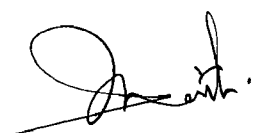


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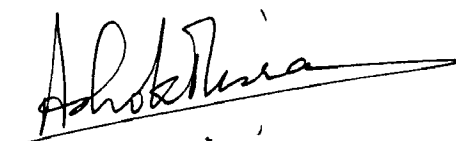
CERTIFICATE

This is to certify that the thesis entitled "Studies on Polybutylene Terephthalate (PBT)/Polyolefin alloys and their short glass fibre reinforced composites" being submitted by Mrs. Mangala Joshi to the Indian Institute of Technology, Delhi for the award of the degree of Doctor of Philosophy, is a record of bonafide research work carried out by her. Mrs. Mangala Joshi has worked under our guidance and supervision and has fulfilled the requirements for the submission of the thesis which to our knowledge has requisite standard.

The results contained in this thesis have not been submitted, in part or in full, to any other University or Institute for the award of any degree or diploma.



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ABSTRACT

Polymer blends and composites have gained enormous importance over past two decades as they frequently meet performance demands that can not be satisfied by currently available commodity polymers. Polymer blends and alloys offer the advantages of ease of processing and/or desirable properties in terms of the cost-performance criterion. Short fibre reinforced thermoplastics on the other hand offer the advantages of enhanced stiffness, strength, dimensional stability, toughness and superior temperature resistance. The present work deals with the studies on polybutylene terephthalate (PBT)/polyolefin alloy systems and their short glass fibre reinforced composites. Composites of blends are expected to show superior performance properties because of dual advantages offered by blending and reinforcement, so that they can find use in load bearing engineering applications.

PBT is a successful thermoplastic polyester but has certain draw backs such as low impact strength, inadequate heat distortion temperature, low melt viscosity and poor optical properties. An attempt has been made to modify the properties of PBT by blending it with polyolefin such as High Density Polyethylene (HDPE) and Polypropylene

(PP), since they have excellent mouldability, good chemical resistance and low cost. PBT/Polyolefin blends are expected to be incompatible owing to their widely varying solubility parameters [PBT - $10.7 \text{ (cal/cc)}^{1/2}$, HDPE - $9.02 \text{ (cal/cc)}^{1/2}$, PP - $8.03 \text{ (cal/cc)}^{1/2}$]. Hence an ionomer based on polyethylene has been used as a compatibiliser to form an alloy. For these ternary blends the PBT/Polyolefin ratio was fixed to 80:20 (by weight) and the ionomer levels chosen were 2%, 4% and 8% by weight. For developing composites, 80/20 blends of PBT/Polyolefin with and without ionomer (5% by wt) were selected. These were then mixed with 10, 20, 30% (by weight) of short glass fibre. The melt compounding of blends as well as composites was done on a single screw extruder, and test specimens prepared by injection moulding.

Various studies have been performed to determine the compatibilizing effect of the ionomer on the PBT/Polyolefin blend system. These include the mechanical properties; crystallisation characteristics by Differential Scanning Calorimetry (DSC) and Wide Angle X-ray Diffraction (WAXD); morphological studies by scanning electron microscopy (SEM), small angle light scattering (SALS) and polarising microscopy (PM); dynamic mechanical thermal analysis and rheological properties by capillary rheometer. Mechanical properties of PBT/polyolefin blends showed

significant improvement on addition of ionomer which was attributed to increase in interfacial adhesion between the blend constituents in presence of ionomer which acts as a compatibilizer. DSC and WAXD results show that the presence of ionomer facilitated crystallisation of PBT thus improving the overall crystallinity of the polymer blend system. DMTA studies show two distinct Tg values one for the PBT rich phase and other for the HDPE rich phase. This confirms that the blend is a two phase system. In presence of ionomer, Tg for the HDPE rich phase shifts to higher value indicating that amorphous PET is dissolving to a greater extent in the HDPE rich phase due to presence of ionomer. DMTA results were compared with published models which further supported the view that interfacial adhesion between the two phases improves with the presence of ionomer. Morphological studies also reveal compatibilizing effect of ionomer by showing a reduced size of dispersed HDPE domains and a more uniform dispersion of the two phases with increasing levels of ionomer. This is attributed to reduction in interfacial tension between the two polymers due to the emulsifying effect of the ionomer. SALS studies show that the superstructure morphology changes from spherulitic for PBT/HDPE system to sheaf-like for the blend with 4% ionomer content. The size of the spherulites also reduces when HDPE is blended with PBT. Rheological studies show that

viscosity of the blend with ionomer is higher than the blend without it. The die-swell values were found to increase on addition of ionomer. The results have been explained on the basis of state of dispersion of two phases with and without ionomer.

Studies carried out to characterize the short glass fibre reinforced composites of PBT/Polyolefin alloys show that Tensile and Flexural properties improve by 50 to 80% on addition of increasing amount of glass fibre as compared to unfilled system. DSC results show that in presence of glass fibre, the crystallisation temperature shifts to a higher value and the degree of supercooling lowers. This proves that presence of glass fibre facilitates crystallisation of the polymers in the blend with the glass fibre acting as a heterogeneous nucleating agent. DMTA studies show that addition of glass fibre reduced the magnitude of $\tan \delta$ peak and widened the transition peak as obtained from E'' vs. temperature plots. Relative storage modulus ($E'c/E'm$) increases as amount of glass fibre increases, at all temperatures. The drop in modulus on passing through transition region decrease as glass fibre is added. Rheological data was obtained at three different temperatures and varying shear rates. Melt viscosity increased whereas die-swell decreased on addition

of increasing glass fibre. Decrease in die-swell is advantageous from the processing point of view. Fibre length distribution studies indicate that extensive fibre damage occurs during compounding and moulding operations. Number average fibre length (l_n) decreases as glass content increases. Composite of the blend without ionomer shows better mechanical properties as compared to that of blend with ionomer which may be because of higher fibre breakage and less wetting of fibres by the matrix for samples with ionomer due to their higher viscosity, thus lowering the effective reinforcement efficiency. SEM examination of fractured tensile specimens show that fibre orientation along the flow direction is better at higher glass fibre content. For samples with ionomer, fibre pull out is more prominent whereas for samples without ionomer, fibre debonding is followed by fibre pullout. Due to this, some polymer is sticking on the surface of pulled out fibre. This indicate that polymer to polymer bonding is stronger than polymer to fibre bonding in presence of ionomer. The reverse is true for blends without ionomer. This is also reflected in the superior mechanical properties of the latter. Using established models, interfacial shear stress τ has been calculated. It is seen that values increase with composite strain. The values obtained using two

models are almost identical and are about 80% of the value of tensile strength for the unfilled matrix.

In the end, the results from various techniques of characterisation have been correlated and major conclusions have been highlighted. Suggestions for future work have also been listed at the end. It has been clearly shown that PBT/Polyolefin can be compatibilized by an ionomer to give an excellent alloy with superior properties. However, when reinforced with short glass fibre, the blend without the ionomer is more desirable. Blends and short fibre reinforcement have been simultaneously employed to produce materials with improved properties, specially the impact strength and melt strength for engineering applications.

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