



## Mechanical, thermal, rheological and morphological behavior of poly(butylene terephthalate)/poly(propylene) blends compatibilized with poly(propylene)-g-acetic anhydride

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### INTRODUCTION

The development of polymer blends, composites and laminates is of great economic importance for the plastics industry and for other industries where the use of such products is becoming increasingly common. Advanced polymer modification techniques have grown in importance during the last two decades as the “point of diminishing returns” has been approached in improving the performance/price balance by altering just the chemical structures of polymers. Most pairs of polymers are immiscible with each other. Even worse is the fact that they also have less compatibility than would be required in order to obtain the desired level of properties and performance from their blends. Compatibilizers are often used as additives to improve the compatibility of immiscible polymers and thus improve the morphology and resulting properties of the blend.

Polyolefins and poly(alkyl terephthalate) are considered immiscible and incompatible. Only a very limited number of studies on these blends have been reported; however, they have received considerable interest lately.<sup>[1-3]</sup> Polypropylene (PP) is neither miscible nor compatible with poly(butylene terephthalate) (PBT) due to a great difference in polarity. In addition, PP does not contain the necessary functional group to react with PBT. Adding a functionalized polymer miscible

with PP to react with PBT may improve the compatibilization of the PP/PBT blends.

In one of the research paper,<sup>[4]</sup> ethylene-co-glycidyl methacrylate (EGMA) was demonstrated to be a good compatibilizer for the PP/PBT blends. EGMA is a reactive copolymer where the ethylene component is compatible with PP, while the glycidyl groups are able to react with hydroxyl or carboxylic terminal groups of the PBT. In this article, a combination of the commercial available maleic anhydride-grafted polypropylene (denoted as PP-MA hereinafter) and a multifunctional epoxy resin (i.e., tetraglycidyl ether of diphenyl diaminomethane) was used as dual reactive compatibilizers for the PP/PBT blends. The PP-MA with a low MA content is totally miscible with PP to make the PP phase quasi-functionalized, so that the multifunctional epoxy has the chance to contact and react with PBT and PP-MA simultaneously to form PP-MA-co-epoxy-co-PBT copolymers at the interface. These desired copolymers are able to anchor along the interface and are expected to serve as efficient compatibilizers. The epoxy resin, tetraglycidyl ether of diphenyl diaminomethane (TGDDM), with the trademark of NPEH-434, was used as a compatibiliser for PBT/PP blends. Tetraglycidyl ether of diphenyl diaminomethane (TGDDM). The reactions between epoxy and anhydride groups of PP-MA can be initi-

ated by a hydroxyl-containing compound to proceed ring-opening reaction.<sup>[5]</sup> The mixture of PP-MA and an epoxy resin was demonstrated to be an efficient dual reactive compatibilizer for immiscible and incompatible PP/PBT blends. The PP-MA with a low MA content is miscible with PP to make it quasi-functionalized, while the multifunctional epoxy has the chance to react with PBT and PP-MA at the interface simultaneously.<sup>[6]</sup> Thus, the in situ-formed PP-MA-co-epoxy-co-PBT copolymers are able to anchor along the interface and serve as efficient compatibilizers. The mechanical properties of the PBT rich blends are improved by increasing the epoxy content to 0.3 phr. An epoxy content of 0.5 phr can lead to a light crosslinking in the PBT phase of the PBT-rich blends and result in a lack of a compatibilization effect. Joung Sook Hong, et al found that when PE was blended with PBT slightly below the melting temperature of PBT, rigid PBT forms a film structure by drag. In this condition, the PBT phase does not evolve to droplet structure unless a significant amount of additional heat or shear is applied, and it can maintain its structure during extrusion. On the contrary, when the processing temperature is higher than the melting temperature of PBT, a thermodynamically unstable film or fibril structure observed at an early stage of morphology development easily evolves to droplet structure during extrusion.<sup>[7]</sup>

In this paper PP-g-AA (containing 0.2 wt% AA level)

is utilized as a compatibilizer for the PBT/PP blend so as to improve its performance properties. The PP-g-AA compatibilized PBT/PP blends were characterized for mechanical, thermal and rheological properties.

## MATERIALS AND METHODS

### Materials

Poly(butylenes terephthale) (Arnite TO 6200) was amiably supplied by DSM Engineering Plastics Ltd., Pune, India. Poly(propylene) (H110MA) was procured from Reliance Industries Pvt. Ltd., Mumbai, India. PP-g-AA (Polybond 1002) was obtained from Uniroyal Chemicals, USA. All the materials were used as obtained without modification or treatment.

### Methods

Prior to blending, PP and PBT pellets were dried at  $100 \pm 5^\circ\text{C}$  for 4–5 hours in an air-circulating oven. The drying is essential to remove the moisture absorbed or adsorbed by the materials. This protocol was followed for all the batches. Also for the compatibiliser, was dried at  $80^\circ\text{C}$ .

All the compositions studied in the present work along with their coding system are presented in TABLE 1. The pre-dried components were dry blended in desired ratio before feeding to co-rotating twin-screw extruder (Model MP 19 PC, APV Baker Ltd, U.K),

TABLE 1 : Prepared uncompatibilized and compatibilized blends of PBT/PP

Code	Poly(butylene terephthalate)		Poly(propylene)		PP-g-AA
	gm	%	gm	%	
CPBT	800	100	0	0	0
PBT/ PP=90/10	720	90	80	10	0
PBT/ PP=80/20	640	80	160	20	0
PBT/ PP=70/30	560	70	240	30	0
PBT/ PP=60/40	480	60	320	40	0
PBT/ PP=50/50	400	50	400	50	0
PBT/ PP=20/80	300	20	500	80	0
CPP	0	0	800	100	0
PBT/PP/ PP-g-AA=90/10/5	800	100	0	0	40
PBT/PP/ PP-g-AA=80/20/5	720	90	80	10	40
PBT/PP/ PP-g-AA=70/30/5	640	80	160	20	40
PBT/PP/ PP-g-AA=60/40/5	560	70	240	30	40
PBT/PP/ PP-g-AA=50/50/5	480	60	320	40	40
PBT/PP/ PP-g-AA=20/80/5	400	50	400	50	40

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TABLE 2 : Processing conditions used for preparing PBT/PP blends

Process		Temperature Profile (°C)						Screw Speed
Blending	Extrusion	Zone 1	Zone 2	Zone 3	Zone 4	Die	Water Bath	40 rpm
		120	180	240	245	250	25-30	
Sample Preparation	Injection Molding	Zone 1	Zone 2	Nozzle	Mold	Cooling Time		Injection Pressure
		150	235	250	30-40	60 sec		

having L/D of 25:1. Mixing speed of 40 rpm was maintained for all the compositions. The extrudate from the die was quenched in a tank containing water at 20-30°C and then pelletized. Processing conditions such as temperature profile in different zones and the extruder die as well as speed of blending etc. for melt blending are given in TABLE 2.

The extrudate were pelletized using Boolani's pelletizer machine. The rpm of the pelletizer was maintained between the ranges of 60-80. Since the systems studied were incompatible with each other, thus they showed high die swell. Thus, the extrudate had to be grinded in a grinder where it was too thick for pelletization.

The pre dried (90°C for 4-5 hours in an air circulating oven) pellets of blends were injection molded using family mould cavity in a microprocessor-based injection molding machine (Boolani Industries Ltd., Mumbai, India). The family mould have tensile, flexural and impact test specimen cavities. The processing conditions for injection molding are given in TABLES 2. The test specimens pertained to ASTM standards (Tensile - ASTM D638, Flexural - ASTM D790, Impact - ASTM D256).

### Characterization

The following methods were employed for determining the specific properties of the polymer blend. Flow chart of the study is shown in Figure 1.

### Mechanical properties

Tensile (tensile strength, tensile modulus and percentage elongation at maximum load) and flexural (flexural strength and flexural modulus) properties were measured at ambient condition using a Universal Testing Machine (LR-50K, Lloyds Instrument, UK), according to ASTM procedures D638 and D790; at a cross-head speed of 50 mm/min and 2.8 mm/min respectively. The notch for impact test was made using a motorized notch-cutting machine (Polytest model 1, Ray

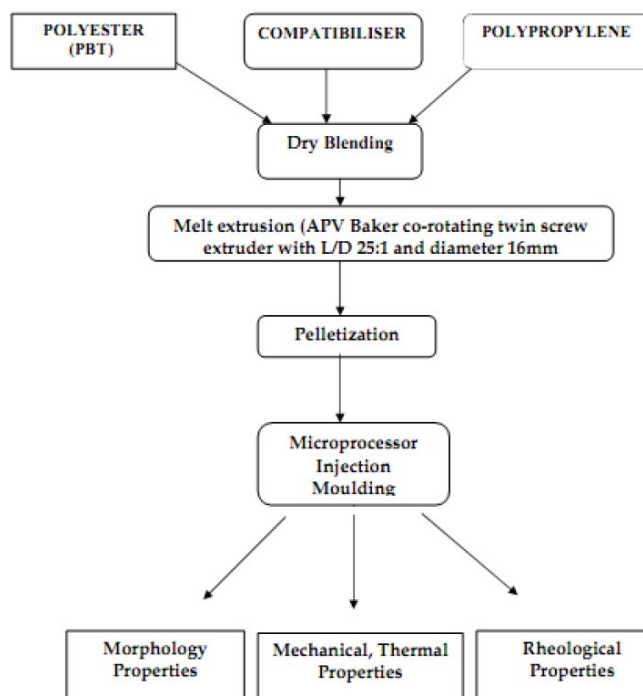


Figure 1 : Flow chart of the study from preparing to analyzing PBT/PP blends

Ran, UK). Notched Izod impact strength was determined at ambient condition according to ASTM D256, using impact tester (Avery Denison, UK) employing a 2.7J striker, having striking velocity of 3.46 m/s.

### Thermal properties

Differential Scanning Calorimetric (Q 100 DSC, TA instruments Ltd., India) characterization was done to investigate the crystallization and melting behaviour of the prepared nanocomposites. Two consecutive heating scans were determined to minimize the influence of possible residual stresses in the material due to any specific thermal history. Scanning rate of 10°C/min was maintained for both heating and cooling cycle; whereas nitrogen gas purge rate was maintained at 50 ml/min. Melting temperature ( $T_m$ ) was determined from the second heating scan, while the crystallization temperature ( $T_c$ ) from the only cooling scan.

## Rheological properties

Rheological properties of different compositions were measured using Haake rheometer RT10 parallel plate viscometer. Melt viscosity ( $\eta$ ) as a function of shear rate ( $\dot{\gamma}$ ) was measured at 250°C. The shear rate was varied over 0.001s<sup>-1</sup> to 1000s<sup>-1</sup>. The samples were predried before rheological analysis. The test was also carried out in oscillatory mode with frequency range varying from 0.01 to 40 Hz at 250°C. Phase angle  $\delta = G3 / G2$  was determined as a function of frequency  $\omega$  (rad /s).

## Morphological properties

The morphology of PBT/PP blends were investigated using SEM with a JSM-840, JEOL instrument at an accelerating voltage of 10 KV. The electrons emitted from the electron gun were focused on the object surface. The secondary electrons emitted from the sample were monitored by suitable detectors. The adjustment of the magnification depends on the size of the scanned surface. The samples were taken from injection molded tensile specimen. The dumbbells were randomly chosen and etched in hot toluene to remove one of the phases. Gold sputtering devices were used to coat polymer samples, which have a most pronounced surface topography with a conducting metal layer.

## Mechanical properties

The CPBT has greater tensile strength than the all compositions of the blend with and without compatibiliser PP-g-AA. Figure 2 shows that the tensile strength of the PBT/PP blends were much lower than that of PBT but was much higher than that of PP. Though, on addition of compatibiliser like PP-g-AA improved the tensile strength, the difference between the values of uncompatibilised and compatibilized blends was about 8-10%. The improvement in mechanical properties of the blend by the addition of the compatibiliser may have resulted due to the improvement in the interfacial adhesion of the blend caused due to decrease in surface tension. 10 wt. % PP addition could make the blends tensile strength decrease significantly. In addition, both the tensile strength and the flexural strength exhibited a near-linear decrease with increasing PP content. Since both PBT and PP are brittle

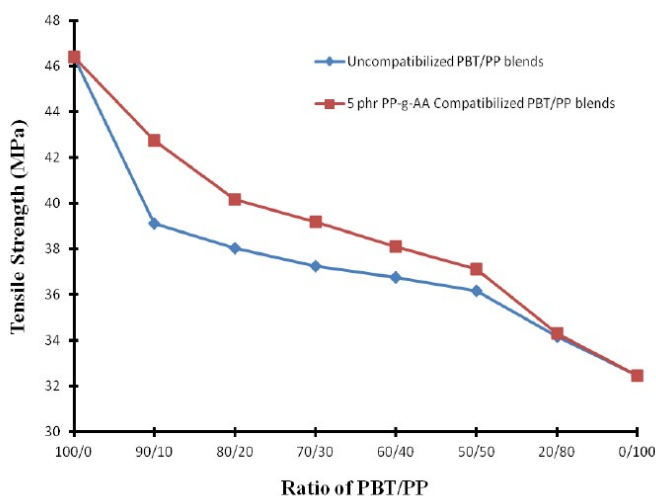


Figure 2 : Tensile Strengths obtained for uncompatibilized and compatibilized PBT/PP blends

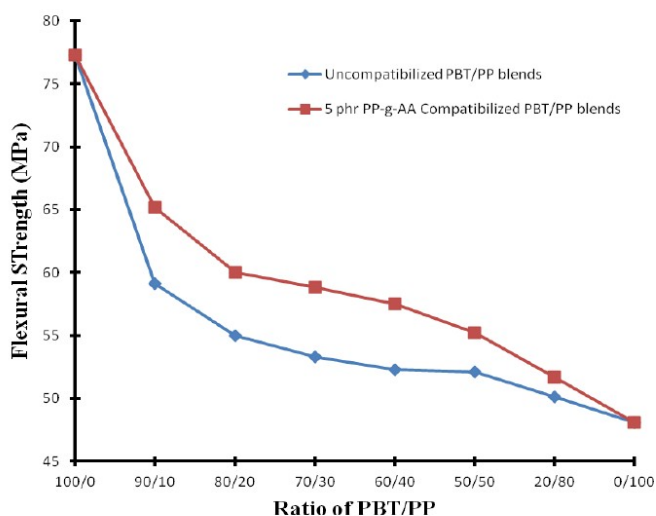


Figure 3 : Flexural Strengths obtained for uncompatibilized and compatibilized PBT/PP blends

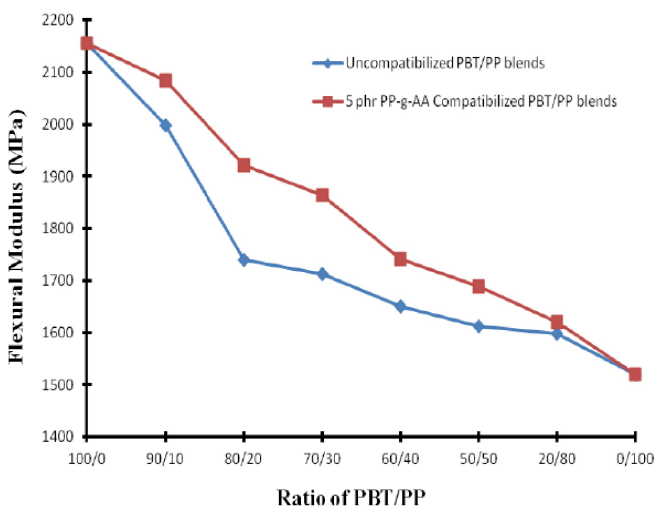
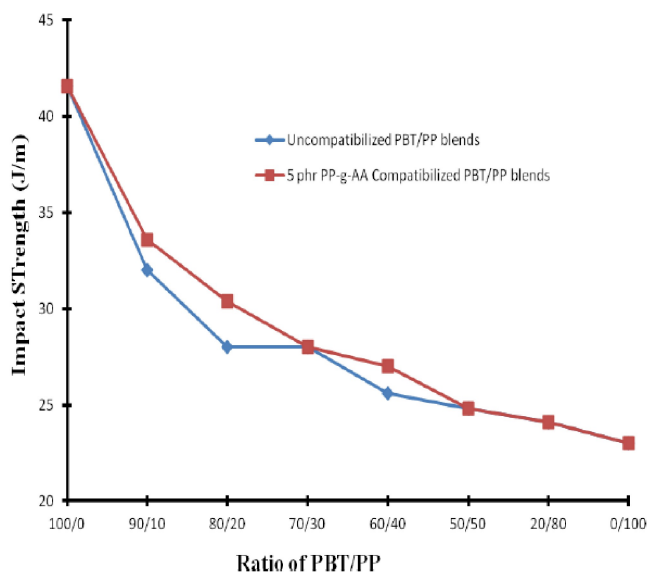


Figure 4 : Flexural Modulus values obtained for uncompatibilized and compatibilized PBT/PP blends

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**Figure 5 : Impact Strength values obtained for uncompatibilized and compatibilized PBT/PP blends**

under standard notched Izod impact testing, all PBT/PP blends, uncompatibilised or compatibilized, are also brittle with nearly identical impact strength as would be expected. The notched impact strengths for PBT/PP blends with compatibiliser were increased than those of uncompatibilised blends. Thus PP-g-AA is successfully able to improve the compatibilization between PBT and PP.

### Thermal properties

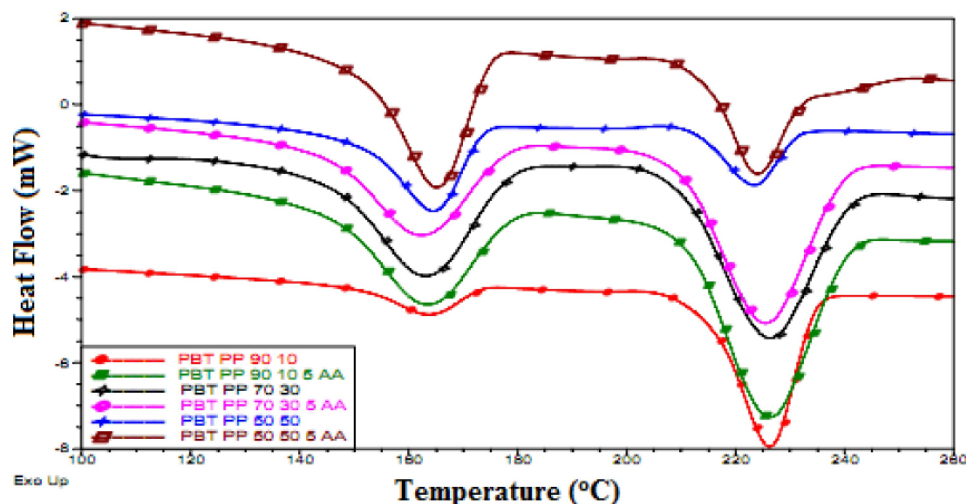
It can be observed that from the table that the addition of PP into PBT has some influence in the melting as well as the crystallization temperature of the PBT.

The addition of PP to PBT results into binodal melting peaks of the blend & there was slight variation in the temperature of the melting peaks of the blends. The crystallinity of the PBT has decreased with addition of PP and the enthalpy values have also reduced with further addition of PP to PBT. The melting behavior of PTT/PP blends are shown in Figure 6.

The crystallization peaks for the blends have also shown the similar trend for these compositions of the blend & reduction in the enthalpy values has confirmed that the addition of PP have definitely interfered with the crystallization process of PBT. The crystallization peaks has shifted to the lower side by addition of the compatibiliser PP-g-AA. The crystallization temperature has reduced by 6°C for PBT/PP (80/20) blend by the addition of the PP-g-AA. The reduction in the crystallization temperature may be due to the partial miscibility of the blends due to addition of the compatibilisers. It is clearly seen that the onset temperature of the blends does not show large variation for all composition of the blends, but the crystallization temperature of the blends were certainly affected due to the addition of the compatibiliser. The following TABLES' viz. 3 & 4 illustrates the melting temperature variation & crystallization temperature variation for PBT/PP blends with addition of component PP and the compatibiliser PP-g-AA.

### Rheological properties

The shear viscosity of a polymer melt is the most important function in characterizing the rheological be-



**Figure 6 : DSC melting curves obtained for uncompatibilized and compatibilized PBT/PP blends**

TABLE 3 : Melting characteristics obtained for PBT/PP blends

Blends	Start (°C)	Onset (°C)	Maximum (°C)	Stop (°C)	Area (J/g)
CPBT	194.64	204	222.4	242.12	51.32
PBT/ PP=90/10	201.63	215.05	226.21	242.12	38.15
PBT/ PP=80/20	196.34	207.12	225.9	242.15	36.45
PBT/ PP=70/30	198.41	214.45	223.93	249.28	22.85
PBT/ PP=60/40	201.63	215.38	225.22	242.12	25.94
PBT/ PP=50/50	201.28	213.21	223.38	242.83	16.25
PBT/ PP=20/80	198.14	210.8	225.17	242.15	22.47
CPP	121.42	149.83	167.55	187.48	84.82
PBT/PP/ PP-g-AA=90/10/5	195.19	209.98	225.34	243.91	30.43
PBT/PP/ PP-g-AA=80/20/5	197.34	208.51	224.9	243.55	35.25
PBT/PP/ PP-g-AA=70/30/5	197.69	210.96	226.13	247.49	29.8
PBT/PP/ PP-g-AA=60/40/5	196.26	209.04	225.07	243.55	32.98
PBT/PP/ PP-g-AA=50/50/5	198.77	209.93	226.15	249.64	26.19
PBT/PP/ PP-g-AA=20/80/5	197.34	209.08	225.07	243.55	22.07

TABLE 4 : Cooling characteristics obtained for PBT/PP blends

Blends	Start (°C)	Onset (°C)	Maximum (°C)	Stop (°C)	Area (J/g)
CPBT	212.53	204.41	191.77	154.95	44.78
PBT/ PP=90/10	215.97	202.69	196	175.84	40.46
PBT/ PP=80/20	212.02	202.1	195.14	176.11	38.85
PBT/ PP=70/30	214.53	202.14	195.34	177.63	34.3
PBT/ PP=60/40	212.74	201.86	194.91	174.41	31.27
PBT/ PP=50/50	209.87	201.41	194.29	176.91	28.9
PBT/ PP=20/80	213.1	202.15	189.36	163.66	26.61
CPP	132.98	118.39	110.71	88.84	101.9
PBT/PP/ PP-g-AA=90/10/5	214.89	204.4	191.47	167.96	33.73
PBT/PP/ PP-g-AA=80/20/5	211.31	203	190.05	167.24	33.07
PBT/PP/ PP-g-AA=70/30/5	217.4	205.02	192.19	168.32	32.11
PBT/PP/ PP-g-AA=60/40/5	209.16	201.32	188.33	167.6	31.22
PBT/PP/ PP-g-AA=50/50/5	210.95	202.9	189.99	166.17	31.97
PBT/PP/ PP-g-AA=20/80/5	213.1	202.15	189.36	163.66	26.61

havior of the polymer melt; it will reflect the variation in interface interaction. The rheological properties of multiphase polymer blends are strongly influenced by their interfacial characteristics. Investigation of the rheological properties of blends can show information about the compatibilization effect, and can also reflect the correlation of rheology-morphology-mechanical properties of the blends.<sup>[8,9]</sup>

Figure 7 gives plots of the shear viscosity versus the shear rate for PP and PBT measured at 250°C. Due to the more rigid chain structure of PBT than that of PP, the PBT shows higher shear viscosity than PP &

less shear thinning behaviour. The increase in viscosity due to the presence of the compatibiliser indicates that the compatibiliser in the blend is able to react with PBT and leads to a higher PBT molecular weight. The existence of reactions between PBT and the compatibiliser in 80/20 blend can be found, where the viscosities of PBT/PP/PP-g-AA blends were increased with incorporation of the compatibiliser. As the percentage of the PBT in PP rich phase i.e. PBT/PP (20/80) blend the viscosity does show any remarkable improvement due to shortage of PBT for reactions with an amount of compatibiliser.

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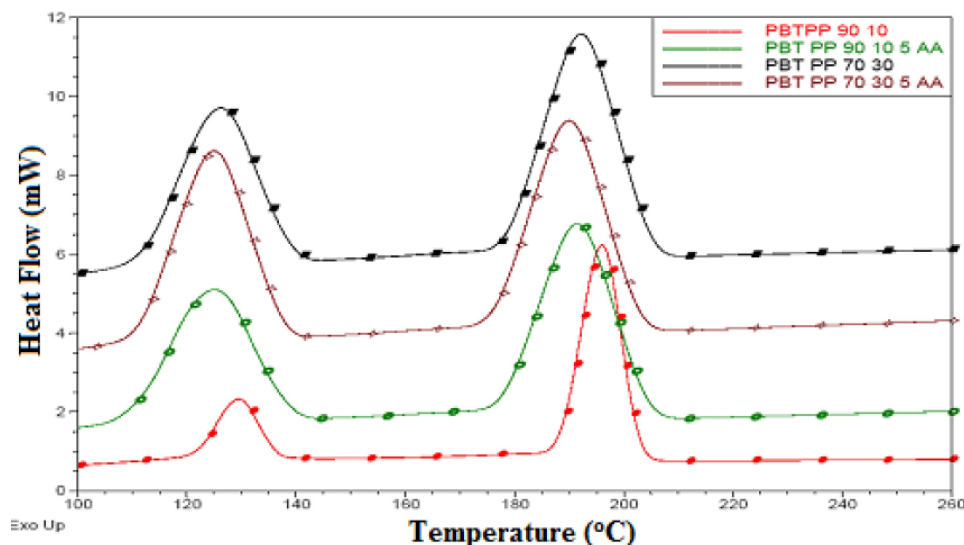


Figure 7 : DSC cooling curves obtained for uncompatibilized and compatibilized PBT/PP blends

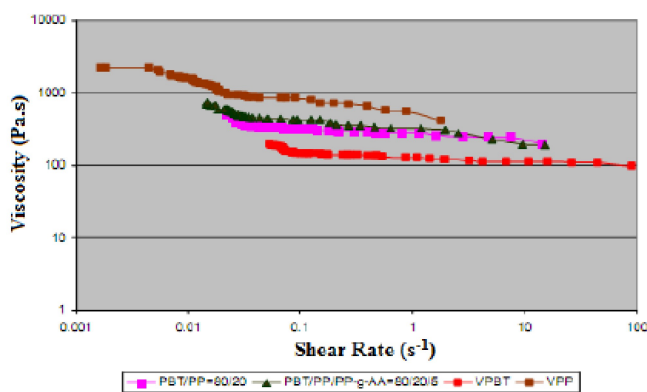


Figure 7b : Graph of Viscosity vs Shear Rate, obtained for uncompatibilized and compatibilized PBT/PP 80/20 blend

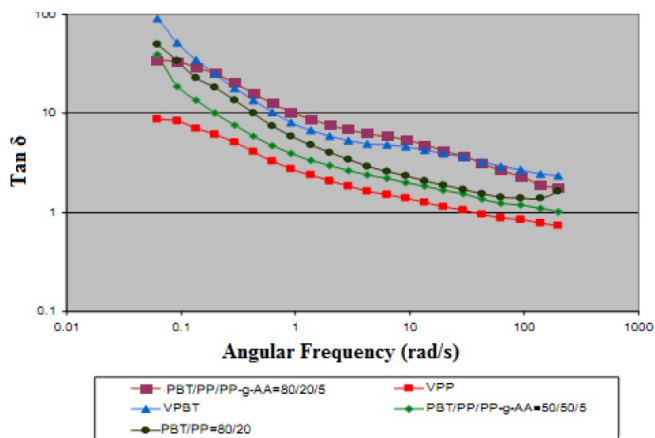


Figure 8 : Graph of Tan( $\delta$ ) vs Angular Frequency, obtained for uncompatibilized and compatibilized PBT/PP 80/20 blend

The Figure 8 shows  $\tan \delta$  values of the blend components at the processing temperature, 250°C. It is clear from the graph that the blend components as well as the blends have shown viscoelastic nature, as their ab-

solute value is close to 1. From the  $\tan \delta$  curves shown in Figure 8 it is observed that, at 20% PP concentration, when PP-g-AA was used as the compatibiliser,

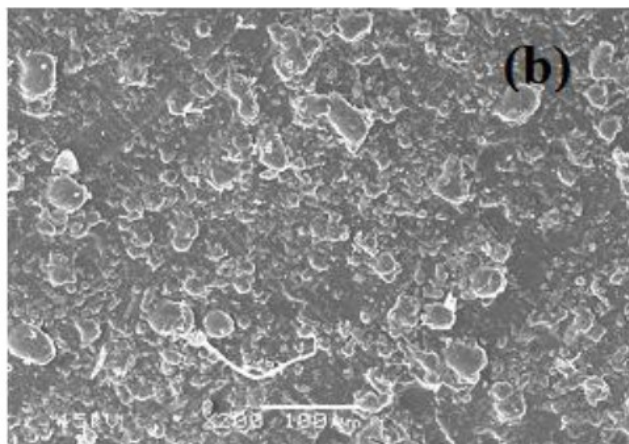
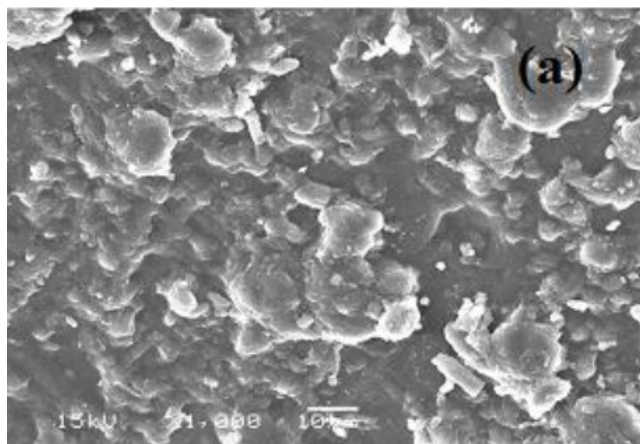


Figure 9 : SEM images obtained for PBT/PP 80/20 (a) and PBT/PP/PP-g-AA (b)

the  $\tan \delta$  curves became increasing than the uncompatibilised one. The  $\tan \delta$  values for the PP-g-AA compatibilized blends were found to be higher than the other corresponding blends.

### Morphological properties

Figure 9 (a) shows SEM micrographs of the PBT/PP (80/20) blend. PTT/PP was a biphasic blend. For the uncompatibilized blends, it can be seen that there is no evidence of interfacial interactions or adhesion, suggesting that the PBT and PP phases were incompatible. It was interesting that for the compatibilized blends there were broken fibers or small bumps on the fractured interfaces with evidence of their being drawn out; some unbroken fibers connecting the PBT and PP phases also appeared. It was concluded that these phenomena were the results of the reactive compatibilization, i.e., the reaction between the acrylic acid functional groups of PP and the hydroxyl end groups of PBT formed polymer connections at the interfaces as seen from Figure 9 (b). These SEM morphological results provided evidence that PP-g-AA could improve the interfacial adhesion of PBT/PP during melt blending and consequently would theoretically enhance its mechanical properties.

### CONCLUSION

The PP-g-AA is potentially reactive towards the carboxylic and/or hydroxyl groups at the chain ends of PBT. The mechanical properties like tensile strength, flexural strength of PBT/PP blend is lower than of vir-

gin PBT, but was much higher than PP. The addition of the compatibiliser PP-g-AA has increased these properties, though the improvement in the impact properties of the blend was not significant. Rheological characterization has confirmed a molecular-weight buildup due to the interfacial reaction between the AA-grafted PP and PBT. Thermal analysis of the blend showed that the melting temperature of PBT has showed a slight decrease, while melting temperature of PP has showed a slight increase about 5°C. This was also confirmed through SEM analysis.

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