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## The Effects of Chain Extender on Viscosity and Mechanical Properties of Poly (Butylene Terephthalate) Blending with Recycled Poly (Ethylene Terephthalate)-glass Fiber Composite

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**Abstract:** In this study, multi-functional styrene-acrylic oligomers was used as the chain extender in poly butylenes terephthalate/recycled glass-filled poly ethylene terephthalate (PBT/RGF-PET) blends. Normally, usage of recycled plastics is favourable but somehow, during the reprocessing causes loss of properties. Thus, chain extender was added to restore the viscosity and mechanical properties of PBT/PET blends due to the incorporation of recycled poly (ethylene terephthalate) has lowered viscosity and molecular weight. Chain extender at 0.50 and 0.65 wt.% was added into 50/50 PBT/RGF-PET using melt compounding method to compare viscosity and mechanical properties with virgin PBT and RGF-PET, respectively. Addition of chain extender has improved the viscosity of the PBT/RGF-PET which was in relation to the increment of molecular weight. When 0.50 wt.% of chain extender was added to PBT/RGF-PET blend at ratio of 50:50, the flexural modulus increased 9.6% to 3530 MPa compared to the original flexural modulus at 3220 MPa. This showed that a small amount of chain extender was successfully to improve the flexural modulus. The increment of molecular weight as induced by the addition of chain extender improved the impact resistance of the PBT/RGF-PET from 17 to 20.3 kJ m<sup>-2</sup> for addition of 0.50 wt.% chain extender.

**Key words:** Poly butylene terephthalate, recycled-glass-filled poly ethylene terephthalate, chain extender, viscosity, mechanical properties, styrene-acrylic oligomers, recycling

### INTRODUCTION

Poly Ethylene Terephthalate (PET) is well known of its applications in fabrication of beverage bottles, packaging containers or engineering materials in combination with glass fibers (Kusuktham, 2011). It has simple processing technology and attachable for extrusion moulding, blow moulding, injection moulding and thermoforming methods. Although, PET is a semi-crystalline, PET possesses slow crystallization rate which limits its application as injection moulding resins before the nucleating agent was introduced to promote fast crystallization rate of PET. On the other hand, Poly Butylene Terephthalate (PBT) has rapid crystallization rate with fast moulding cycle (Scheirs and Long, 2003). When PET and PBT are blended together, the blend can be well moulded at normal mould temperature around 80°C without addition of compatibilizing agent.

However, the polycondensation thermoplastics such as PET and PBT tend to undergo severe degradation

when they are processed at high temperature, i.e., hydrolysis and thermal cleavage regularly happens especially during recycling process (Frounchi, 1999; Omrani *et al.*, 2005). Camacho and Karlsson (2000) reported that it is very frequent for the recycled plastics to undergo undesirable change of mechanical, physical and chemical properties as compared to the original polymers due to excessive heat and shear effects. For instance, Mark (2004) has reported the initial intrinsic viscosity (IV) of the virgin PET resin (V-PET) dropped from 0.75-0.85 to 0.62-0.65 dL g<sup>-1</sup> resulted from the postconsumer recycled recovery process and subsequent melt-compounding.

Many study have reported the recycling effects on physical, mechanical, thermal, or rheology properties of recycled PET (R-PET) by comparing to V-PET as well as the chemical modification or chain extension to improve the intrinsic viscosity and molecular weight of V-PET or R-PET (Shah and Gasaway, 2007; Haralabakopoulos *et al.*, 1999; Villalobos *et al.*, 2006). Type of chain extenders

such as hydroxyl chain extender, diisocyanates, bis-oxazolines, diepoxides, polyepoxides, bis-N-acyllactam and biscaprolactam have been studied in polycondensates polymers for typical PET and PBT (Loontjens, 2003). On the other hand, recycled glass-filled PET (RGF-PET) is seldom reused as compared to post-consumer PET. This is due to R-PET has low viscosity and molecular weight with possible post-contamination which might lead to inferior properties (Felix *et al.*, 2011). Hence, this study was focusing on comparing the flexural and impact properties as well as the viscosity number of the chain extender effects on the RGF-PET blending with PBT.

### MATERIALS AND METHODS

**Materials and preparation of blends:** PBT resin, Ultradur® B2550 was supplied by BASF. Recycled glass-filled PET source was supplied by 3T Industrial Sdn. Bhd. Styrene-acrylic multi-functional oligomer, a polyepoxides type of chain extender, Joncryl ADR-4368, was supplied by BASF. PBT resins and recycled glass-filled PET (RGF-PET) were dried in air circulation oven at 120°C for 4 h to reduce moisture content prior dry blending and compounding. Several samples as listed in Table 1 were prepared to determine the effects of chain extender to the PBT/RFG-PET blends. Co-rotating twin screw extruder Berstorff ZE25 UTX (25 mm diameter and L/D 44), was used for melt compounding. The barrel temperatures were varied from 240 to 260°C and the melt temperature was controlled at 260°C. The extrudate was cooled at water bath and cut into pellet form. The pellet samples were dried in air circulation oven at 120°C for 4 h. Injection moulding machine Krauss-Maffei 130-380 CX was used to mould ISO 3167 Type 1A specimens. Barrel temperatures were varied from 240 to 265°C to achieve the melt temperature 260°C. Mould temperature was controlled to 80°C and total cycle time was 40 sec. The specimens were conditioned in environment 23°C and 50% relative humidity for minimum 88 h prior testing.

**Determination of viscosity and mechanical properties:** Viscosity number was determined according to ISO 1628-5. Small amount of pellet sample was dissolved in phenol/1,2-dichlorobenzene (1:1) to a 0.005 g mL<sup>-1</sup> concentration solution. An Ubbelohde viscometer type

capillary Ic was used to determine the flow times of the sample solution and solvent at a water bath temperature of 25°C. On the other hand, mechanical properties were conducted in controlled laboratory atmosphere with (23±2)°C and (50±10)% relative humidity. Meanwhile, the Melt Volumetric Rate (MVR) of the molten blends was determined using Ray-Ran Melt Flow Indexer according to ASTM D1238-10 at load 2.16 kg and temperature 260°C. Flexural properties were conducted using Zwick/Roell Universal Tester Machine with 2.5 kN loadcell according to ISO 178, with testing speed of 2 mm min<sup>-1</sup>. Unnotched Charpy impact strength was determined using Zwick/Roell Pendulum Impact Tester with 2 J pendulum according to ISO 179-1, with striking speed of 2.9 m sec<sup>-1</sup>.

### RESULTS AND DISCUSSION

**Viscosity behavior:** As shown in Table 2, the viscosity number of PBT has reduced after blending with RFG-PET. The low RFG-PET viscosity is because reprocessing has caused chain cleavage and depolymerization. This was also reported by Jabarin and Lofgren (1984) that thermal oxidation was the main degradation approach of PET. In this study, it was observed that the viscosity number of pre-processing RFG-PET has viscosity number at 62 mL g<sup>-1</sup> and dropped to 57 mL g<sup>-1</sup> after subjected to heat and shear effects in the extrusion process. In consequence, the PET-RGF has lack of entanglement which directly promotes chain sliding lowered the viscosity. Beside chain scission has caused dropped of macromolecular sizes, there was possibility breakage of glass fibers during the shear effect in the extrusion process. Repetitive processing is unfavourable because prolong exposure to extreme conditions is the main cause of severe degradation.

In contrast, the MVR outcomes showed that the molten viscosity of the PBT/RFG-PET has reduced at the higher composition of RGF-PET. The higher MVR indicates higher flow ability thus lower in viscosity. This observation can explain that PET has higher melting point (255°C) than PBT at 223°C. When measurements were done at the similar temperature, inherently PET has higher

Table 1: Blending formulation

Samples	PBT (wt.%)	RGF-PET (wt.%)	Chain extender
100/0-N	100	0	-
50/50-N	50	50	-
0/100-N	0	100	-
50/50-E50	49.75	49.75	0.50
50/50-E65	49.675	49.675	0.65

Table 2: Viscosity number and MVR of PBT, RGF-PET and blends with the addition of chain extender

Samples	RGF-PET content (wt.%)	Chain extender content (wt.%)	Viscosity No. (mL g <sup>-1</sup> )	MVR (cm <sup>3</sup> /10 min)
100/0-N	-	-	108	64
50/50-N	50	-	81	49
0/100-N	100	-	57	16
50/50-E50	49.75	0.50	110	7.47
50/50-E65	49.675	0.65	Not dissolved	5.54

viscosity that PBT. This was well exhibited in the MVR of RGF-PET where it was four-fold lower than the PBT when tested at 260°C. In fact, this temperature was slightly above the melting point of PET. This observation was also important to decide the melt temperature to be used in injection moulding process to enable good processability by controlling the melt temperature at 260°C to avoid severe degradation where virgin PBT can withstand temperature up to 275 °C (Samperi *et al.*, 2004; McNeil and Bounekhel, 1991).

In this study, it was found that viscosity number for sample 50/50-N was 81 mL g<sup>-1</sup> before adding chain extender. After adding 0.50 wt% of chain extender, the sample 50/50-E50 possessed viscosity number at 110 mL g<sup>-1</sup> which was 36% higher than the viscosity number without chain extender. However, there was no viscosity number result for sample 50/50-E65. It was because the respective sample was not fully dissolved in the phenol/1,2-dichlorobenzene solvent in the Ubbelohde viscosity test. During the test, polymer gel was observed in the solution which means that the molecular weight of sample containing 0.65% chain extender were very high with the expected viscosity number was more than 110 mL g<sup>-1</sup>. According to Guo and Chan (1999) who have found that the polymer gel was formed when diepoxides chain extender was added into PBT. The gel content was 8 and 15%, respectively when 1.4 and 1.6 wt% of chain extender was added. The branching and crosslinking have occurred when the chain extender amount was high (Guo and Chan 1999; Villalobos *et al.*, 2006). In order to obtain gelation free PBT/RGF-PET blend at ratio of 50:50, the chain extender should be applied at the range 0.50 and 0.65 wt.% to achieve the optimum physical outcomes. The limited dosage (maximum 0.65 wt.% in this study) of multi-functional styrene-acrylic oligomers in PBT/RPET-GF blends may be due to the polymer blends has reached high branching and crosslinking environment which caused the polymer gel to form extensively. Although, the determination of viscosity number was difficult for sample 50/50 E65, MVR measurement was another good indication of high viscosity number of these both samples. It was observed that the MVR has reduced when the amount of chain extender increased. This indicated that higher chain extender remained in an increasing trend when the amount is >0.50 wt%. In other words, the addition of chain extender increased the molecular weight of PBT/R-GF-PET blends.

**Flexural properties:** Flexural modulus as summarized in Fig. 1 exhibited an increasing trend when the amount of RGF-PET was increased in the blend system. From flexural strength results in Fig. 2, it was found that sample 100/0

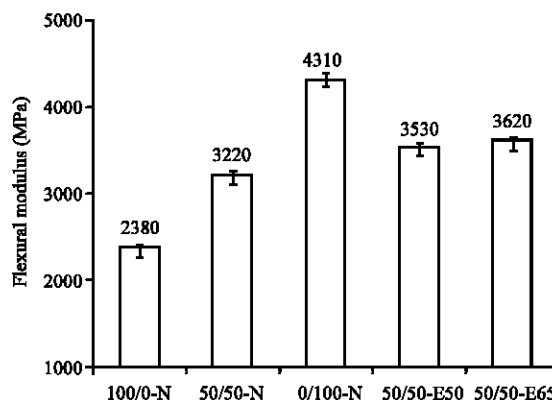


Fig. 1: Flexural modulus of PBT, RGF-PET and blends with the addition of chain extender

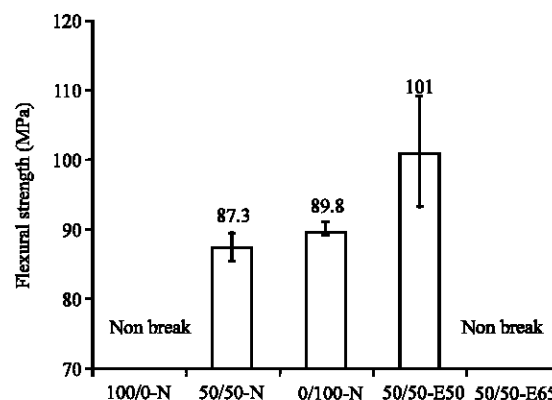


Fig. 2: Flexural strength of PBT, RGF-PET and blends with the addition of chain extender

did not break when the preset 5% maximum deformation was reached. The non-break behavior in virgin PBT showed a good ductility. When RGF-PET was added into PBT/RGF-PET blend system, the flexural strength was lowered indicated the inferior of the RGF-PET. In contrast, the flexural modulus of chain extended PBT/RGF-PET showed some increment after chain extender Joncryl ADR-4368 was added to the blends. When 0.50 wt.% of chain extender was added to sample 50/50-E50, the flexural modulus increased 9.6% to 3530 MPa, compared to sample 50/50 with flexural modulus at 3220 MPa. It showed that a small amount of chain extender was successfully to improve the flexural modulus. A slight increased in flexural modulus also reported in Zhou and his co-workers' study using epoxy-cyclohexyl polyhedral oligomeric silsesquioxane (epoxy-POSS) as chain extender in PBT.

Moreover, addition of chain extender has shown improvement of flexural strength of PBT/RGF-PET blend. When the amount of chain extender increased up to

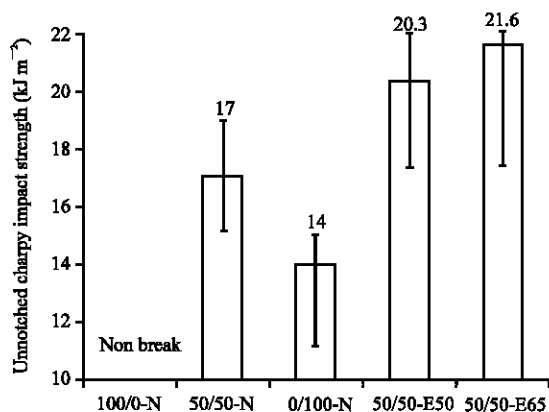


Fig. 3: Unnotched Charpy impact strength of PBT, RGF-PET and blends with the addition of chain extender

0.65 wt.%, the PBT/RGF-PET did not break when the preset 5% maximum deformation has been reached (Fig. 2). It was further expected the chain extender treated PBT/RGF-PET having equivalent flexibility as virgin PBT. Generally the branching and crosslinking of polymer chains increase the deformation strength as the result of limited chain mobility or flexibility. Such observation has been reported by Guo and Chan (1999) work about the chain extender was the preferable method to induce higher flexural strength when PBT was processed by reactive extrusion method.

**Impact properties:** As shown in Fig. 3, the sample 100/0-N consisted of virgin PBT did not break when subjected to the impact hammer indicated that PBT has high ductility or toughness. When the amount of RGF-PET was increased in the blend systems, the impact strength was decline. According to Chivers and Moore (1994), the toughness of the polymers is in relation to the molecular weight. The reduction of impact strength for the blends of PBT/RGF-PET happened at high contain of RGF-PET was mainly attributed to the affect of inferior characteristic of chain scissioning in recycled PET. Meanwhile, the impact strength of the blends showed somewhat increment after chain extender was added. The sample 50/50-E65 exhibited the highest strength due to higher molecular weight built up by chain extender has improved the impact resistance. The decrement in percentage of crystallinity and increment in toughness explained the improvement of impact resistance whereby the cracks can propagate more readily in the crystallite.

### CONCLUSION

In conclusion, the blending of RGF-PET has caused inferior effects to PBT. However, addition of chain styrene-acrylic polyepoxide type of chain extender helps

to improve the blending properties. Addition of chain extender has improved the viscosity of the PBT/RGF-PBT which was in relation to the increment of molecular weight. When PBT/RGF-PET blend at ratio of 50:50, the chain extender should be applied at the range 0.50 and 0.65 wt.% to achieve the optimum physical properties. When 0.50 wt.% of chain extender was added to PBT/RGF-PET blend at ratio of 50:50, the flexural modulus increased 9.6% to 3530 MPa compared to the original flexural modulus at 3220 MPa. This shows that a small amount of chain extender has successfully improved the flexural modulus. Meanwhile, 0.65 wt.% of chain extender added PBT/RGF-PET has exhibited equal flexibility as virgin PBT. The increment of molecular weight as induced by the addition of chain extender improved the impact resistance of the PBT/RGF-PET from 17 to 20.3 kJ m<sup>-2</sup> for addition of 0.50 wt.% chain extender.

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