FRACTURE BEHAVIOUR OF INJECTION MOULDED RUBBER TOUGHENED POLY (BUTYLENE TEREPHTHALATE) COMPOSITES

Z.A Mohd Ishak* and J Karger-Kocsis+

The fracture behaviour of poly butylene terephthalate (PBT), rubber toughened PBT (RTPBT) and their glass fibre composites have been investigated. The RTPBT systems are comprised of three different types of impact modifiers namely crosslinked acrylate (XLA), core-shell styrene-acrylonitrile (CSR) and ethylene acrylate functional (EAF) rubbers. The fracture mechanical characterisation of was evaluated by using notched compact tension (CT) specimens. The fracture toughness, K_C was observed to be strongly dependent on both internal (types of impact modifiers and glass fibre content) and external (temperature, strain rate and hygrothermal ageing) parameters.

INTRODUCTION

Poly butylene terephthalate (PBT) is becoming more attractive engineering thermoplastic owing to the fact that it possesses a good combination of properties (thermal, physical, electrical and mechanical) and processability (short extrusion and injection moulding cycles and excellent mould flow). However it suffers two major drawbacks in that it has limited toughness especially at high deformation rates and tendency to hydrolyse and embrittle in hot and moist environment (1). Thus considerable R&D efforts are targeted to solve these problems. Recently Czigany et.al, (2) have reported that toughness and hydrolytic stability of PBT can be enhanced through the incorporation of impact modifiers and short glass fibres into PBT matrix. The enhancement has been observed to be dependent on the quality of impact modifiers used.

It is the objective of this paper to assess and compare the fracture properties of short glass fibre(GF)-reinforced, injection moulded rubber toughened PBT (RTPBT) composites. Emphasis will be given in studying the effect of temperature, strain rate and hygrothermal ageing.

*School of Industrial Technology, Universiti Sains Malaysia, 11800 Penang, Malaysia.
+Institute For Composite Materials GmbH, University Kaiserslautern, PO Box 3049, D-67653 Kaiserslautern, Germany.

EXPERIMENTAL

GF-reinforced PBT at 30wt.% (PBT-GF) were supplied by BASF AG, Ludwigshafen Germany in the form of injection moulded plaques of dimensions 180x190xca.4 mm^3. The composites studied contained both neat PBT and RTPBT. The later is comprised of three different types of impact modifiers, namely crosslinked acrylates (XLA), core-shell type styrene/acrylonitrile (CSR) and funtionalised ethylene/acrylates (EAF) based rubbers, each of them present in 20wt.% in the compounds. Fracture mechanical characterisation of the various materials were performed on notched compact tension (CT) specimens (notch length, a=10mm, free ligament width, W=29mm).Testing of the specimens was carried out on a Zwick 1445 type tensile machine as a function of testing temperatures (T= -40°, 20° and $80^{\circ}C$) and strain rates (crosshead speed; 1 and 1000 mm/min). The effect of hygrothermal ageing was investigated by immersing the CT specimens in the water at $90^{\circ}C$ for about a week. The fracture toughness, K_C were calculated in accordance to the recommendation of the protocol of the ESIS TG-4. The detailed descriptions of all the experimental procedures followed in this investigation is given elsewhere (2).

RESULTS AND DISCUSSION

Effect of temperature Figure 1 shows the effect of testing temperature on the fracture toughness of PBT and RTPBT. At ambient, the K_C value of about 6 $MPam^{1/2}$ indicates that PBT is basically a semi-ductile material. The plastic deformation in the form of matrix tearing and shear yielding provided supportive evidence for the ductility of the PBT matrix(2). PBT-EAF and PBT-XLA showed the formation of large stable damage zone due to extensive plastic blunting at the crack tip. Thus in such cases the maximum force was calculated from the intercept of the 95% slope with the load-displacement curve, thereby K_Q values were derived instead of K_C. Perhaps a better approach to characterise the toughness of these materials is to utilise the J-Integral technique. At $-40^{\circ}C$, there is no significant changes in the K_C value of PBT. However, remarkable improvement in the toughness of PBT-EAF system can be observed in Fig.1. This may be attributed to the relaxation behaviour of the rubber particles as observed in dynamic mechanical thermal analysis (DMTA) study (3). When the temperature approaches the glass transition, T_g of the rubbery phase, the relaxation of the rubber becomes active and absorbs considerable amount of energy. As for the PBT-XLA, inspite of having a sub-ambient transition at about $-50^{\circ}C$, there is no significant change in K_C. This may be due to the presence of relatively rigid crosslinks which suppress the mobility of the macromolecules and consequently reduces the energy absorbing capabilities. The rather low K_C value of PBT-CSR may again be explained by considering the nature of the rubber particles. The CSR rubber which consisted of SAN is obviously rigid particles , thus it can be anticipated that at sub-ambient, the particles were only acting as a rigid particulate filler in PBT. Increasing the temperature to $80^{\circ}C$ has resulted in the reduction of all K_C values. The lowering of yield strength of PBT matrix above its T_g i.e. about $60^{\circ}C$ (3) combined with the dissappearance of the toughening effects of rubber particles

are the possible reasons for the observed trend. The presence of fibre-related micromechanisms such as fibre debonding and fibre pull-out (2) and increased strength associated with additional load-bearing capability of the fibres obviously resulted in the significant enhancement of K_C values shown in Fig. 2. In general, similar trend to that of unreinforced versions can still be observed as far as the effect of temperature is concerned.

Effect of strain rate Table 1 illustrates the effect of strain rates i.e. in the form of crosshead speeds on K_C for PBT, RTPBT and their composites. A drastic reduction in K_C value of PBT at high strain rate indicates that the time scale is too short for the plastic deformation process to takes place. Thus PBT fails in a brittle manner. As expected, significant improvement in fracture toughness was observed in the case of PBT-XLA and PBT-EAF at high strain rate. This is believed to arise from the rubber-related toughening mechanisms such as cavitation and shear yielding. SEM micrograph of PBT-EAF revealed the existence of extensive shear yielding and fibrillar structures on the fracture plane(2). A similar observation has been reported by Wu et.al, (4) for other rubber toughened PBT systems. In Table 1 it can be seen that the effect of incorporating short glass fibres into PBT and RTPBT matrices does not influence the toughness remarkably. This seems to suggest that at a high strain rate the fracture process of the materials is more or less dominated by matrix- and rubber-related toughening mechanisms. As before, PBT-EAF-GF exhibits the highest K_C value as compared to other PBT formulations. A sheathed fibre pull-out whereby the fibre surface well coated with a thick layer of matrix material has been observed (2).

Effect of hygrothermal ageing Table 2 illustrates the effect of hygorthermal ageing on the fracture properties of PBT, RTPBT and their composites. It is clear that the ageing process has resulted in a dramatic reduction of K_C values of all materials tested. It is known that under such an adverse condition the chemical interactions between water molecules and the ester groups has resulted in the hydrolysis of PBT. This will then leads to the lowering of the molecular mass of the PBT matrix. PBT with low molecular mass is prone to undergo brittle fracture. Detailed investigations on the hydrolysis of PBT and its blend have been reported by Golovay, et.al, (5). The appearance of the fracture plane which displayed a smooth surface with no evidence of plastic deformation has been reported by Ishak and Lim (6). The highest K_C value of PBT-EAF as compared to other types of RTPBT indicates that the extent of toughness deterioration is strongly controlled by the quality or the chemical nature of the impact modifiers. However, in general the toughening effect of the rubber particles appear to diminish due to the serious embrittlement and disintegration of the PBT matrix.

As for the PBT and RTPBT glass fibre composites, the higher K_C values of the later indirectly implies that the hydrolytic stability of PBT was improved through the combination of both short glass fibres and impact modifiers. Again, the highest retention in K_C is observed in the case of PBT-EAF composites. SEM micrograph has provided a clear evidence for an extraordinary good bonding between GF and the PBT-EAF matrix.

The fibre surface was well coated with a thick layer of matrix materials. On the contrary, a poor bonding quality was observed in the case of unmodified PBT composites (2).

Further investigation on the extent of deterioration of the fracture properties has been performed by drying the hygrothermally aged PBT, RTPBT and their composites in a vacuum oven at 100°C for 48h.. As expected, poor recovery was observed especially in the case of unreinforced PBT and RTPBT. The percentage recovery for all the materials tested is shown in Table 2. This clearly indicates that hydrolysis of PBT matrix has resulted in a permenant damage or irreversible deterioration of the fracture performance. This is in agreement with earlier works reported by other workers(2, 6). The superior retention and recovery of the RTPBT composites, especially that of PBT-EAF, again provides a clear indication that the hydrolytic stability of PBT was improved through the combination of short glass fibre reinforcement and impact modifiers. Failure mechanisms characterised in-situ and post-fracture using acoustic emmision and SEM techniques, respectively, have revealed that matrix-, rubber- and fibre-related mechanisms have been restored to some extent by redrying the hygrothermally aged materials (2).

CONCLUSIONS

The fracture mechanical performance of PBT is strongly influenced by both internal (types of impact modifiers and glass fibre loading) and external parameters (temperature, strain rate and hygrothermal ageing). The combination of short glass fibre and impact modifiers produced a synergistic effect on the fracture behaviour of PBT, especially when the materials were subjected to testing conditions of high strain rate and low temperature. The extent of toughness deterioration as a results of hydrolysis of PBT matrix via hygrothermal ageing can be improved by incorporating glass fibre and EAF impact modifier into PBT matrix.

REFERENCES

1. Schultz J M and Fakirov S; Eds Solid State Behaviour of Linear Polyesters and Polyamides, Prentice Hall, Eaglewood Cliffs, New Jersey, U.S.A., 1990.

2. Czigany T, Mohd Ishak Z A, Heitz T and Karger-Kocsis J, Poly. Comp. (in press).

3. Mohd Ishak Z A and Karger-Kocsis J, Technical Report, BASF AG, Ludwigshafen, Germany, 1995.

4. Wu J, Mai Y W and Cotterell B, J Mater. Sci., Vol. 28., 1993, pp.3373-3384..

5. Golovay A, Cheung M F and Van Oene H, Polym. Eng. Sci., Vol.28., 1988, pp. 200-207.

6. Mohd Ishak Z A and Lim N C, Polym. Eng. Sci., Vol.34., 1994, 1645-1655.

TABLE 1- Effect of Crosshead Speed on the Fracture Toughness of PBT, RTPBT and Their Composites.

Materials	K_C (MPam$^{1/2}$)	
	1 (mm/min)	1000 (mm/min)
PBT	5.9	2.2
PBT-XLA	3.7	5.2
PBT-CSR	3.3	1.9
PBT-EAF	4.2	5.1
PBT-GF	7.3	3.3
PBT-XLA-GF	5.8	4.5
PBT-CSR-GF	5.9	3.1
PBT-EAF-GF	6.0	6.7

TABLE 2- Effect of Hygrothermal Ageing on the Fracture Toughness of PBT, RTPBT and Their Composites

Materials	K_C (MPam$^{1/2}$)		
	Unaged	Aged at 90°C	Redried
PBT	5.9	0.4	0.5 (7.8)
PBT-XLA	3.7	0.4	0.5 (14.2)
PBT-CSR	3.3	0.6	0.5 (16.3)
PBT-EAF	4.2	0.7	0.7 (16.8)
PBT-GF	7.3	2.5	2.8 (38.0)
PBT-XLA-GF	5.8	2.9	3.2 (55.0)
PBT-CSR-GF	5.9	3.3	3.6 (60.9)
PBT-EAF-GF	6.0	3.2	3.8 (63.2)

( )% Recovery with respect to the unaged samples.

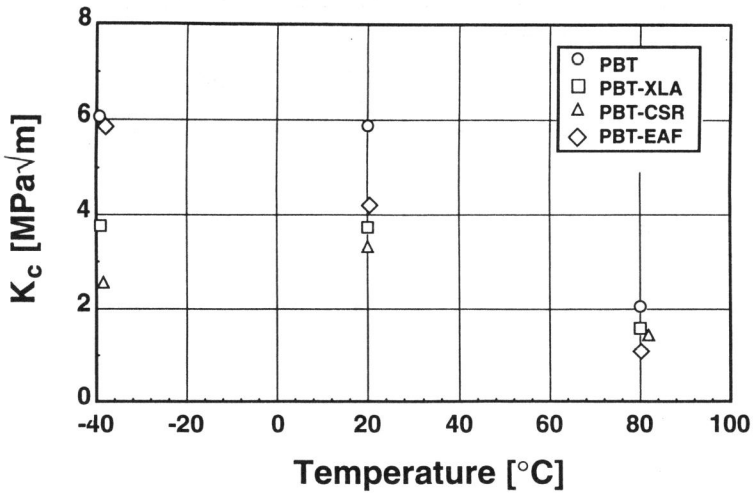

Figure 1 Effect of temperatures on the fracture toughness of PBT and RTPBT.

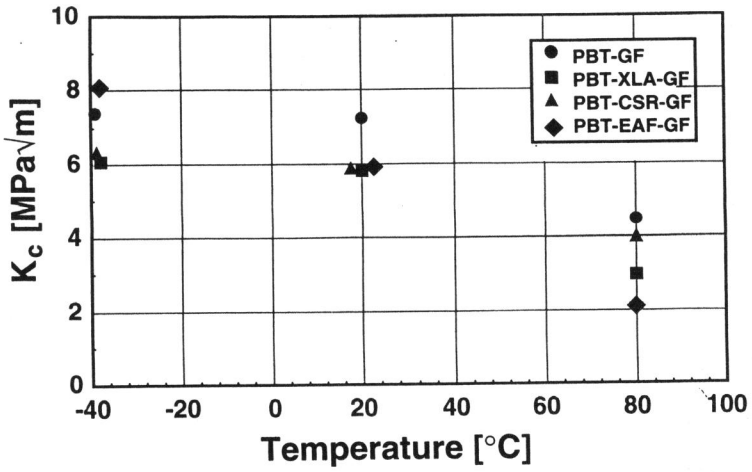

Figure 2 Effect of temperatures on the fracture toughness of glass fibre reinforced PBT and RTPBT.