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INFLUENCE OF FIBER-MATRIX ADHESION ON MECHANICAL PROPERTIES OF GLASS/POLYBUTYLENE TEREPHTHALATE UNIDIRECTIONAL COMPOSITES

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Abstract

Interface failure plays an important role in determining the mechanical properties of composites [1]. We studied the effect of matrix-fiber interfacial bonding on the transverse bending properties of glass fiber reinforced polybutylene terephthalate (G/PBT) unidirectional (UD) composites. Six types of specimens were manufactured using three different processing methods, namely reaction-based resin, prepreg and commingled yarn systems. Transverse bending properties of the UD composites were measured. Further tension failure zone after the transverse bending test was examined using scanning electron microscopy (SEM) fractography analysis. Additionally, the composites quality was evaluated using complementary microscopic techniques (optical microscopy, OM and SEM).

1 Introduction

Interface between reinforcing fiber and resin is a key factor in determining the mechanical properties of thermoplastic composites [1-2]. Microscopy was widely used to examine the fracture surface of composites to pave light into the nature of bonding at the matrix-fiber interface and information relating structure to mechanical properties [3-4]. Although the reinforcements mechanisms are similar, most of these studies were reported on epoxy based composites [5-6]. However, specific applications and complex structures require precise predictions of mechanical behavior with respect to a particular fiber-matrix combination. Transverse bending is widely used for the comparison and screening of different polymer systems [7]. Herein, we measured the transverse bending properties, visualized the fiber-matrix interfacial bonding of fractured specimen using SEM and evaluated the quality of glass fiber reinforced polybutylene terephthalate (G/PBT) composites.

2 Experimental

2.1 Materials

The cyclic butylene terephthalate (CBT160) supplied by Cyclics Corporation (USA) was in powder form at room temperature. Glass fiber rovings used with CBT160 in order to make a UD composite was supplied by PPG industries (USA) and Ahlstrom (Finland). The prepreg tapes received from Ticona has 60wt% glass fibres whereas the prepreg tapes supplied by Jonam has 63wt% glass fibres, with a 0° orientation in both cases. The commingled G/PBT systems are supplied by Owens corning (France) and Comfil (Denmark). The G/PBT system delivered by Owens corning has copolyester Twintex, 65% GF by weight.

2.2 Processing of G/PBT Composites

Unidirectional G/PBT composites were manufactured by vacuum consolidation technique using the three processing methods namely, commingled yarn, prepreg and reaction-based resin systems. G/PBT systems were processed by *in-situ* polymerization of powdered CBT (for reaction-based CBT resin) and PBT for prepreg and commingled yarn). The recommended process temperature for CBT and PBT used are 230°C and 240°C, respectively.

2.3 Transverse Bending

The tests were conducted using Zwick/Z100 testing machine at crosshead displacement control (3.3mm/min) at room temperature... During the test, load applied to the specimen and the deflections were recorded. The samples were loaded until the failure was complete. Further the fractured area was examined using SEM microscopy. The fixture used was custom build to fulfill the requirements specified in the standard for the specific sample size. The test setup with important dimensions is shown in Figure 1. The pins supporting and loading the test sample were not allowed to rotate freely during test. The G/PBT laminates were cut in to rectangular samples, based on ISO 14125 class III standards, with a dimensions $120 \times 15 \times 5$ (mm3). The thickness of samples was measured using vernier caliper which is in the range $4.5 - 5$ mm. The span, L, was adjusted to fit 100mm $\pm$ 0.2mm.

2.4 Microscopic Evaluations

The quality of the manufactured UD composite was evaluated (relatively) using several complementary microscopic methods on both cross sections perpendicular to the fiber orientation of polished specimen as well as fractured surface. The microscopy gives information on voids, delaminations, fiber distributions and matrix rich areas. The following evaluation methods are chosen to compare the composite systems considered in this article.

2.4. 1 Optical Microscopy (OM)

Reflected light micrographs of polished Glass/PBT specimens were obtained using Olympus BX 60 (Denmark) connected to Leica DFC320 camera. Leica IM50 (UK) software was used to capture the images. For sample cross-sectional analysis, specimens were grinded and polished with the following sequence of abrasive paper with grain sizes 500, 1200, and 4000 until obtaining a smooth surface. Samples were rinsed using de-ionized water during each step.

2.4. 2 Scanned Electron Microscopy (SEM)

The morphology of both polished cross sections and fractured surface (tensile tested) specimens were examined using Zeiss EVO 60 SEM with an electron source 10-25 keV in the secondary electron mode.

To reduce the extent of sample arching, both polished cross sections and fractured specimens were coated with a thin layer of metallic gold in an automatic sputter coated prior to examination by SEM. The sputter coater uses argon gas.

3 Results and Discussion

3.1 Mechanical Properties - Transverse bending

A series of glass fiber (GF) reinforced PBT thermoplastic unidirectional (UD) composites with GF volume fractions of 41-52 wt. % have been manufactured. The fiber content of the composites manufactured from commingled yarn was ca. 50 wt. % which allows the matrix to fully consolidate during vacuum consolidation. The fiber content for prepreg systems was slightly lower and mixed fiber volume fractions were used for the case of reaction-based resin systems. Depending on the glass fiber sizing formulation assigned by the supplier and the composite processing conditions; the composites were expected to have slightly different fiber-matrix interfacial properties and thereby mechanical properties. Here, we tried to unveil the correlation between transverse bending properties and composite quality factors governing the composite failure.

The flexural strength and modulus of G/PBT UD composites were measured. A summary of the test results are showed in Table 1. In most cases, the flexural modulus was higher for composites with higher fiber volume fractions. Both commingled yarn and reaction-based resin systems were reported higher flexural modulus values. However there was no direct relationship for flexural strength values with different fiber volume fractions. Figure 2 shows the typical (single data) stress-strain curves plotted using flexural tests results. It was observed that the composite responses can be linearly and non-linearly under transversal flexural test. Specimen, C1 exhibit a purely linear behavior, which is directly related to the brittle and sudden failure. This type of failure behavior may generally due to the presence of non-wetted fibers (clear fibers) and therefore poor interface bonding. On the other hand, specimen C2 exhibit a non-linear behavior (ductile) may due to good matrix-fiber bonding. For a few cases, the strain-stress curves exhibited some steps in the initial stages of curves (linear portion). This phenomenon might be due to step-wise (continuous)

damage process of the matrix and interface (debonding). Specimens, B2 and C2 with fiber volume fractions ca. 50 wt. %, showed better flexural properties.

3.2 Microscopic Observations

3.2.1 Cross-sectional analysis of polished G/PBT UD composites

Microscopy has been used widely for the evaluation of nature of adhesion between the fiber and resin [3-6]. Both optical microscope and SEM were used to analyze the distribution of fibers, degree of wetting, resin-rich areas and voids in the matrix. Figure 3 show optical micrographs (OM) of the cross-sections of polished specimens, A1 to C2 and their corresponding higher resolution SEM micrographs are showed as inset figures. Micrographs of reaction-based resin specimens, A1 and A2 revealed large areas of resin-rich regions and a few voids.

In specimen B1, arrangements of fibers as bundles with uniform fiber wetting and packing were observed. Micrographs of specimens C2, C1 and B2 exhibited good fiber distribution. However, specimen C1 showed several non-wetted areas (clear fibers) and voids in the composites. In both commingled (C1 and C2) and reaction-based resin (A1 and A2) specimens, non-uniform fiber wetting and voids were observed in a few areas whereas the adjacent areas exhibited good fiber wetting which may due to the uneven consolidation process conditions of the composites.

3.2.2 SEM fractography observations - Post failure analysis

Information relating structure to mechanical properties can be correlated by assessing SEM micrographs of the fractured composite area. Figure 4 show SEM micrographs of fractured surface around tension failure zone after the flexural test [8]. The reaction-based resin specimens, A1 and A2 exhibited a significant amount of resin around the fiber surface (Figure 4 A1 and A2) and many hackles on the fiber surface (green arrows). However, debonding of fibers observed in a few areas whereas no clear fibers present in the fractured specimens. Several matrix cracks (brittle behavior) were appeared along the resin-rich regions. In the failed specimen, C1 clear fibers (yellow arrows) were present with or without resin residue and have

observed many debonded fibers (Figure 4 C1). In specimens C2, B2 and B1, the individual glass fiber surface showed a thin layer of resin residue bonded to the fibers.

3.3 Influence of matrix-fiber interface bonding on the mechanical properties of G/PBT UD composites

Matrix-fiber interface bonding, fiber volume fraction and void content of a composite play an important role in determining the various mechanical strengths and weakness of composites [1]. Microscopic methods were able to examine the matrix-fiber interface of the fractured surface and to correlate with the mechanical properties of composite. Table 2, summarizes the main evaluation criteria and the observed matrix-interfacial behavior according to the SEM and OM micrographs of G/PBT composites. In summary, prepreg specimens (B1 and B2) exhibited better composite quality with the least void content whereas commingled specimens (C1 and C2) showed good fiber distribution. Based on microscopic evaluation three types of failure modes were observed, namely matrix failure, interface failure and a mixed mode of failure.

The reaction-based resin specimens, A1 and A2 revealed a significant amount of resin residue and many hackles the around the fiber surface (Figure 4, green arrows), indicating high level of adhesion. Therefore, mainly a matrix dominant mode of failure was inferred for these specimens. However in a few cases, fiber debonding were observed (Figure 4 B2, red arrow) revealed a mixed mode of failure. Both the specimens A1 and A2 exhibited cohesive failure at the resin-rich area [6]. The brittle failure observed at the resin-rich areas (Figure 4, A1 and A2) is in good agreement with the sudden failure as seen in stress-strain curves. Further, the steps appeared in the curves may due to the delamination or the initial failure of voids present in the composites.

It is well known that the void content of a composite may significantly affect its mechanical properties and therefore it is an important indicator for composite quality. Particularly, micrographs of specimen C1 showed many non-wetted areas (red arrows) and clear fibers (yellow arrows) which may the reason for the sudden failure as observed in stress-strain curves. Thus the fracture occurs mainly at the interface (interfacial failure) as inferred from

SEM fractography micrograph (Figure 4, C1). It is well-known that the fiber dominated properties like tensile and bending in the fiber direction are not very sensitive to matrix-fiber bond strength [1]. Here, we found a similar behavior as in the case of specimen, C1where the test was performed in fiber transverse direction. This supports their higher flexural modulus values for a fiber volume fraction of 51 wt. %.

Both prepreg specimens (B1 and B2) showed good fiber wetting with the least void content. The fractured areas of the specimen, B1 showed fiber bundles surrounded by resin rich phase as revealed in both fractography and cross-sectional micrographs, inferred a matrix failure mode. In specimens, B2 and C2 (figures 4), the individual glass fiber surface showed a thin layer of resin residue. Further, the stress-strain curves (Figure 2) showed a linear behavior which could be due to high fiber volume fraction and uniform fiber distribution in these specimens. Both the specimens exhibited a few hackles around the fiber and/or matrix areas and fiber debonded areas (Figures 4, B2 and C2) inferring a mixed mode of failure (matrix and interface failure). In summary, SEM micrographs (figure 4) inferred primarily a matrix dominant mode of failure for specimens, A1, A2 and B1; mixed mode of failure for specimens, B2 and C2 and interface failure mode for specimen, C1.

4 Summary

The mechanical properties of UD laminates of G/PBT were measured and tried to elucidate the correlation with respect to the interfacial bond strength, visualized using microscopy. Therefore we could be able to test the effects of interfacial bond strength on the transverse bending properties. In most cases, we found that specimens processed with higher fiber volume fractions follow the trend. Although, here the matrix-fiber interfacial bond strength is not significantly vulnerable to the mechanical properties of G/PBT UD composites, this study provides a better understanding of the relationships between processing, composite quality, fiber-matrix characteristics and thereby performance of composites.

Processing Method	Materials (G/PBT)	Fiber volume fraction, (V_f)	Flexural Modulus, E_f (GPa)	Flexural Strength, σ_u (MPa)
Reaction-based Resin (CBT 160)	A1	0.41	9.2 ± 1.3	49.8 ± 3.4
	A2	0.52	8.1 ± 1.1	45.3 ± 4.1
Prepreg Tapes	B1	0.46	5.9 ± 0.2	39.2 ± 3.5
	B2	0.49	7.8 ± 0.6	69.3 ± 5.1
Commingled Yarns	C1	0.51	8.4 ± 0.2	36.2 ± 2.5
	C2	0.50	7.1 ± 0.9	45.4 ± 1.8

Table.1. Summary of flexural test of G/PBT UD composites.

Processing Method	Materials (G/PBT)	Fiber volume fraction, (V_f)	SEM Observations		
			Clear Fibers	Hackles	Voids/non-wetted areas
Reaction-based Resin (CBT 160)	A1	0.41	No	Yes	A few
	A2	0.52	No	Yes	Yes
Prepreg Tapes	B1	0.46	No	Yes	No
	B2	0.49	No	Yes	Yes
Commingled Yarns	C1	0.51	Yes	No	Many
	C2	0.50	No	Yes	Yes

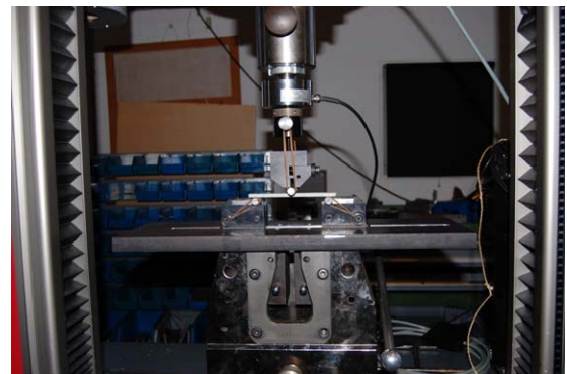Table.2. Summary of SEM observations of interfacial morphology of G/PBT UD composites.

Figure 1. Dimension sketch of fixture used for performing the three-point transverse bending l = 120[mm], h = 5[mm], L = 100[mm], R1 = 5[mm] and R2 = 5[mm].
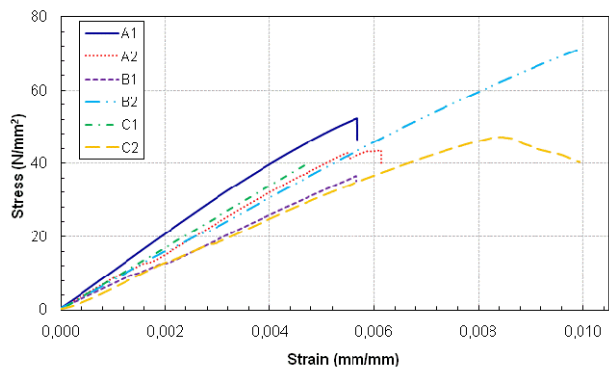
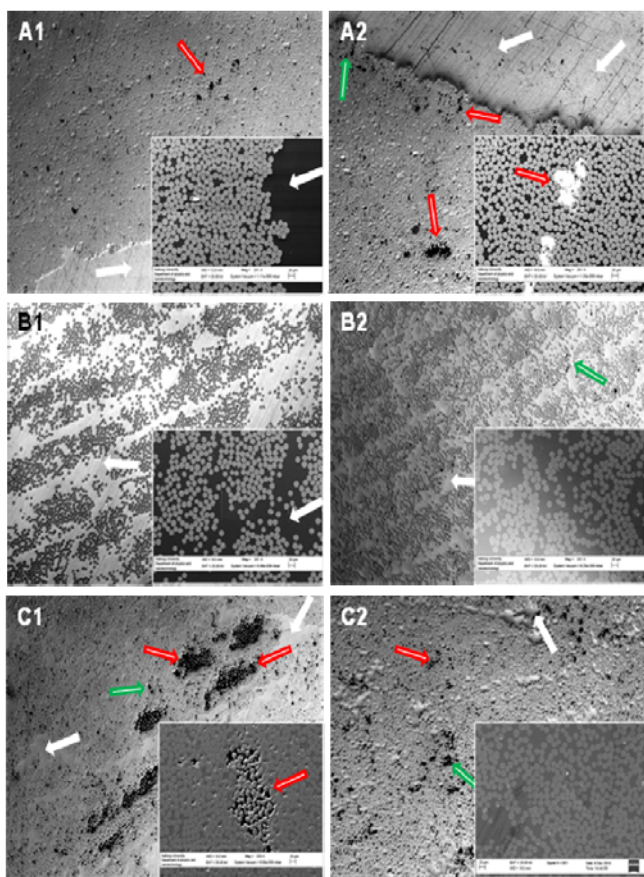Figure 2. G/PBT specimens: Typical (single data) stress-strain plot from flexural test.

Figure 3. Optical micrographs of cross-sections of polished specimens, A1, A2, B1, B2, C1 and C3, respectively. Corresponding high resolution SEM micrographs are showed in inset.
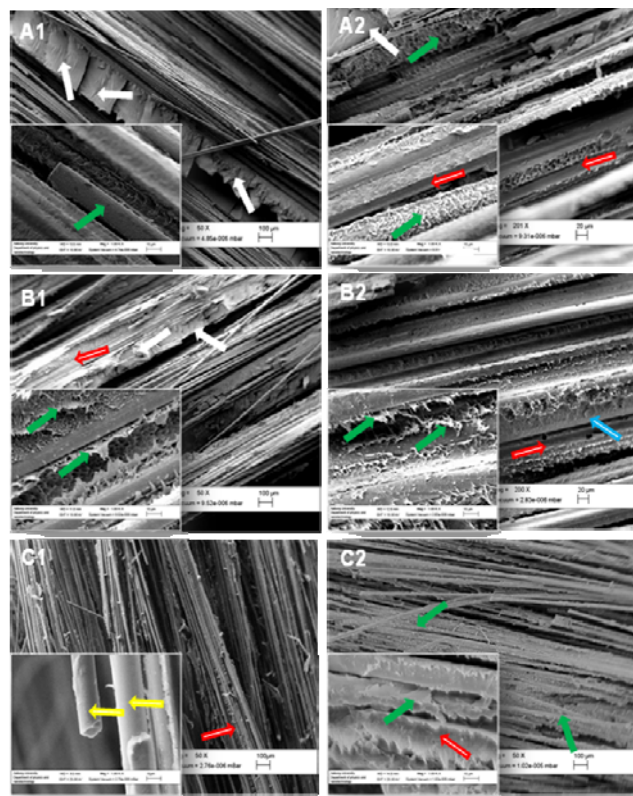

Figure 4. SEM fractography: Micrographs of fractured specimens, A1, A2, B1, B2, C1 and C3, respectively. Corresponding high resolution SEM micrographs are showed in inset.

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