FRACTURE BEHAVIOUR OF ALL-POLY(ETHYLENE TEREPHTHALATE) COMPOSITES

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Abstract
In this study all-poly(ethylene terephthalate) composites were developed and their (fracture)mechanical properties were investigated. Glycol modified poly(ethylene terephthalate) (PETG) and high tenacity PET multifilament were used as matrix material and reinforcement, respectively. The reinforcement was aligned in unidirectional (UD) and cross-ply (CP) manner and its content varied between 30...70 wt%. The composite plates were prepared via winding the reinforcing filament onto alumina plates and matrix film were placed in-betweens. This pre-product was consolidated by compression molding. The tensile mechanical properties and the density of the composite plates were determined. To characterize fracture behavior of the matrix material and the all-PET composites, Essential Work of Fracture (EWF) and J-integral methods were adopted. Based on the results one can state that the UD and CP reinforcement could enhance significantly the mechanical properties respect to PETG but in case of fracture toughness only the UD composite passed matrix value and CP composite remained much below.

1 Introduction
Among thermoplastic composites, self-reinforced polymer composites (SRPCs) are the most researched ones. The components of SRPCs belong to the same polymer family which ensures the easy and fully recyclability and further lower density than the usual most used glass fiber reinforced polymer composites. To produce SRPC sheets different methods can be used (hot compaction, consolidation of coextruded tapes, film-stacking), however, by creating suitable processing windows [1]. Over the recent years, large body of works has been devoted to study and develop polyethylene (PE) and polypropylene (PP) based self-reinforced composites with success. Since both polymers have low glass transition (T_g, below 0°C) and low melting temperature (T_m, 130-170°C) their use is limited for high temperature applications. Since poly(ethylene terephthalate) has higher T_g and T_m (~70 and ~260°C,

respectively), it is a suitable candidate in this case. To create polyethylene terephthalate based SRPC only a few studies are available so far [e.g. 2,3].
The goal of our study is to develop all-poly(ethylene terephthalate) composites and investigate their fracture and failure behavior. As matrix material glycol modified poly(ethylene terephthalate) was selected and used which ensured a wide processing window (~50°C). The SRPCs were produced by compression molding according to the film-stacking method by aligning the reinforcing fibers unidirectionally and in cross-ply manner. The reinforcing content was 30, 50 and 70 wt%. Beyond the usual mechanical characterizations (static tensile and dynamic falling weight impact) the fracture and failure behavior of the composites prepared was assessed by fracture mechanical methods.

2 Materials and testing methods

2.1 Materials and sample preparation

Highly stretched PET fibers were used as reinforcement. The reinforcing fiber has a diameter of 25,2±1,4 μm, Young´s modulus of 12,4±1,6 GPa and tensile strength of 930,5±70,2 MPa (measured on single fiber tests). The melting temperature of the fibers is 263°C. A commercial grade of amorphous glycol modified poly(ethylene terephthalate) (PETG) was used as the matrix material (Eastar Copolyester 6763, Eastman). 0.05 mm thickness films of PETG were prepared by extrusion film blowing.

2.2 Composite preparation

Composite plates with different nominal reinforcing contents (30, 50, 70 wt%) were prepared according to the film-stacking technique. The reinforcing filaments were wound on an alumina plate in unidirectional (UD) and cross-ply (CP) manner. Between the wound reinforcing layers matrix foil was placed and the layered package on both side was covered also by matrix films. Thereafter, this package was inserted in a preheated hot press (215°C) and hold for 5 min without pressure and then compression molded for 5 min with 5.3 MPa pressure. The composite was cooled (cooling rate 30°C/min) under pressure. The thickness of the resulting composite plates is between 1.7 and 1.9 mm. Table 1 lists the determined reinforcing content prior to compression molding. Note that a slight loss of material occurred during consolidation (in case of 30 and 50 wt% UD composites), so these values remained nominal values. Further on in the discussion they are referred to as 30, 50 and 70 wt%, but in the figures the correct nominal values are used.

Composite	Nominal reinforcing content [%]	Number of reinforcing layers [pieces]
PET-30-UD	32	7
PET-30-CP	30	3/3
PET-50-UD	53	7
PET-50-CP	52	4/4
PET-70-UD	71	7
PET-70-CP	72	4/4

Table 1. Nominal reinforcing content of the composites

2.3 Composite characterization

The density of the samples was determined in absolute ethanol at 23°C according to EN ISO 1183-1.
The consolidation and resulting matrix/reinforcing layer structure was inspected by reflection light microscopy (LM; Olympus BX51M) on polished cross sections of the composite sheets.
Tensile tests were performed on a Zwick Z020 type instrumented universal testing machine on a Shape 3 type dumbbell specimen (according to ISO 8256 standard) with 5 mm/min

loading rate. Tensile tests were performed at room temperature and at least five specimens were tested in all composites.

2.4 Fracture mechanical tests

Fracture characterization was carried out on mode I double edge-notched tensile specimens (DENT) cut from the plaques (nominal width W was 25 mm and nominal length S was 150 mm), at a crosshead speed of 10 mm/min.

As the matrix and all-PET composites did not present the same fracture behavior, different Fracture Mechanics approaches were adopted.

It is well established in the literature that if there is not a significant crack growth resistance, a good measure of toughness can be obtained from the value of the J-integral at initiation, J_{Ic}. The J-integral is conventionally defined as a path independent line integral for non-linear elastic materials and it can be used to characterize ductile fracture in polymers [4-5] and also materials that fail by cleavage. The J_c parameter [4, 6] is applicable to characterize quasi-brittle failure behavior (non-linear load-displacement curves with sharp load drop at the point of fracture) of specimens with a crack to depth ratio close to 0.5. In the present work, J_c was evaluated at the instability load point by calculating the fracture energy required to produce cleavage behavior of pre-cracked specimens having a crack depth to width ratio of $0.45 \leq a/W \leq 0.55$ as:

$$J_c = \frac{\eta U_{tot}}{B(W-a)} \tag{1}$$

Where U_{tot} is the overall fracture energy, *i.e.* the total area under the load-deflection curve, B specimen thickness, and η a geometry factor that for DENT specimens can be expressed as [7]

$$\eta = -0.06 + 5.99\left(\frac{a}{W}\right) - 7.42\left(\frac{a}{W}\right)^2 + 3.29\left(\frac{a}{W}\right)^3 . \tag{2}$$

The J-integral approach is a natural extension of Linear Elastic Fracture Mechanics and is very useful for semi-ductile fractures. The Essential Work of Fracture (EWF) is a better approach for polymer composites that exhibit completely ductile fracture. The aim of the EWF approach is to separate the work performed in the fracture process zone, W_e, from the total work of fracture, W_f, that in ductile polymers is often dominated by the work of plastic deformation, W_p.

$$W_f = W_e + W_p \tag{3}$$

The work performed in the fracture process zone, termed the essential work of fracture, is a quasi-material property only dependent on specimen thickness. The EWF method makes use of the fact that the essential work and the plastic work scale differently:

$$W_f = w_e l B + \beta w_p l^2 B \tag{4}$$

where B is the plate thickness, β is the shape factor, and w_e and w_p are the specific essential work of fracture and the specific non-essential work of fracture, respectively. By dividing W_f by the ligament area lB, the specific total work w_f can be expressed as:

$$w_f = w_e + \beta w_p l \tag{5}$$

If the entire specimen ligament deforms plastically before fracture initiation, then the specific essential work can be found by testing different ligament lengths and extrapolating the specific total work of fracture to zero ligament length. The EWF method has been extensively

applied to polymers. It delivers a single fracture parameter that is representative of crack propagation.

3 Results and discussion

3.1 Composite characterization

The density values of the manufactured composite and matrix are shown in Figure 1. It can well be seen that the density increases significantly with increasing nominal reinforcing fiber content up to 50 wt%, but above this value it does not increase further in case of CP and decreases for UD. This is due to the fact that the matrix material can not impregnate completely the reinforcing filaments when 70 wt% reinforcement is applied. Consequently, the entrapped air decreases the density especially in the case of UD composites.

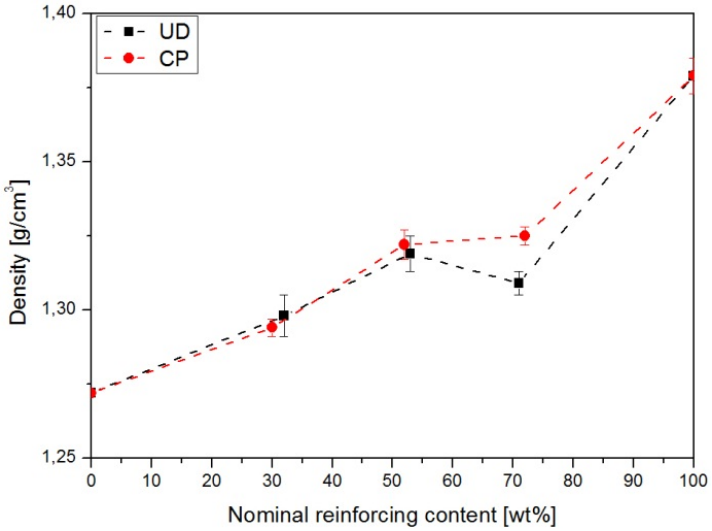
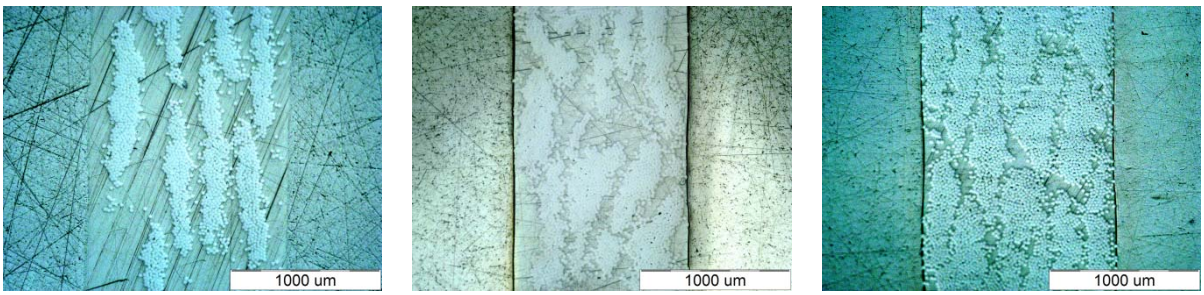


Figure 1. Density of the cross-ply and unidirectional composites as a function of reinforcing content. Note that the density at 100 wt% was determined on the reinforcing fibers

The poor consolidation is also observable on the light microscopic photographs taken from the polished cross section. Figure 2 shows the LM pictures of UD composites with different reinforcing contents.



a. b. c.

Figure 2. LM pictures taken from the polished cross section of UD composites having a reinforcing content of 30 (a), 50 (b) and 70 wt%

Tensile test results (tensile strength, tensile modulus and elongation at break values) are presented in Figure 3 along with their deviations.

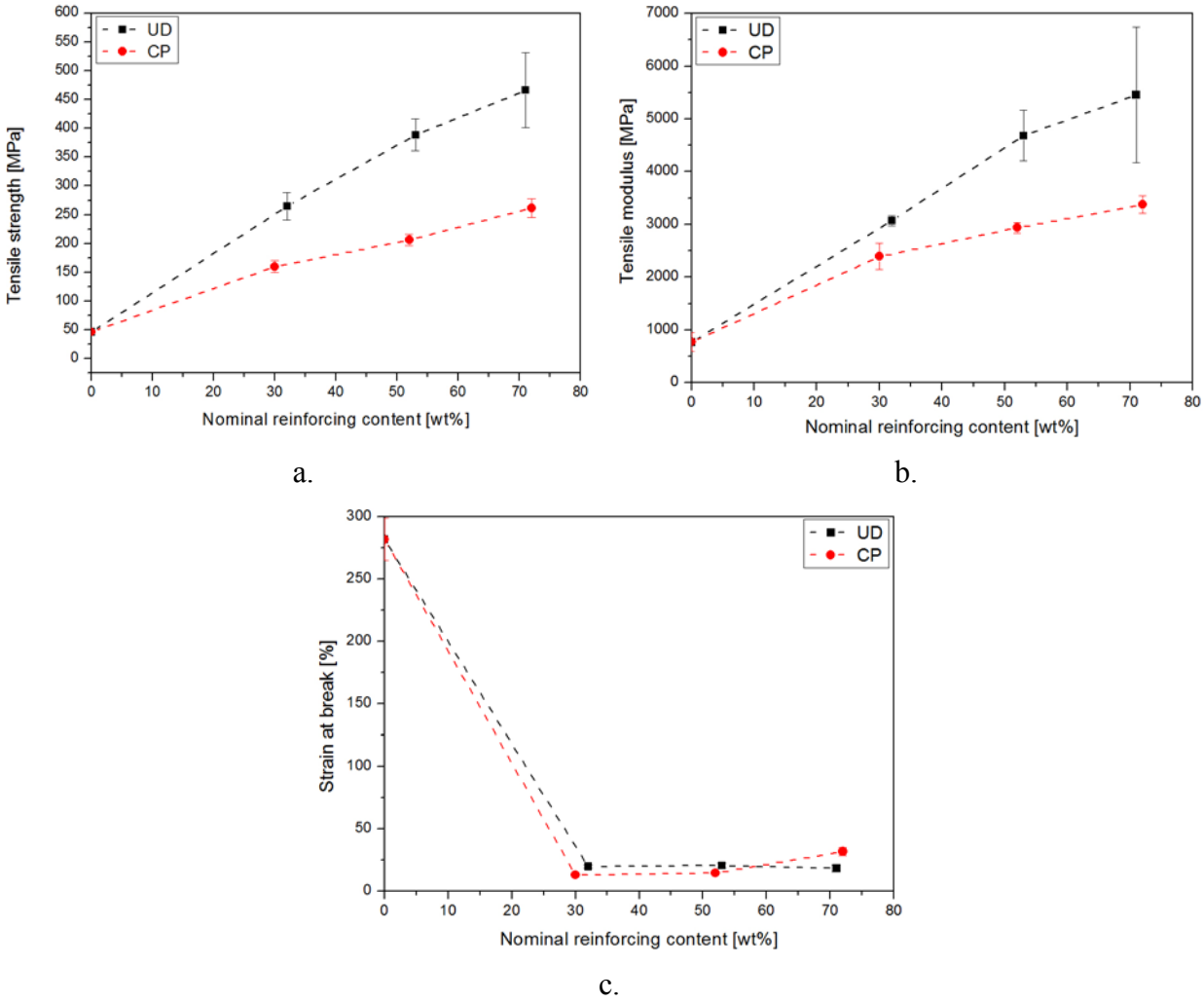


Figure 3. Tensile strength (a), tensile modulus (b) and elongation at break values for the cross-ply and unidirectional composites as a function of nominal reinforcing content.

Tensile strength and modulus values increase with increasing reinforcing content. The UD reinforced composites surpass the CP reinforced ones. The elongation at break decreases with increasing fiber content respect to the matrix value. In case of 70 wt% CP composites the strain at break value increased (comparing to 50 wt% CP composite), because the specimen showed yield-like failure behavior.

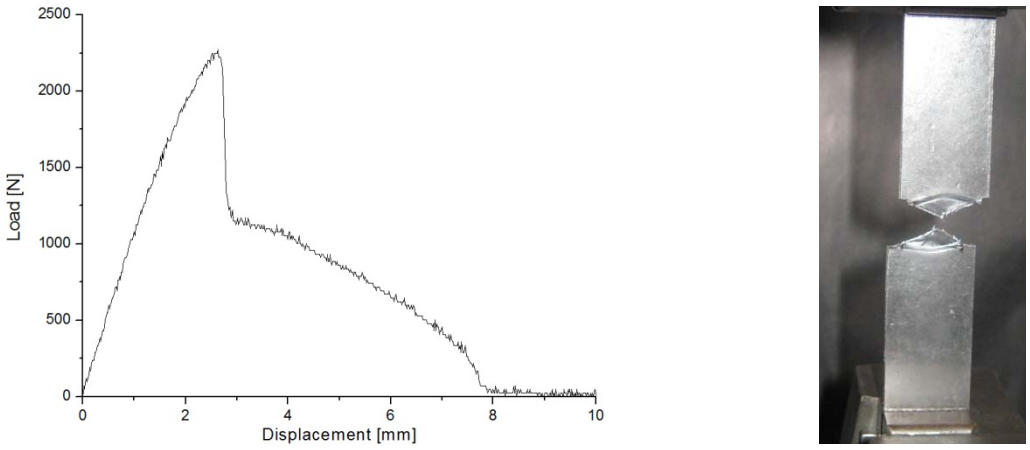
Figure 4 shows the macroscopic failure behavior of tensile specimens. In case of UD alignment the head of the dumbbell specimens (in the clamping) cleaved and this weaker part slipped out of the clamping. Therefore the specimens of 50 and 70 wt% UD composites were tabbed and the tensile parameters were determined these specimens.

a. b.

Figure 4. Typical macroscopic failure behavior of tensile specimens – 50 wt% UD (a) and 50 wt% CP (b)

3.2 Fracture tests

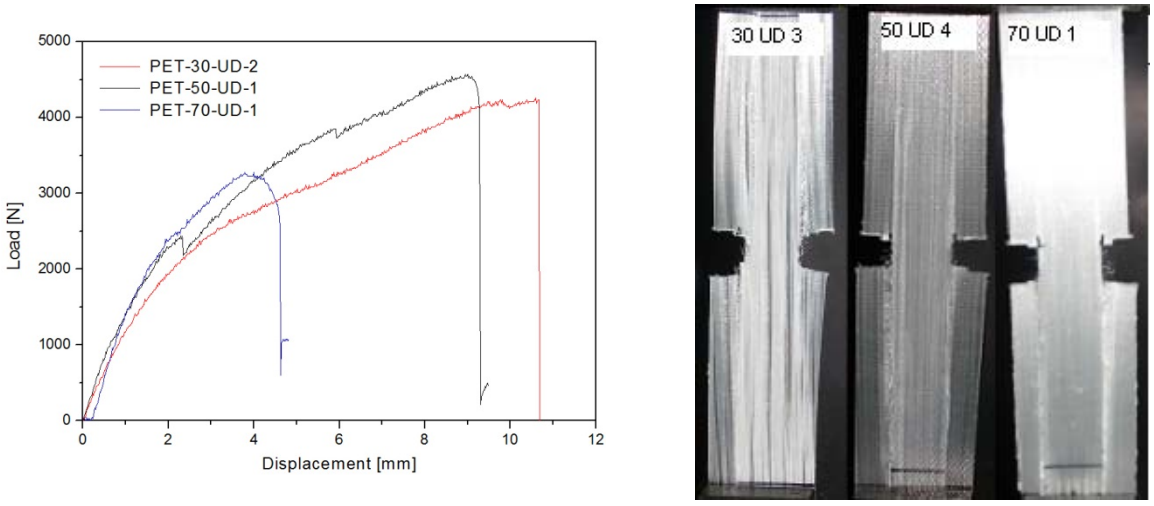
Figure 5-7 show typical load-displacement curves along with typical failure behavior for the matrix material and the different composites investigated, respectively. The matrix displays completely ductile fracture with fully yielded ligament before crack propagation, whereas all composites exhibit non-linear load-displacement records with unstable fracture at some point. Hence, the EWF method and the J_c evaluated at the instability are adopted to characterize the fracture behavior of the matrix and the composites, respectively.



a. b.

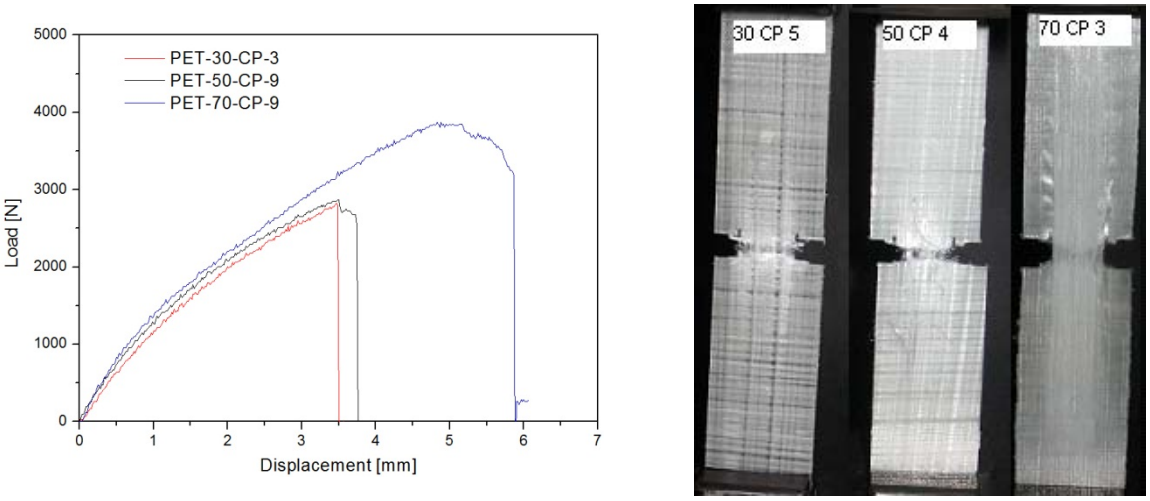
Figure 5. Load-displacement curves and macroscopic failure behavior for the matrix material

As a result of a non-uniform distribution of fibers in the unidirectional composites (Figure 7b), crack propagates along the longitudinal direction especially in the composite with the highest fiber content investigated (70 wt%).



a. b.

Figure 6. Load-displacement curves (a) and macroscopic failure behavior (b) for cross-ply PET composites



a. b.

Figure 7. Load-displacement curves (a) and macroscopic failure behavior (b) for unidirectional PET composites

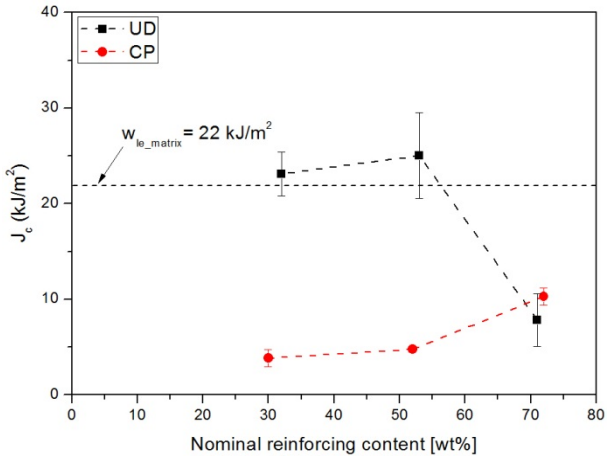


Figure 8. Toughness-reinforcing content curves for CP and UD all-PET composites

Finally, UD composites exhibit significantly higher values of fracture toughness than CP composites except in the case of the composite with 70 wt% as a result of the non-uniform distribution of fibers and the poorer matrix impregnation (Figure 8). However, only a slight increase in toughness is observed even for UD composites respect to the matrix initiation value (w_{Ie}). In the case of CP composites, on the other hand, an important improvement in J_c at instability is found for 70 wt% of fibers as a result of a more ductile fracture behavior. Fracture results are in good agreement with tensile tests results.

4 Conclusions

The tensile and fracture behavior of all-PET composites having unidirectional and cross-ply reinforcements was investigated.

It was found that tensile strength values increase with increasing reinforcing content for all compositions investigated, whereas tensile modulus increases only up to 50 wt%. The UD composites surpass the CP ones. On the other hand, elongation at break decreases with increasing fiber content for UD composites but increases for CP composites. The latter result is attributed to the yield-like failure behavior exhibited by the 70 wt% CP composite.

In fracture tests, the matrix material displays completely ductile behavior whereas the all-PET composites present crack instability at some point in the load-displacement curve. Therefore,

different fracture mechanical methodologies (EWF and J_c at instability) were adopted to characterize the materials fracture behavior.
Significantly higher values of fracture toughness are found for UD composites respect to CP composites except for the composite with 70 wt% of fibers. However, those composites do not exhibit any important increase in J_c at instability respect to the matrix initiation value.
CP composites exhibit the highest toughness value for 70 wt% of reinforcement due to the higher energy consumed by this composite during both crack initiation and propagation steps.
The results obtained in fracture tests are in good agreement with tensile tests results and also confirmed the poor impregnation of the fibers by the matrix material observed in density measurements for higher fiber contents.

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