Conference Paper

A Mechanical Analysis of *In Situ* Polymerized Poly(butylene terephthalate) Flax Fiber Reinforced Composites Produced by RTM

C. Romão,¹ C. M. C. Pereira,² and J. L. Esteves³

¹ Department of Mechanical Engineering and Industrial Management (DEMGI), School of Technology and Management, Polytechnic Institute of Viseu, 3504-510 Viseu, Portugal

² Institute of Mechanical Engineering and Industrial Management (INEGI), University of Porto, 4200-465 Porto, Portugal

³ Department of Mechanical Engineering (DEMec), Faculty of Engineering, University of Porto, 4200-465 Porto, Portugal

Correspondence should be addressed to C. Romão; romao@estv.ipv.pt

Received 16 September 2013; Accepted 18 November 2013

Academic Editors: R. Figueiro, H. Hong, and A. T. Marques

This Conference Paper is based on a presentation given by C. Romão at “International Conference on Natural Fibers—Sustainable Materials for Advanced Applications 2013” held from 9 June 2013 to 11 June 2013 in Guimarães, Portugal.

Copyright © 2013 C. Romão et al. This is an open access article distributed under the Creative Commons Attribution License, which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited.

This work addresses mechanical characterization in tension of woven flax fabric reinforced *in situ* polymerized poly(butylene terephthalate) composites, produced by the RTM technique. A brief description of the developed RTM set-up is made and the composite manufacturing details are presented. A morphological analysis of the mechanically characterized materials by Scanning Electronic Microscopy (SEM) is also made. The produced neat polymer (pCBT) showed a brittle behavior and mechanical properties lower than those found in the literature. Its reinforcement with woven flax fabric resulted in an enhancement of both tensile strength and stiffness. The obtained results can be significantly improved by the polymer modifying chemically, optimizing the control of the processing parameters, and subjecting flax fibers to a surface treatment compatible with the CBT 160 resin.

1. Introduction

Thermoplastic composites offer some interesting advantages over thermosets counterparts, such as higher toughness and impact resistance, recyclability, and faster production cycles. The use of vegetable fibers as reinforcement, replacing glass fibers, further increases the range of benefits: they are renewable, less expensive, and not abrasive and have lower density, specific properties (σ_r/ρ and E/ρ) that can be comparable to those of glass fibers (e.g., hemp and flax) and have lower environmental impact, since they are biodegradable and easily recyclable. The thermoplastic matrix selection needs, however, to take into account the vegetal fibers low thermal resistance and the processing technique that will be used. The cellulosic component of the fibers experiences a fast and irreversible degradation at temperatures about 200°C [1, 2].

The common thermoplastic processing techniques do not allow the combination of engineering or high performance thermoplastics with vegetal fibers due to its high melting temperature. Reactive processing of thermoplastics is a recent technique, currently in development and optimization, which makes use of mono- or oligomeric precursors that, after heated and mixed with an activator system, impregnate the fibers and polymerize *in-situ* to form the desired matrix. Due to its low molecular weight the precursors have extremely low melt viscosity (in order of mPa.s) allowing appropriate impregnation of short and continuous reinforcement at lower processing pressures and moderate temperatures (180 to 250°C for PA12 and PBT). The traditional liquid molding technologies of thermoset composites (like RTM, VARTM, SRIM, and RRIM) can be used to process this new generation of thermoplastic materials. Though, attending to the existing

FIGURE 1: CBT 160 resin and its polymerization to pCBT (polymerized CBT).

differences in the processing of these two types of polymeric materials, it is necessary to make adjustments in the traditional RTM equipment [3, 4].

This paper describes the mechanical behavior in tension of neat and flax fiber reinforced *in-situ* polymerized poly(butylene terephthalate), produced by the isothermal RTM technique, starting from the precursor Cyclic Butylene Terephthalate (CBT resin). A morphological analysis of the mechanically characterized materials by Scanning Electronic Microscopy (SEM) is also made.

The main objective was to investigate the possible compatibility between these two kinds of materials, both very sensitive to humidity. Reactive processing of vegetal fiber reinforced thermoplastics, as far as the authors know, had not been tested so far.

2. Materials and Methods

2.1. Materials. The polymeric precursor used in this study is the one-component CBT 160 resin, a Cyclic Butylene Terephthalate oligomer mixture that already contains the catalyst (Figure 1), supplied by Cyclics Corporation (Schwarzheide, Germany). It is reinforced with nontreated flax woven fabric, supplied by Composites Evolution (Chesterfield, United Kingdom), suitable for processing by traditional liquid molding techniques such as hand lay-up, vacuum infusion, and RTM. The woven fabric type is 4×4 Hopsack, based on 250 tex yarns, with 7 warp ends/cm, 11 weft picks/cm, and a surface weight of 510 g/m² (Figure 2).

2.2. RTM Set-Up and Composite Manufacturing Details. The developed RTM set-up consists mainly of a heated mold, a system for melting the resin under nitrogen atmosphere

FIGURE 2: Biotex Flax 4 × 4 Hopsack, 510 g/m².

(fusion system), a heated system to inject the resin into the fiber bed, and three different temperature control units connected to each of these thermal systems (Figure 3). The CBT 160 resin was heated under a nitrogen atmosphere to 190°C (approx.) and then injected under a nitrogen pressure of 2–50 kPa into the closed mold, at the same temperature. Once the mold was completely filled, the temperature was maintained for 30 min, in order to complete the polymerization reaction and to allow for cold crystallization. In principle, the part could be demolded at this temperature. However, since demolding at such high temperatures is rather troublesome with the current mold set-up, the part was allowed to cool before demolding. Before processing, CBT 160 was dried at 80°C for 2 h. Composites were produced using a 32% volume fraction of 4 × 4 Hopsack woven flax fabric undried and dried at 80°C for 24 h.

2.3. Mechanical Properties. Tensile tests were performed according to ISO 527-4, on a universal INSTRON 3367

FIGURE 3: The developed RTM set-up.

FIGURE 4: Pure pCBT (a) and flax reinforced pCBT (b) plates produced by RTM.

FIGURE 5: Typical tensile stress—strain plots for pure and reinforced pCBT.

machine equipped with a 30 kN load cell and an extensometer with a gauge length of 25 mm. The test speed was 2 mm/min.

2.4. Morphological Analysis. The morphology of the developed materials was analyzed by SEM, using a scanning electron microscope with integrated X-ray microanalysis: FEI QUANTA 400 FEG ESEM/EDAX PEGASUS.

3. Results and Discussion

3.1. Production. Flat plates ($100 \times 100 \times 4 \text{ mm}^3$) of neat and reinforced pCBT (polymerized Cyclic Butylene Terephthalate) were successfully produced using the RTM set-up and the production method described previously (Figure 4). The neat pCBT plates exhibit a higher mold shrinkage, during which fissuration also took place. The incorporation of the reinforcement reduced the matrix shrinkage; however, the presence of microcracks randomly distributed on the surface of the produced plates was observed. The presence of microcracks is also reported by other researchers for glass fiber reinforcements [5].

3.2. Mechanical Properties. Both neat and reinforced pCBT (polymerized Cyclic Butylene Terephthalate) were mechanically characterized. The results from the tensile tests are shown in Figure 5 and Table 1.

FIGURE 6: SEM micrographs of neat pCBT (a) and flax reinforced pCBT ((b), (c), and (d)).

TABLE 1: Uniaxial tension test results.

Samples		σ_t [MPa]	E_t [GPa]
Neat pCBT		15.0 ± 1.0	2.48 ± 0.26
pCBT/flax	Undried fibers	20.0 ± 0.5	3.89 ± 0.57
pCBT/flax	Dried fibers	26.1 ± 1.4	3.46 ± 0.91

Analyzing the results presented above, it is possible to conclude the following. (i) Neat pCBT isothermally processed at 190°C is brittle and has a tensile strength lower than that obtained by Mohd Ishak et al. [6] and Miller [7]. The pCBT brittleness has been already reported by several authors that investigate physical and/or chemical modification methods of pCBT [8, 9]. (ii) The reinforcement of pCBT with woven flax fabric results in an enhancement of both tensile strength and stiffness (iii) The use of dried fibers improves substantially the tensile strength of the composites.

3.3. Morphological Analysis. SEM micrographs are provided in Figure 6. Its analysis revealed (i) the presence of CBT oligomer crystals in both neat pCBT (Figure 6(a)) and reinforced pCBT (Figure 6(b)) samples; according to Abt et al. [9] these crystals act as a rigid filler and contribute to the

brittleness of unmodified pCBT; the amount of CBT crystals apparently exceeds the equilibrium content (usually between 1 and 3%) which may have contributed to an increase in pCBT brittleness and to the decrease in its characteristic tensile properties; (ii) the presence of porosities in both neat (Figure 6(a)) and flax reinforced pCBT (Figure 6(c)) that influence, also, the mechanical properties; (iii) a low adhesion between the fibers and the matrix in most of the observed samples that can be improved by performing surface treatments on flax fibers.

4. Conclusions

The developed RTM production system enabled the successful production of the materials with the desired geometry. Notwithstanding, the neat polymer showed a brittle behavior and lower mechanical properties than those found in the literature. The analysis of the process and the results allowed concluding that neat pCBT mechanical properties can be further improved by optimizing the control of the processing parameters (pressure and temperature) and modifying chemically the pCBT polymer in order to decrease its brittleness. The composites exhibited better mechanical properties than those of pure polymer, although a weak

adhesion fibers/matrix that can be improved by subjecting flax fibers to a surface treatment compatible with the CBT 160 resin has been observed.

Acknowledgment

The authors gratefully acknowledge the funding by Ministério da Ciência, Tecnologia e Ensino Superior, FCT, Portugal, under the SFRH/BD/40522/2007 grant.

References

- [1] C. Romão, *Estudo do Comportamento Mecânico de Materiais Compósitos de Matriz Polimérica Reforçados com Fibras Naturais*, Tese de Mestrado, Universidade do Porto, 2003.
- [2] C. Bailie, *Green Composites: Polymer Composites and the Environment*, Woodhead, Canada, 2004.
- [3] H. Parton, *Characterisation of the in-situ polymerisation production process for continuous fibre reinforced thermoplastics [Ph.D. thesis]*, Katholieke Universiteit Leuven, 2006.
- [4] K. van Rijswijk and H. E. N. Bersee, "Reactive processing of textile fiber-reinforced thermoplastic composites—an overview," *Composites A*, vol. 38, no. 3, pp. 666–681, 2007.
- [5] R. T. D. Prabhakaran, T. L. Andersen, and A. Lystrup, "Glass/CBT laminate processing and quality aspects," in *Proceedings of the 10th International Conference on Flow Processes in Composite Materials (FPCM '10)*, Monte Verità, Switzerland, 2010.
- [6] Z. A. Mohd Ishak, Y. W. Leong, M. Steeg, and J. Karger-Kocsis, "Mechanical properties of woven glass fabric reinforced *in situ* polymerized poly(butylene terephthalate) composites," *Composites Science and Technology*, vol. 67, no. 3-4, pp. 390–398, 2007.
- [7] S. Miller, *Macrocyclic polymers from cyclic oligomers of poly(butylene terephthalate) [Ph.D. thesis]*, UMI, 1998.
- [8] J. Baets, A. Godara, J. Devaux, and I. Verpoest, "Toughening of isothermally polymerized cyclic butylene terephthalate for use in composites," *Polymer Degradation and Stability*, vol. 95, no. 3, pp. 346–352, 2010.
- [9] T. Abt, M. Sánchez-Soto, and A. Martínez de Ilarduya, "Toughening of *in situ* polymerized cyclic butylene terephthalate by chain extension with a bifunctional epoxy resin," *European Polymer Journal*, vol. 48, no. 1, pp. 163–171, 2012.

The Scientific World Journal

Hindawi Publishing Corporation
<http://www.hindawi.com>

Volume 2014

Hindawi

- ▶ Impact Factor **1.730**
- ▶ **28 Days** Fast Track Peer Review
- ▶ All Subject Areas of Science
- ▶ Submit at <http://www.tswj.com>