NATURAL FABRICS AND BIODEGRADABLE POLYMERS FOR THE MANUFACTURE OF ENVIRONMENTALLY FRIENDLY COMPOSITE MATERIALS

I. Bavasso[1], J. Tirillò[1], P. Russo[2], V. Fiore[3] and F. Sarasini[1]

[1] Department of Chemical Engineering Materials Environment & UdR INSTM, Sapienza-Università di Roma, Via Eudossiana 18, 00184 Roma, Italy, www.uniroma1.it

[2] Institute for Polymers, Composites and Biomaterials, National Research Council, Pozzuoli, Italy, www.cnr.it

[3] Department of Engineering, University of Palermo, Viale delle Scienze, 90128 Palermo, Italy, www.unipa.it

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ABSTRACT

An experimental investigation has been carried out to evaluate the effect of eight plies of flax and jute fabrics on quasi-static (tensile and flexural) and impact properties of poly (lactic acid)/poly (butylene adipate-co-terephthalate) (PLA/PBAT) laminates. The composites and the neat matrix were tested under low velocity impact events at different energy levels (2 J and 6 J) and temperature conditions (25 °C and -50 °C). The results demonstrated that the natural fabrics enhance the load bearing capability of PLA/PBAT blend: at 2 J and 25 °C in both composites an increase in the peak force of about 127 % (flax) and 105 % (jute) with respect to neat blend was recorded. In addition, at 6 J no perforation of composites was observed contrary to what obtained for PLA/PBAT specimens at both investigated temperatures. Quasi-static tests revealed that the introduction of flax and jute fabrics contributed to an increase in the tensile and flexural strength of more than five and three times compared to the mechanical properties of the neat blend, respectively.

1 INTRODUCTION

The challenge we are currently facing is the replacement of conventional polymer materials with biodegradable products that minimize waste disposal from post-consumer materials. In this context, Poly (lactic acid) (PLA) is an example of a biodegradable polymer in the family of aliphatic polyesters and, thanks to the research on polymerization synthesis, the production of PLA is economically competitive with respect to traditional polymers but its application is limited by the low toughness, low impact resistance, low thermal stability and poor crystallinity [1]. For this reason, this polymer is usually blended with other polymers to balance such limits, and, among biodegradable thermoplastics, poly (butylene adipate-co-terephthalate) (PBAT) seems to be a valid candidate [2-3]. It is a semi-aromatic copolyester obtained from the condensation of 1-4 butandiol adipic acid and terephthalic acid derived from petroleum products but resulting in a completely biodegradable product. It features high ductility and toughness [4].

It should be noted that the biodegradability of these polymers has a slow rate and, for this reason, it is necessary to improve their mechanical properties to make them efficient for a long-life cycle.

A good strategy could be the use of natural materials (flax [5,6], sisal [7], hemp [8], jute [9], kenaf [10]) to improve the mechanical properties of biopolymers while maintaining biodegradable nature in a cost-effective product [11-12]. Although quasi-static mechanical properties of biodegradable composites are well addressed in literature, a limited number of papers discuss the impact properties as Charpy [6,13] and Izod [14] impact test results. Low velocity impact test through drop-weight impact represents a more realistic impact event, which is crucial for the prediction of damage propagation and composites failure [15], and only a few papers refer to this topic [16-18].

In this work, flax and jute fabrics were used for the manufacture of PLA/PBAT/FLAX and PLA/PBAT/JUTE composite laminates. A preliminary investigation of mechanical properties (tensile, flexural and low velocity impact tests) and thermal stability (thermogravimetric analysis) of both composite materials is proposed and results were compared with those of the neat polymer blend.

2 MATERIALS

Commercial PLA/PBAT blend with a formulation of 20 wt.% and 80 wt.%, respectively, and natural fabrics as flax (areal density of 220 g/m^2) and jute (areal density of 290 g/m^2) were used for the manufacture of composite laminates. The laminates were produced by hot compression moulding by using a P400E moulding machine (Collin GmbH) and applying the film stacking technique. In particular, for each laminate, eight plies of fabric were interleaved with polymeric films produced with a Teach-Line E20T flat die extruder equipped with a CR72T calender (Collin GmbH). Compression moulding was carried out at 180 °C under the following pressure profile: 1 bar-5 bar-10 bar-15 bar-20 bar-25 bar-30 bar (each step lasting 2 min). The laminates were cooled at room temperature under a pressure of 40 bar.

3 METHODS

Low Velocity Impact (LVI) tests were conducted at two different energy levels (2 J and 6 J) and temperatures (25 °C and -50 °C) using a drop-weight impact testing machine (CEAST/Instron 9340) equipped with a hemispherical tip (diameter of 12.7 mm). Before the impact tests the samples were conditioned at the selected temperature for 60 min. The kinetic impact energy was modified by varying the height of the weight (3.055 kg) and the specimens were clamped between two steel plates with a circular unsupported area with a diameter of 40 mm. All tests were conducted in triplicate.

The damaged area has been inspected by a non-contact laser profilometer (Talyscan 150, Taylor Hobson) for the measurement of the dent depth of specimens with a scanning speed of 8500 μm/s. In addition, mechanical properties such as tensile (in displacement control with a crosshead speed of 10 mm/min at room temperature and l_0 = 30 mm), flexural (three-point bending tests with a crosshead speed of 5 mm/min) were addressed. The thermal stability of composites was also investigated and compared to the neat polymer blend (by heating the samples from room temperature to 800 °C under nitrogen atmosphere and with 10 °C/min as heating rate; Setsys Evolution system by Setaram). Fracture surface morphology was investigated by scanning electron microscopy (FE-SEM Mira3 by Tescan).

4 RESULTS AND DISCUSSION

Low velocity impact tests at two energy levels (2 J and 6 J) and two temperatures (25 °C and -50 °C) were conducted on PLA/PBAT, PLA/PBAT/FLAX and PLA/PBAT/JUTE laminates and results are summarized in Figure 1 and table 1. In Figure 1 the evolution of force – displacement curves recorded at 2 J and 25 °C (Fig.1a) and -50 °C (Fig.1b) are shown and it is possible to notice that flax and jute composites revealed an effective strengthening effect on PLA/PBAT reaching the highest peak force values. PLA/PBAT/FLAX and PLA/PBAT/JUTE laminates exhibited a similar initial linear elastic behaviour and final unloading trend after the achievement of the peak force at lower displacement values with respect to PLA/PBAT because the occurrence of a transition from a ductile nature of PLA/PBAT to a stiffer material when flax and jute fabrics are stacked in the laminate. At 25 °C, such improvement in load bearing capability of PLA/PBAT could be quantified as the increase in peak force of about 105 % in the case of jute and an additional increase of 12% in the case of flax fabrics with reference to jute composite. Similar results were observed during the impact at 6 J where the increase in the peak force was about 51% for PLA/PBAT/JUTE and 128% for PLA/PBAT/FLAX compared to neat PLA/PBAT. Although the different fibre volume fractions in the final laminate (0.27 and 0.33 for flax and jute, respectively), the major difference in impact resistance can be attributed to the superior mechanical properties of flax fibres to jute fibres [19]. It is possible to exclude any degradation effect due to composite manufacture as the maximum processing temperature adopted in this work was 180 °C: thermogravimetric analysis revealed that the laminates' degradation starts to be significant at temperatures above 180 °C, as supported by the onset degradation temperature (defined as the temperature at 5 % of weight loss) values equal to 342.1 °C (PLA/PBAT), 297.3 °C (PLA/PBAT/FLAX) and 293.2 °C (PLA/PBAT/JUTE). The reduction of the onset temperatures of composite specimens suggests a slight loss in thermal stability with respect to PLA/PBAT and this is ascribed to the partial replacement of the more stable polymer matrix with the less thermally stable hemicellulose and cellulose components from flax and jute [8,20].

The aforementioned difference in impact resistance between PLA/PBAT/FLAX and PLA/PBAT/JUTE is emphasized when composites become more brittle at -50 °C, a temperature lower than PLA and PBAT glass transition temperatures [21] (Fig.1b and Table 1). A survey on permanent indentation (Table 1) revealed the occurrence of PLA/PBAT penetration at 6 J and at both adopted temperatures while, at the same energy level, a permanent indentation of about 772 µm (impact at 25 °C) and 1236 µm (-50 °C) was measured for PLA/PBAT/JUTE laminate. The flax-based composites featured a significant decrease in permanent indentation extent in both cases.

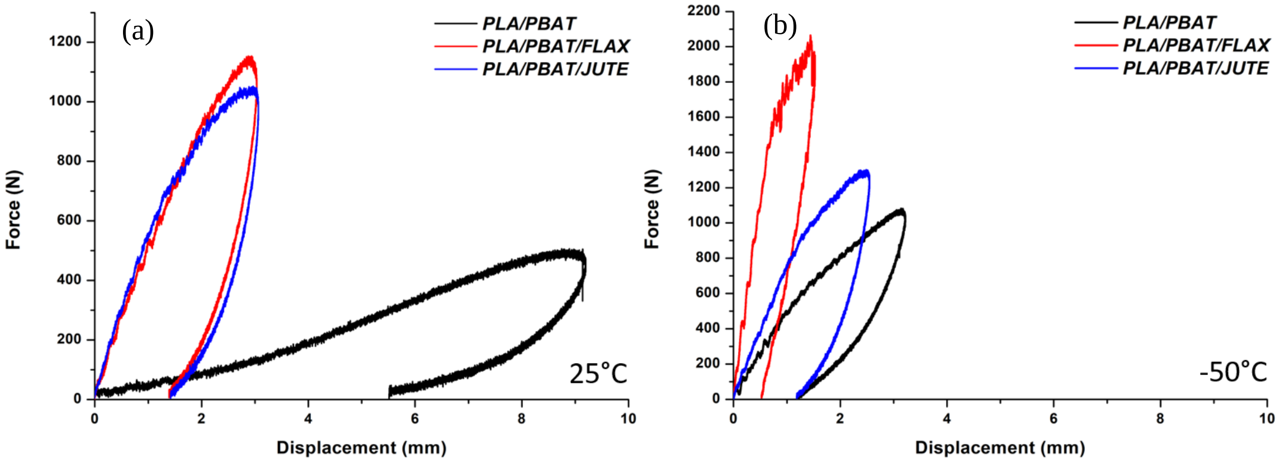

Figure 1: LVI results: force vs. displacement curves at (a) 25 °C and (b) -50 °C for samples impacted at 2 J.

Sample	Impact temperature [°C]	Peak Force [N]	Dent Depth [µm]
PLA/PBAT		676.2 (±1.1)	-
PLA/PBAT/FLAX	25	1540.4 (±30.0)	614.8 (±44.5)
PLA/PBAT/JUTE		1376.9 (±8.2)	722.0 (±76.8)
PLA/PBAT		1364.0 (±268.3)	-
PLA/PBAT/FLAX	-50	2585.9 (±108.3)	145.2 (±54.7)
PLA/PBAT/JUTE		1540.3 (±69.0)	1236.0 (±49.5)

Table 1: Peak force and dent depth of the laminates after LVI tests at 6J.

To evaluate the effect of flax and jute fabrics on the quasi-static mechanical performances of laminates, tensile and flexural tests were conducted and results are summarized in Table 2. Samples with flax and jute fabrics showed to be significantly more performant in both tensile and flexural properties than PLA/PBAT. In addition, PLA/PBAT/FLAX has demonstrated higher tensile and flexural strengths of about 86% and 58%, respectively, with a substantially higher modulus than PLA/PBAT/JUTE. These differences can be explained by considering the composition of the natural materials. Flax has higher cellulose content (≈81%) than jute (≈72%) [22] and seems to have better interfacial adhesion with polymers such as PLA [23]. In this experimental study, even to a slight extent, the compatibility between the fibres and the polymer matrix was pointed out through micrographs in Figure 2, where the mild interfacial adhesion between the flax (Fig.2a) and the polymer matrix is highlighted by the presence of polymer anchor points to fibre surface (white arrows), while an almost clean surface of jute fibre (Fig.2b) is visible and the presence of trace generated by the single-fibre pull-out.

Sample	Tensile strength [MPa]	Young's modulus [GPa]	Flexural strength [MPa]	Flexural modulus [GPa]
PLA/PBAT	10.4 (±1.9)	0.13 (±0.06)	8.5 (±0.2)	0.2 (±0.01)
PLA/PBAT/FLAX	56.6 (±2.3)	4.2 (±0.1)	41.9 (±0.6)	3.4 (±0.1)
PLA/PBAT/JUTE	30.5 (±3.0)	1.8 (±0.2)	26.6 (±0.9)	1.8 (±0.03)

Table 2: Quasi-static mechanical properties.

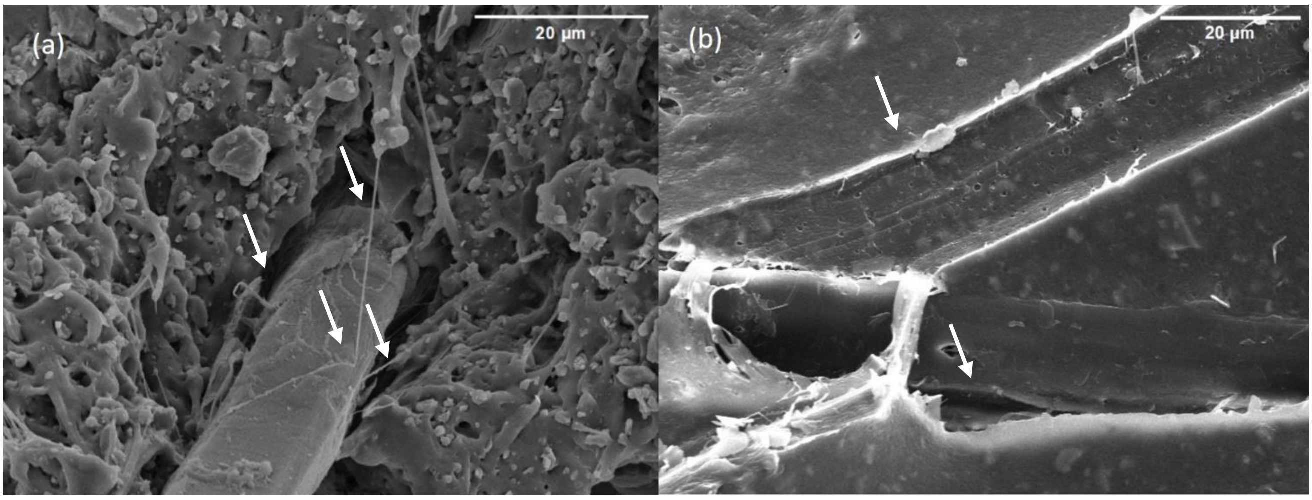

Figure 2: SEM micrographs of fracture surface of (a) PLA/PBAT/FLAX and (b) PLA/PBAT/JUTE.

5 CONCLUSION

This work aimed to demonstrate the possibility to enhance the mechanical performances of a biodegradable polymer blend PLA/PBAT with flax and jute fabrics. The composites were manufactured through hot compression moulding by stacking eight plies of fabric with polymeric films. The specimens were tested under low velocity impact events at different impact energy levels (2 J and 6 J) and temperature conditions (25 °C and -50 °C). The quasi-static mechanical properties were evaluated through tensile and bending tests. The composites showed a higher attitude to withstand the impact energy up to 6 J without perforation at both investigated temperatures with respect to PLA/PBAT. In addition, PLA/PBAT/FLAX and PLA/PBAT/JUTE have exhibited higher quasi-static mechanical properties such as tensile and flexural strength and modulus with respect to the neat blend. From the comparison between the two composites, PLA/PBAT/FLAX was superior to PLA/PBAT/JUTE and this result can be ascribed to both cellulose content and fibre mechanical properties jointly with the slightly better interfacial adhesion between flax fibres and the polymer matrix.

Such results highlight the feasibility of manufacturing composite materials with good mechanical performances without altering the biodegradable attitude of polymers. Possible modifications of the natural fabrics could be considered to ameliorate the compatibility between the polymers and the fabrics.

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