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Thermoforming–process of biopolymer composites

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This work aims to study the behavior of two dimensional composites (2-D composites) during a thermoforming process. Polylactide acid (PLA) films were prepared in a hot press at a temperature of 155˚ C and pressure of 90 kN/m^2. The obtained PLA-films were used as a base for further composite materials synthesis. Various glass fiber types and viscose rayon were used in order to improve the composite behavior. Those materials were further processed to final product packaging, in KIV Kreis GmbH via vacuum thermoforming. The mechanical properties were tested by means of tensile strength, tensile modulus, three point bending and Charpy impact strength. The morphological properties of the composite materials were investigated by digital microscope.

Keywords: *PLA, Viscose rayon, Glass fiber, Thermoforming process*

1. INTRODUCTION

Plastic thermoforming is one of the fastest growing methods of producing plastic packaging. During the thermoforming process, the polymeric sheet is heated to the so-called forming temperature. This temperature depends on the different type of thermoplastic material. The polymer sheet is supple and elastic plastic deformable at the forming temperature [1]. Thermoforming is widely used in manufacturing industries to produce large and labor intensive products. Compared to other manufacturing techniques, thermoforming is an extremely efficient process that is suitable for high-efficiency mass production [2].

PLA is biodegradable and biocompatible polyester, which can be produced from renewable sources [3, 4]. Due to new techniques which allow economical production of high molecular weight PLA, this polymer has attracted great attention recently, being considered as one of the most promising materials for replacing synthetic polymers [5]. PLA has reasonably good optical, physical-, mechanical and barrier properties compared to existing petroleum-based polymers like high density polyethylene (HDPE), polystyrene (PS), polypropylene (PP) and polyethylene terephthalate (PET). The building blocks of PLA can exist in an optically active state. Depending on the enantiomers ratio, the PLA can obtain different mechanical, barrier and surface properties. This allows the production of a wide spectrum of PLA grades matching the performance requirements [6]. The thermoplastic property of those composites enables commercial manufacturing.

Extrusion, injection molding, casting of films and sheets, thermoforming, foaming and fiber spinning are among the methods utilized for PLA processing [7, 8]. The PLA technological waste can be reused to a great extent, which is another significant benefit of this polymer [9].

Despite the above-mentioned advantages the PLA materials show some disadvantages which limit its application, such as high brittleness, poor heat stability, low impact strength. To improve these properties, different modification methods like annealing, adding nucleating agents, fibers or nanoparticles were investigated. Alternatively, some chemical and physical treatments were applied to facilitate the cross linking between PLA molecules. Plasma treatment, UV-light and laser exposures were applied in order to enhance the material adhesion properties. [10, 11].

In this work, glass fiber reinforced, viscose rayon reinforced, and unreinforced PLA sheet were used in experimental thermoforming operation. The results obtained from the experiment and mechanical properties of composite materials were compared with results from unreinforced PLA.

2. EXPERIMENTAL

2.1 Materials

Poly (lactic acid) (PLA; molecular weight, M_w = 20kDa;M_n=10.1 kDa) was obtained from Biomer

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Krailling, Germany. Viscose rayon (Cordenka). Cordenka is Super 2 Rayon yarn with high elongation at break and optimized (= reduced) water shrinkage. It is used in mechanical rubber goods, with a focus on radiator hoses. Viscose rayon was obtained from Cordenka GmbH Germany. Glass fiber (stitch) density =300 g/m^2, glass – wool density =440 g/m^2. Both products were obtained from DHD Technology GmbH & Co.KG/ Germany.

2.2 Composites processing

The composites were obtained using press (Servitec Polystat 400S). The fibers were added between two PLA films and pressed. The processing parameters for press are: pressure = 90 kN/m^2, temperature = 155˚ C for 10 minutes. The compositions of the composites prepared are shown in Table 1.

Table 1. Compositions of composites

Materials	Time (min)	Filler (g/m^2)
PLA	5	-
PLA/Cordenka (short fibers)	10	90.5
PLA/Cordenka (stitch)	10	180.5
PLA/Glass fibers (stitch)	10	270.5
PLA/Glass - wool	10	320

The end products were manufactured by KIV Kreis GmbH. The thermoforming process was realized using Visual Thermoforming (HDVS 3036) vacuum skin packager. The processing conditions are shown in Table 2.

Table 2. Thermoforming conditions

Sheet temperature	160 – 170 º C
Heater temperature	350 º C
Heater distance from sheet	10 cm
Mold temperature	35 – 40 º C

Figure 1 shows the thermoform (A), and the thermoforming process (B). Figure 2 shows the thermoformed composites. Figure 2 (A) shows PLA, (B) shows PLA/Viscose rayon (short fibers), (C) shows PLA/Viscose rayon (stitch), (D) shows PLA/Glass fibers (stitch) and (E) shows PLA/Glass – wool.

2.3 Measurements

2.3.1 Mechanical testing. A mechanical testing machine, "Zwick 2000" Zwick GmbH & Co. KG (20 kN), Germany was used to measure the tensile properties according to ISO 14129. The flexural properties of a thermoformed composite were preformed according to the ISO 178. System control and data analysis were preformed using Datum software. All results presented are the average values of five measurements.

An EN ISO 179 charpy impact test was carried out using 10 unnotched samples. In each case a standard deviation < 15% (drop weight) was used to calculate the charpy impact strength.

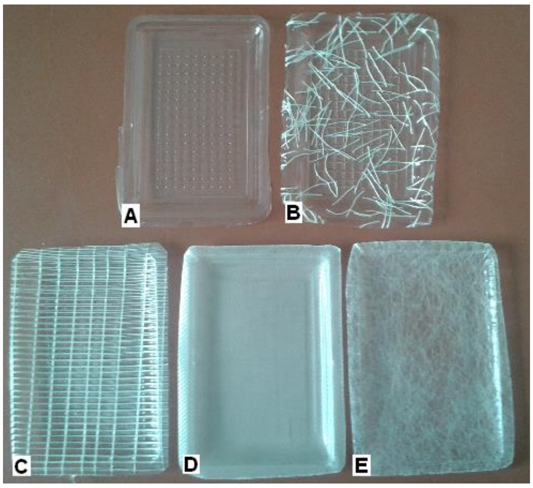


Fig. 1 (A) the thermoform, (B) the thermoforming process.

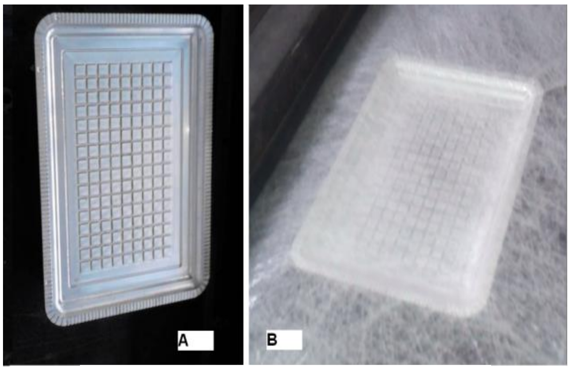


Fig. 2. The thermoformed composites.

2.3.2 Optical microscopy. The morphology of impact cross section of the composites was observed by optical microscopy at room temperature. A microscope (model VHX-600 Series) was used to collect microscopy images for the composite specimen.

4. RESULTS AND DISCUSSIONS

The results of the tensile properties investigation of PLA and PLA - composites are shown in Figs. 3 and 4.The pure PLA has a tensile strength of 58 MPa and a modulus of 3.8 GPa. The addition of glass fiber (stitch and wool) improved the tensile strength 178 MPa, 80 MPa and the modulus of 7 GPa and 5.7 GPA, which indicates that the stress transfers from PLA polymer matrix to the glass fibers. Whereas the addition of Viscose rayon fibers decreased the modulus of 3.9 GPa and the tensile strength of 30 MPa.

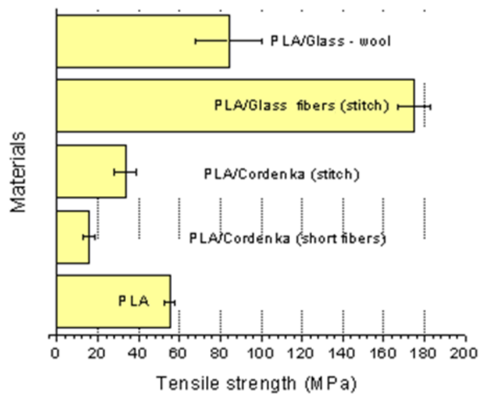


Fig. 3 Tensile strength of composites.

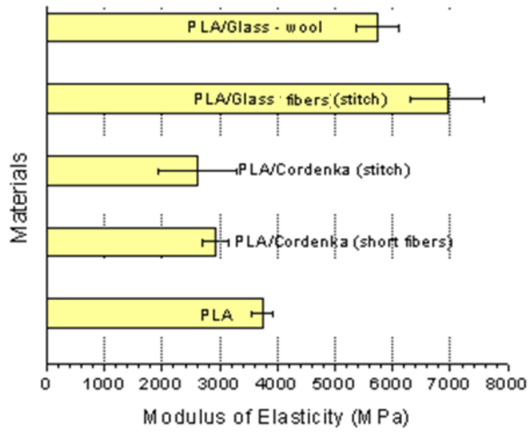


Fig. 4 Tensile modulus of composites.

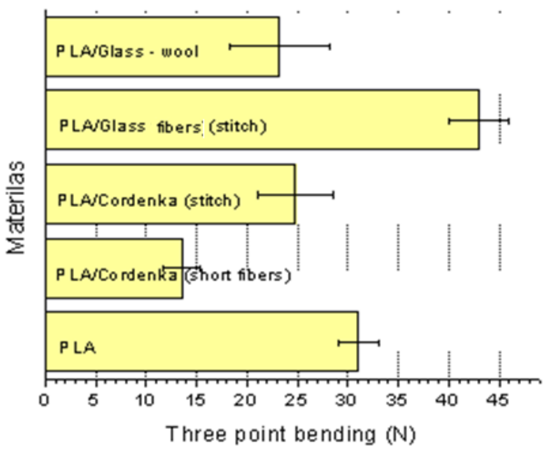


Fig. 5 Three point bending of thermoformed composites.

The three point bending tests of PLA and reinforced PLA composites (Fig. 5) were performed at room temperature. The pure PLA has a flexural tensile of 31 N. Glass fibers (stitch) composite improve the flexural tensile of PLA 42 N. No improvement could be observed in the Viscose rayon composites. Charpy impact samples were prepared by cutting and sanding to dimensions of 80 x 10 x 3.9 mm (length x width x thickness). The results of Charpy impact strength test are summarized in Fig. 6. All composites show improvement in impact strength. Composites with the Viscose rayon (stitch) improved the impact strength of fracture greater than 90 kJ/m^2.

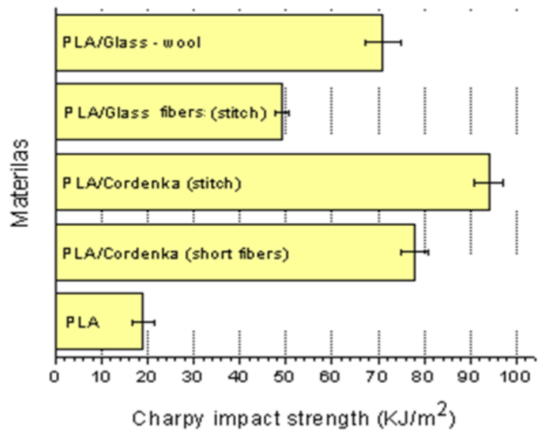


Fig. 6 Charpy impact strength of composites.

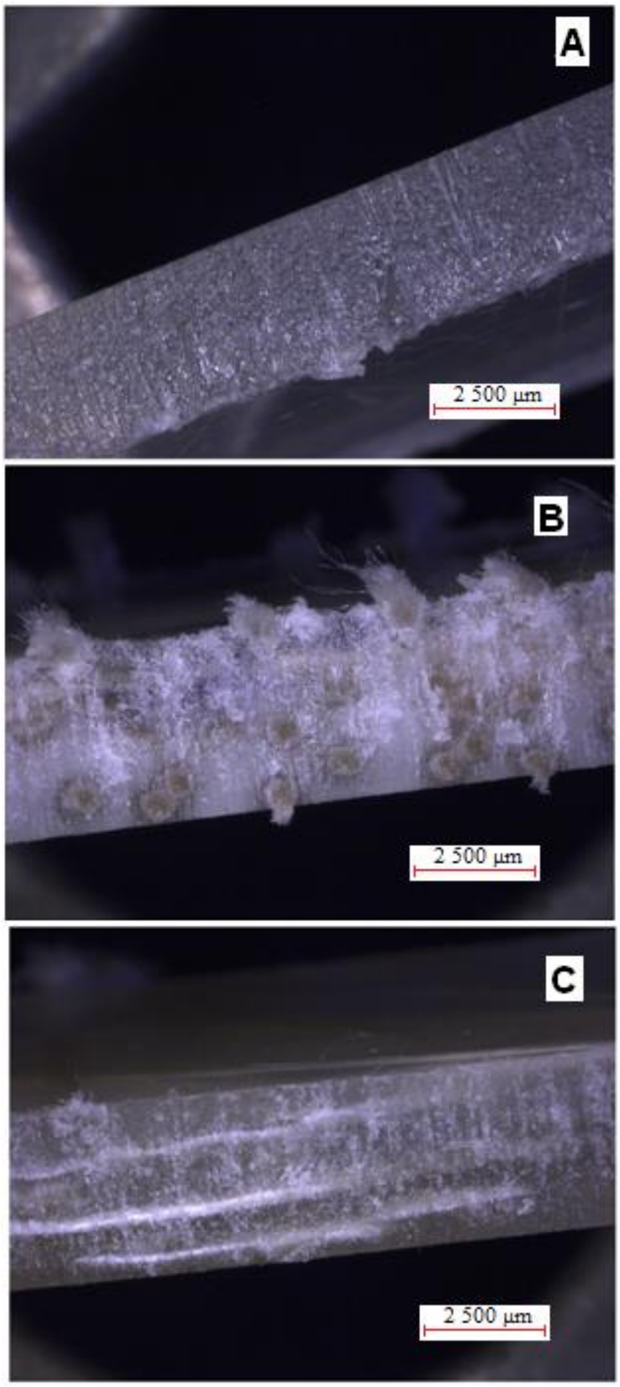
The digital microscopy images are shown in Figure 7. The fibers are illuminated white; the matrix is grey and the voids black. Microscopy analysis allows important individual void characteristics to be assessed. For example, the missing adhesion between the polymer matrix and the fibers can be clearly seen in Fig 7B, Fig. 7C and D proof of the bad adhesion based on the greater contrast between fibers and matrix. The best adhesion from all materials is that of the PLA/Glass wool, Fig. 7 (E). One explanation could be the fiber thickness; the thinner the fiber the better the adhesion.

5. CONCLUSIONS

Due to the variety of interesting composites properties, further development of the thermoforming process is highly recommended. One of the biggest advantages of those materials is tailoring their properties according to the application, followed by the economically efficient production process.

In many cases, the physical properties are enhanced dramatically compared to that of the sub-components. The lack of elasticity and the lack of compatibility in adhesion remains a problem as seen in PLA/Viscose rayon composites. The PLA/Glass fibers composites manufactured in this study have a higher tensile strength and a higher specific E-modulus than PLA/Viscose rayon composites. The three point bending test showed that only the glass (stitch) improved the flexure of composite.

PLA/Viscose rayon (stitch) improved the impact strength almost five times. The promising impact properties of the presented PLA/Viscose rayon composites show their potential as an alternative to traditional composites.



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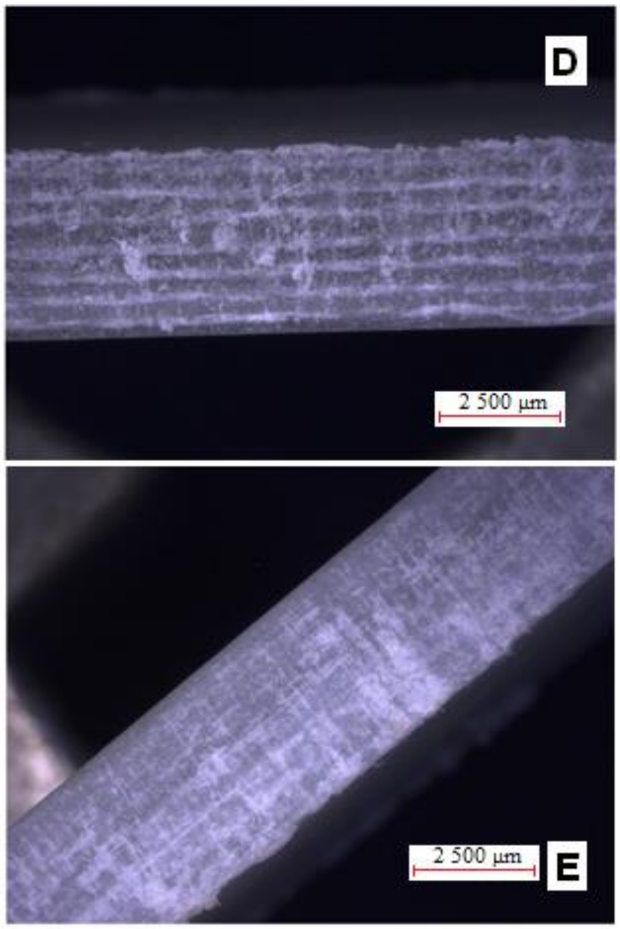

Fig. 7. Composite cross section (A) PLA, (B) PLA/Viscose rayon (short fiber), (C) PLA/Viscose rayon (stitch), (D) PLA/Glass (stitch), (E) PLA/Glass wool.

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ТЕРМОФОРМУВАНЕ НА БИО БАЗИРАНИ БАЗИРАНИ КОМПОЗИТИ

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(Резюме)

Тази работа цели изучаване поведението на дву-дименсионални композити (2-D композити) чрез термооформящ процес. За получаването на композитите е използвана полимлечна киселина (ПМК), стъклени влакна и вискозни влакна. ПМК беше предварително подготвена във формата на тънки полимерни листове чрез пресоване при температура равна на 155^{o} C и налягане от 90 kN/m^{2}. Така приготвените листове от ПМК са използвани в по-следващия процес при получаването на композитните материали. Крайните дву дименсионални продукти бяха получени във фабирката за производство на термопластични опаковки (KIV Kreis GmbH – Германия), чрез вакум термооформяне. Механчините свойства, якост на опън, модул на еластичност при опън, три точково огъване и якост на удар бяха изследвани. Морфологията на композитите бяха изследвани с дигитален микроскоп.