

PREPARATION AND CHARACTERIZATION OF SPARTIUM JUNCEUM L FIBER FILLED POLYCARBONATE COMPOSITES APPLICABLE IN THE AUTOMOBILE INDUSTRY

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<https://doi.org/10.37904/nanocon.2025.5017>

Abstract

The aim of this study is the modification of the polycarbonate (PC) as synthetic polymer with the natural polymer - Spartium junceum L (SJL) fibers to obtain a new biodegradable polymer material. Spanish broom (Spartium junceum L) was chosen as reinforcement for PC composites because of her low cost, light weight, renewability, biodegradability and easily availability in nature. The effect of different content of SJL fiber on the properties of PC/SJL composites was investigated. The PC composites reinforced with randomly oriented, short Spartium junceum L. fibers were prepared in Brabender mixer and hydraulics press. Mechanical properties of prepared PC/SJL fiber composites were analysed using dynamic mechanical analysis (DMA) and tensile tests. The morphology of the composites was analysed using scanning electron microscopy (SEM). The chemical properties were characterized by Fourier transform infrared spectroscopy (FTIR). The obtained results show that the SJL fibers increased the tensile strength and loss modulus as well as decreased the elongation of composites. The SEM analysis revealed that good adhesion between SJL fiber as filler and PC matrix was observed. The resulting PC/SJL fiber composites have the potential to be used in the automotive industry.

Keywords: Polycarbonate, Spartium junceum L (SJL) fibers, DMA, mechanical properties, SEM

1. INTRODUCTION

Synthetic polymer materials are mostly used as disposable packaging which makes them major environmental pollutants. Because of the large consumption of polymer materials, the large amounts of waste are created, which cause the creation of large landfills of synthetic polymers which are not biodegradable. Therefore, interest in biodegradable materials stems from the growing concern of people about the potential harm and the problem of waste disposal arising from the use of materials based on non-renewable sources of energy. Due to all mentioned above, a good waste management system is important, which includes prevention of waste generation, minimization of the amount of waste, reuse whenever is that possible and different disposal procedures, of which various recycling procedures bring exceptional importance and environmental benefit. In the last few years, there has been a growing interest in the commercial use of composites based on natural fibers, i.e. biocomposites, especially in the automotive industry. A significant amount of research has been dedicated to the use of development of biocomposites due to recyclable, environmentally friendly, and compostable alternatives to petrochemical based conventional plastics such as polypropylene (PP) and polyethylene (PE). Natural fibers represent sustainable materials, easily available in nature, with several advantages such as low cost, light weight, renewability, biodegradability and high specific properties [1]. The introduction of reinforcements of natural fibers is a practice for improving the properties of polymer materials. Also, the use of natural fibers as reinforcements for polymers is increasingly being investigated due to the

environmental acceptability of natural fibers compared to synthetic fibers. Some of the most used natural fibers are barely, oats, rye, sugar cane, bamboo, rice husks, hemp, etc. [2]. Polycarbonate (PC) is a thermoplastic polymer that is easily processed, moulded, and thermoformed. It is highly transparent to visible light, with better light transmission than many kinds of glass and because of this property PC finds many applications in optical devices. This polymer is a durable material, but although it has high impact resistance, its scratch resistance is poor and so a hard coating is generally applied to polycarbonate eyewear lenses and polycarbonate exterior automotive components. The biggest application of PC in automotive industry is for headlights, instrument panels and wheel cover as well as windows. In the Mediterranean region of Croatia Spanish broom are of great agricultural crop. It is composed of cellulose, hemicellulose and lignin. Lignocellulosic materials have a lot of positive features, such as low density, low requirements on processing equipment, no abrasion during processing, abundance and biodegradability.

2. EXPERIMENTAL

2.1 Materials

In this study Polycarbonate (PC) *Makrolon 2805* was supplied in pellet form by Bayer, Germany. As indicated by the manufacturer, the material has a melt flow index of 10 g / 10 min at 300°C. The *Spartium junceum* L fibers were obtained from the *Spartium junceum* L plant which was harvested around town Šibenik, Croatia.

2.2 Preparation of composites

To determine the influence of the *Spartium junceum* L fibers on the properties of the PC matrix, a series of composites was manufactured with different content of randomly oriented, short *Spartium junceum* L fibers (0, 5, 10, 15, and 20 % by weight). The samples were prepared using Brabender mixer at a temperature of 200°C and a screw speed of 60 min⁻¹ for 5 min, moulded in a hydraulic hot press Dake Model 44-226 (Holland) at 200 °C and a pressure of 25 kPa for 5 min and then cooled down between thick metal blocks at room temperature. **Figure 1** shows a photo of the hot pressed samples of investigated PC/SJL fiber composites used for further characterization.

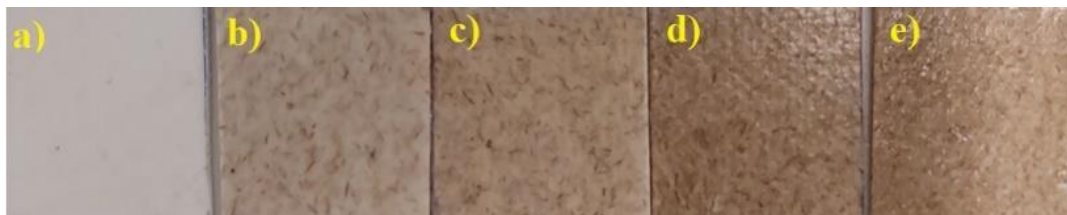


Figure 1 Photograph of PC/SJL fiber composites with a) 0; b) 5; c) 10; d) 15 and e) 20 wt.% of SJL fiber

2.3 Characterization

Dynamic mechanical measurements are performed with a TA Instruments model DMA 983 Analyzer. The measurements are conducted over temperature range of 25 °C to 200 °C at a constant frequency of 1Hz with amplitude of 0.3 mm. The heating rate was 3 °C min⁻¹. Tensile test was conducted on a universal testing machine (Zwick Roel Z020) with a testing speed 50 mm/min. At least five specimens were repeated. Infrared spectroscopy (FT-IR) spectra were obtained with a Perkin Elmer Spectrum 100 FTIR spectrometer (Waltham, MA, USA) using attenuated total-reflection (ATR) method. The analyses were carried out at room temperature and ambient humidity. All spectra were registered from 4000 cm⁻¹ to 650 cm⁻¹ with a resolution of 4 cm⁻¹. A scanning electron microscope (SEM) was used to evaluate the fracture surfaces, respectively, the interphase adhesion. Images of individual samples were taken on a JOEL JSM 7610F scanning electron microscope at different magnifications. The applied voltage was 1,00 kV.

3. RESULTS AND DISCUSSION

3.1 Viscoelastic properties

To investigate the viscoelastic properties (loss modulus, E'' and storage modulus, E') of the neat PC and PC/SJL fibers composites, we tested the dynamic mechanical properties using DMA. Generally, in biocomposites in which fibers are used as filler, the dynamic mechanical properties depend on the nature of the polymer and the interactions between the filler and the polymer matrix, as well as of the dispersion and orientation of the filler. **Figure 2a** shows the storage modulus (E') of the neat PC and PC/SJL fiber composites with different SJL fiber content. From **Figure 2a**, the storage modulus below the T_g (glassy region) of the neat PC and PC/SJL fiber composites maintains plateau and shows no significant difference among the samples. Above the T_g (rubbery plateau region), the storage modulus of the neat PC and PC/SJL fiber composites decreases with increasing the temperature. For the rubbery plateau region, the storage modulus of the composites is significantly increased with increasing the SJL fibers content, except for the composite with a content of 20 wt. % of SJL fiber (**Table 1**). This increases in storage modulus probably due to the increase in the crystal phase fraction derived from the crystal structure of cellulose present in SJL fiber. **Figure 2b** shows the curves of the loss modulus (E'') vs temperature for the neat PC and PC/SJL fiber composites. According to **Figure 2b** loss modulus curve of the neat PC exhibit one broad relaxation maximum at 55 °C which is related to the β -relaxation in PC due to the mobility of shorter segments in the polymer chain [3]. At a higher temperature (153,3 °C), a relaxation maximum of high intensity corresponding to the T_g of the PC was obtained. The T_g 's obtained for PC and PC/SJL fiber composites are shown in **Table 1**. The T_g values of the PC/SJL fiber composites decrease with increasing the SJL fiber content compared with neat PC. Similar results have been obtained for nanocellulose/PC composites [4]. This decrease in the T_g may be due to increased chain mobility of the amorphous PC with higher amount of lignin which also has an amorphous structure.

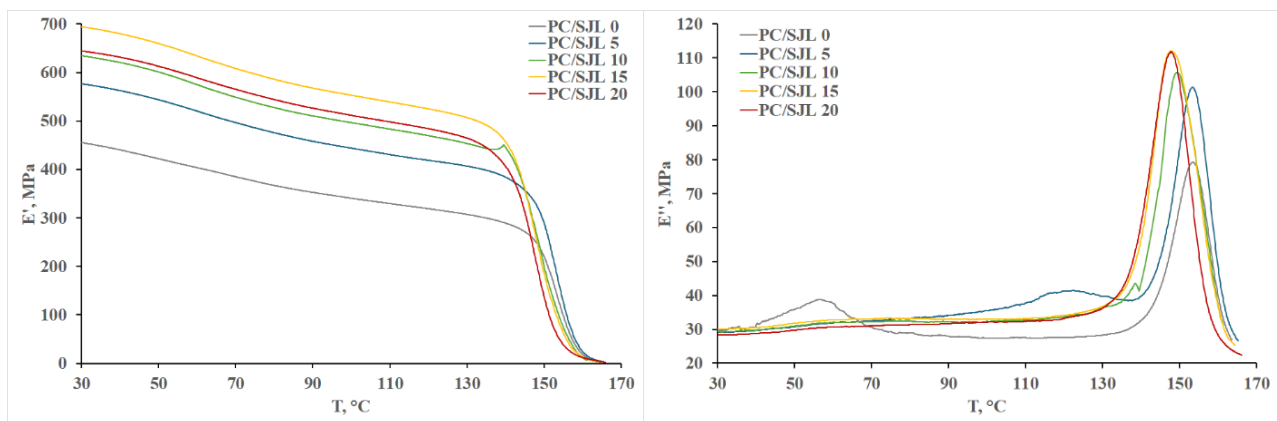


Figure 2 a) storage modulus (E') and b) loss modulus (E'') as a function of temperature for the neat PC, and PC/SJL fiber composites

3.2 Mechanical properties

The stress/strain curves of composites at different SJL fiber content were shown in **Figure 3**. **Table 1** present the results of tensile strength (σ) and elongation at break (ϵ) for the neat PC and PC/SJL fiber composites. It can be seen from **Table 1** that tensile strength of SJL fiber reinforced PC composites was higher when compared with the neat PC. Tensile strength of fiber reinforced PC composite increased with the increasing of SJL fiber content [5] and only slightly decreased when SJL fiber content reached 20 wt.%, as shown in **Table 1**. The reduction of tensile strength at higher SJL fiber content may possible due to the increasing of the probability of fiber agglomeration. On the other hand, the improvement in tensile strength was believed to be due to the better interfacial adhesion between SJL fibers and PC matrix. The interfacial bonding between fiber

and matrix played an important role for the mechanical property of fiber reinforced composite. The increased of tensile strength agrees with the increase of storage modulus value observed from DMA measurements. However, the elongation at break of SJL fiber reinforced PC composites were lower than the neat PC and decreased with increasing of fiber content.

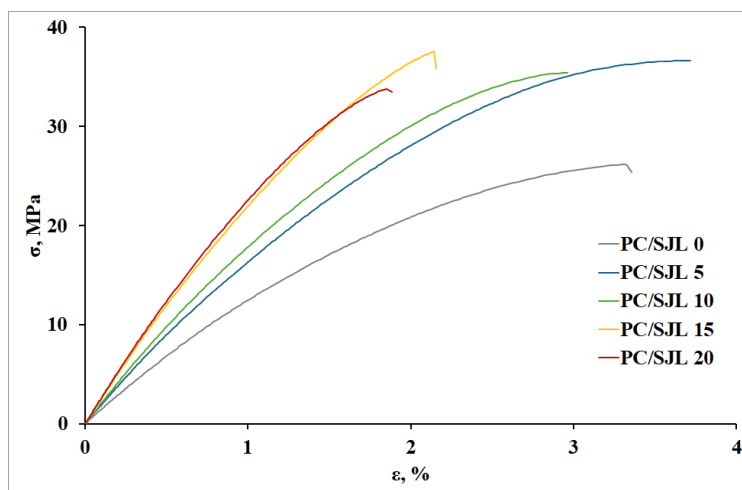


Figure 3 Stress/strain curves for the neat PC, and PC/SJL fiber composites

Table 1 DMA and mechanical properties of the neat PC and PC/SJL fiber composites

Sample	T_g E'' (°C)	$E'_{25^\circ C}$ (MPa)	σ (MPa)	ϵ (%)
PC/SJL 0	153.3	461.6	25.2±0.4	3.7±1.3
PC/SJL 5	153.5	580.9	36.6±0.2	3.1±0.1
PC/SJL 10	149.5	634.7	36.4±3.3	2.9±0.2
PC/SJL 15	147.9	698.6	39.2±5.2	2.1±0.6
PC/SJL 20	147.0	646.0	33.1±8.1	1.8±0.6

3.3 FTIR results

The characteristic absorption bands for the neat PC are observed in **Figure 4** and summarized in **Table 2**. PC is characterized by absorption bands at 2968 cm^{-1} , 2920 cm^{-1} , 2873 cm^{-1} and 2841 cm^{-1} which can be attributed to the C-H stretching vibrations of the CH_3 groups while absorption bands at 1768 cm^{-1} are derived from molecular vibrations of C=O groups [6]. The absorption band at 1502 cm^{-1} is associated to stretching C=C vibrations in the aromatic ring, the absorption bands at 1409 cm^{-1} and 1363 cm^{-1} deformation OH, and 1218 cm^{-1} , 1185 cm^{-1} , 1157 cm^{-1} and 1109 cm^{-1} , are attached to the deformation of acetyl C-O group [7]. The absorption peak at 1012 cm^{-1} corresponded to the stretching vibrations of the C-O-C group. **Table 2** shows the IR spectra of SJL fibers which are usually characterized by several absorption bands. The broad absorption band at 3334 cm^{-1} represents vibration of free OH groups of the cellulose while bands at 2915 cm^{-1} and 2899 cm^{-1} are attributed to the C-H symmetrical stretching of cellulose, hemicellulose, pectin, fats and waxes. The absorption band at 1604 cm^{-1} corresponds to the C=C stretching from the amorphous lignin. The absorption band at 1025 cm^{-1} corresponds to the C-C, C-OH, C-H ring and side group vibrations from cellulose. The band at 1100 cm^{-1} is related to the C-O-C glycosidic ether from cellulose and band at 896 cm^{-1} of the COC, CCO and CCH deformation and stretching in the cellulose. The typical band of O-H at 3334 cm^{-1} was visible in the FTIR spectrum of the PC/SJL fiber composites, indicating the presence of SJL fibers. The results in **Figure 4** show that was not significantly different of shape and position in FTIR spectra with increasing of SJL fiber content. The overlap of some absorption bands of SJL and PC was observed.

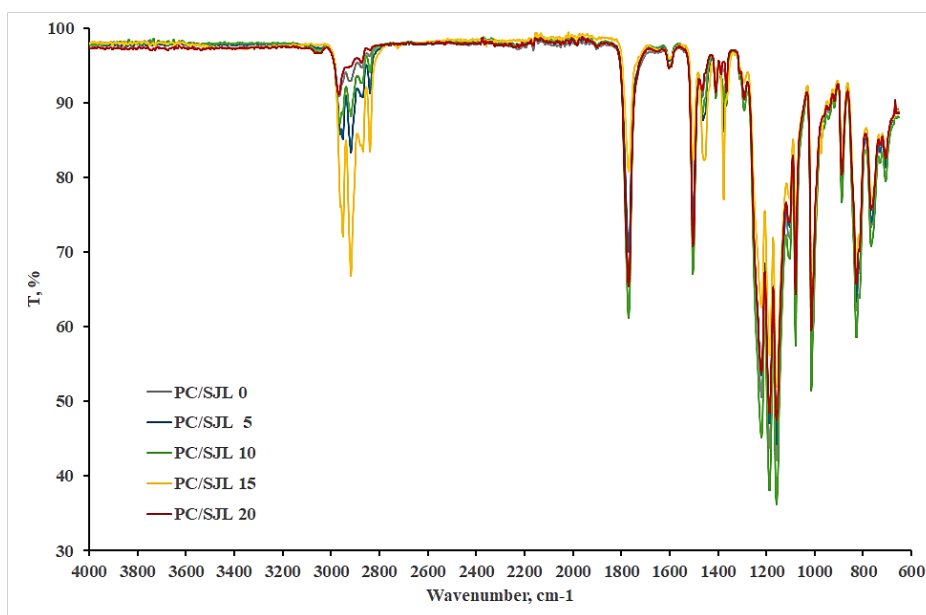


Figure 4 FTIR spectra for neat PC, and PC/SJL fiber composites

Table 2 FTIR data peak band assignments for PC and SJL fiber

PC	
Assignment	Peak position (cm ⁻¹)
CH stretching	2968 ,2920, 2873, 2841
C=O stretching	1768
C=C stretching	1502
OH deformation	1409, 1363
C-O deformation	1218, 1185,1157, 1109
C–O–C stretching	1012
SJL fiber	
Assignment	Peak position (cm ⁻¹)
OH stretching (Cellulose, Hemicellulose)	3334
C-H symmetrical stretching (Cellulose, Hemicellulose)	2915,2899
C=O stretching vibration (Pectin, Waxes)	1730
C=C stretching (lignin)	1604
C-O-C glycosidic ether (cellulose)	1100
C-C, C-OH, C-H ring and side group vibrations (cellulose)	1025
COC, CCO and CCH deformation and stretching (cellulose)	896

3.4 Morphology of the neat PC and PC/SJL fibers composites

The morphology of the neat PC and PC/SJL fiber composites was measured by scanning electron microscopy (SEM). The matrix-fillers interface plays critical role for ensuring that the properties of each component contribute optimally to the properties of the final product. The SEM micrograph of fracture surface after tensile test of the neat PC and PC/SJL fiber composites are shown in **Figure 5**. As observed from SEM images a good dispersion of fibers is observed for composites with content of fiber up to 15 wt.%. Also, it is possible to

note that the better the interfacial adhesion between SJL fiber and PC matrix was observed in composites with less fiber content (**Figures 5b-5d**). We can conclude that a good adhesion between reinforcing fiber and PC matrix interphase resulted in tensile strength improvement up to 15 wt.% of SJL fiber content (**Table 1**).

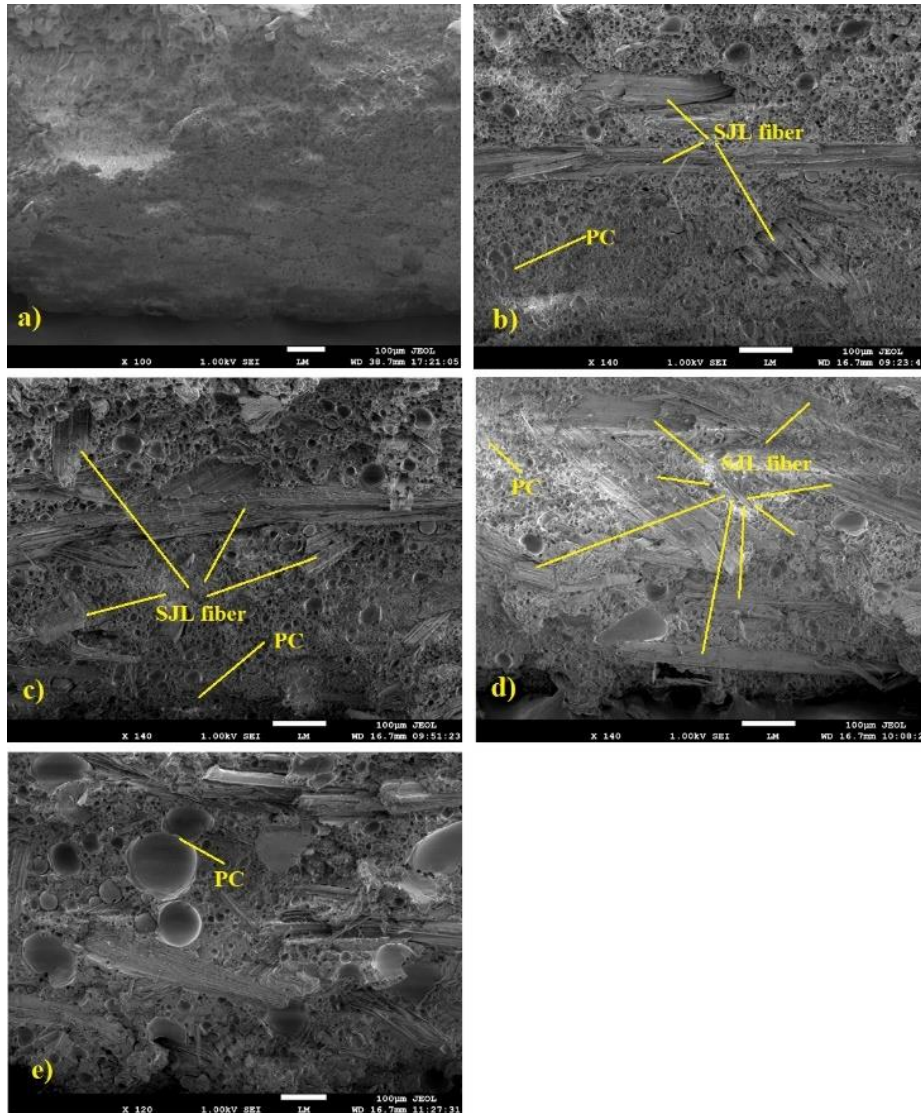


Figure 5 SEM images of the fracture surface of (a) neat PC; (b) PC/SJL 5; (c) PC/SJL 10; (d) PC/SJL 15 and (e) PC/SJL 20 composites

4. CONCLUSION

In this work, the polymer composite samples of polycarbonate (PC) matrix with *Spartium junceum* L (SJL) fibers were successfully prepared via hot melt in Brabender mixer. The results of dynamic mechanical analysis (DMA) according to storage modulus (E') and the tensile strength (σ) of PC/SJL fiber composites showed that E' and σ increased with increasing of fiber content. Good adhesion between SJL fiber and PC matrix resulted in tensile strength improvement reinforced PC composites. However, the elongation at break (ϵ) and glass transition temperature (T_g) decreased with increasing fiber content compared with the neat PC due to the increased chain mobility of the amorphous PC. The FTIR analysis results showed the presence of SJL fibers in PC/SJL composites and the overlap of some absorption bands of SJL and PC. Upon SJL fiber addition, there are not major changes of shape and position of the characteristic absorption bands, and it can be

concluded that FTIR spectra of the composites are practically identical to the spectra of neat PC, except of the absorption bands intensity. The results of morphological studies of PC/SJL fiber composites clearly showed that a good dispersion of fibers is observed for composites with content of fiber up to 15 wt.% and this resulted in tensile strength improvement. Considering the results of mechanical, chemical and morphological studies, it is evident that the SJL fiber as reinforcement might improve the investigated properties of PC. Based on these results we assumed that SJL fibers represent good candidate as potential application in the automobile industry.

ACKNOWLEDGEMENTS

The research in this work was supported by grant no.121174 University of Zagreb, Croatia.

We would like to acknowledge to the “Functional integration of the University of Split, PMF-ST, PF-ST and KTF-ST through the development of scientific research infrastructure in the building of three faculties” (KK.01.1.1.02.0018), a project co-financed by the Croatian Government and the European Union through the European Regional Development Fund—the Competitiveness and Cohesion Operational Programme.

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