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Development of PLA based nanocellulose films for packaging applications

R. K. Gond and M. K. Gupta*

Department of Mechanical Engineering, Motilal Nehru National Institute of Technology Allahabad, Prayagraj-211 004, Uttar Pradesh, India

E-mail: mkgupta@mnnit.ac.in, mnnit.manoj@gmail.com

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Owing to environmental concern, demand of nanocellulose of natural fibres has been increasingly increased in packaging applications. In this work, nanocellulose of sugarcane bagasse was isolated through a series of chemical treatments followed by the mechanical grinding process. The films of neat PLA and nanocellulose reinforced PLA were prepared by the solvent casting method with changeable weight percentages (i.e. 1, 2, 3, and 4) of nanocellulose of sugarcane bagasse. Tensile properties, optical properties and morphological analysis of the prepared films were studied. The better performance of tensile properties was exhibited by the film with 2 wt.% of nanocellulose. Transparency values of the films were found to decrease with adding of nanocellulose of sugarcane bagasse.

Keywords: Polymer film, nanocellulose, tensile property, optical properties.

Introduction

In the world, around 275 million tones of plastic waste are produced each year and approximate 8.5 million tones of plastic is dumped into sea. In current scenario, India contributes 9.6 million tones of plastic waste annually which is neither biodegradable nor recyclable, and also hazardous to health as well as environment. Because of the biodegradability and recyclability, biopolymer materials are being used opposite to petroleum-based polymers for packaging materials. Polylactic acid (PLA) is one of the most utilized biodegradable biopolymer extracted from renewable plant resources i.e. wheat, corn, sugar beet and potatoes[1]. Recently, PLA has been used in many areas such as packaging materials, textiles industry, biomedical field and automobiles industry because of its good mechanical strength, thermal properties, biocompatibility, easy processing and good optical properties. On the other hand, for food packaging applications, PLA has some limitations also like poor barrier properties, high brittleness, low tensile strength and low heat resistance[2].

Nanocellulose is one of the commonly used filler material isolated from the cellulose. A cellulose is the most abundant, renewable, light weight and biodegradable material derived from wood, rice husk, stalk, hemp, flax, arecanut, bamboo, cotton, sugercane bagasse, kenaf, and other plant based materials[3]. The term nanocellulose indicates that at least one dimension of cellulosic material must be in nanometer scale (0.1–100 nm). The mechanical, thermal, and optical and barrier properties of PLA was found to be enhanced by incorporation of nanocellulose[3]. Most of the nanocellulose extracted from the source materials by various chemical treatments and various mechanical processes[4]. The nanocellulose from jute fibres was extracted using ball milling process and observed enhanced mechanical properties of PLA film after incorporation of nanocellulose from jute[5]. Extraction of nanocellulose from rice hulls was done by bleaching and ultrasonication technique[6]. Author examined the thermal stability, microstructure and mechanical analysis of films. Solvent casting method was obtained to develop alginate based nanocomposite film subjected to mechanical and thermal tests[4]. Same method was also used to prepare the films of polymers reinforced by nanocellulose[7]. In another study, nanocellulose of sugarcane bagasse was obtained, and morphology and surface topography analysis of its films was carried out[8]. Nanocellulose of the natural fibres was extracted and its polymer based films were prepared for the various analysis[9].

Based on literature review, it was found that characterization of PLA based films reinforced with nanocellulose of sugarcane bagasse was unreported so far. In this work, PLA based film of the nanocellulose was prepared by solvent casting method subjected to analysis of the tensile strength, surface morphology and optical property.

Materials and methods

Materials:

Sugarcane bagasse was obtained by K. M. Sugar Mills, Faizabad, Uttar Pradesh, India. PLA in pellets form was purchased from Nature-Tech India Pvt. Ltd., Chennai, India. The properties of sugarcane bagasse fibre[10] and PLA[11] are provided in published literature. Consumable materials such as sodium hydroxide, concentrated hydrochloric acid and chloroform were purchased from Science Corporation Johnstonganj, Prayagraj, Uttar Pradesh, India.

Preparation of films:

In this study, bagasse nanocellulose (BNC) was obtained through a series of chemical treatments (i.e. alkali treatment, acid hydrolysis and alkali treatment) followed by a grinding process. The films of neat PLA and nanocellulose reinforced PLA with varying weight percentages (1, 2, 3 and 4) were prepared by the solvent casting technique, as shown in Fig. 1. Firstly, PLA pellets were dried in a hot air oven at 60°C for 24 h to eliminate the moisture content before use. Dried PLA (5 g) were dissolved into 50 ml solvent (chloroform). The solution was stirred for 30 min at 70–80°C with help of a magnetic stirrer. When PLA was completely dissolved in solvent then BNC was poured into solution and was kept on magnetic stirrer for uniform mixing. Dissolved PLA/BNC was evenly spread out at room temperature in a Petri disk. After 2–3 days dried film were removed from the Petri disk and stored in a dry place. The thickness of the prepared film was found between 0.015–0.025 mm. Nomenclature used for prepared films with varying wt.% is as such: P(0) = neat PLA, NC(1) = 1 wt.% BNC film, NC(2) = 2 wt.% BNC film, NC(3) = 3 wt.% BNC film and NC(4) = 4 wt.% BNC film.

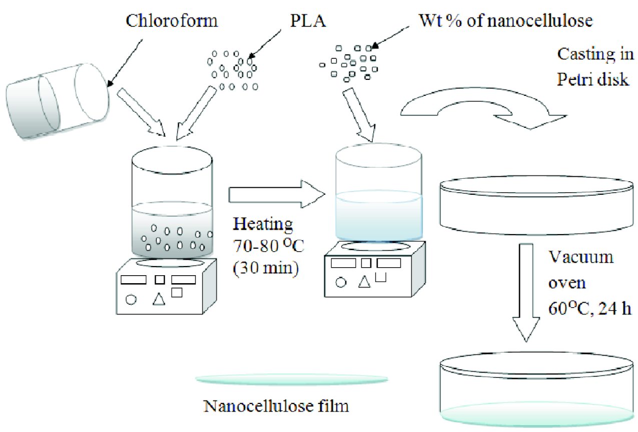

Fig. 1. Prepration of bagasse nanocellulose film.

Characterization

Film thickness:

Thicknesses of the films were measured by digital thickness measuring micrometer (0.015–0.025 mm) to the nearest 0.001 mm. The measurement of films was taken at 5 different locations of each sample, and mean values of thickness at those locations were used to calculate the properties of the films.

Morphological analysis:

The morphological analysis of prepared neat PLA and BNC/PLA films was carried out using FESEM (Model: Nova nano SEM 450), with gold coating of each film for 2 min.

Tensile properties:

The tensile properties of each film were determined according to ASTM standard D882-97. The test was carried out on Tinius-Olsen UTM with 10 kN load cell. The films were cut into a strip (15 mm×80 mm) for tensile test. Five samples of each film were tested and their average values are reported.

Optical property:

Transparencies of the films were obtained using UV-Vis spectrometer (Model: Shimadzu, UV-3600 Plus). The film samples were cut into rectangular shape and inserted into the test cell. A neat PLA sample was set as a reference for evaluating the optical properties of each composite film. The transparency of the films was calculated using the following eq. (1).

$$TV = \frac{\log(T)}{X} \quad (1)$$

where TV is transparency value of film, T is the transmittance at different wavelength range and X is the thickness of the film in mm. From above equation; higher value of T indicates the lower transparency and higher degree of opacity.

Results and discussion

Morphological analysis:

The FESEM micrographs were obtained to analyze the morphology of PLA based nanocellulose films at different magnifications. The FESEM micrographs of neat PLA and PLA/BNC films are given in Fig. 2. Fig. 2a shows the micrograph of neat PLA with voids, whereas Figs. 2b and c indicate that the nanocellulose was homogeneously dispersed and was not aggregated within the PLA matrix. The similar surface morphologies were seen in past literture[7]. From Figs. 2d and e, it was observed that some nanocellulose particles (white dots) are aggregated at different locations as compare to Figs. 2b and c. Furthermore, it was also observed that due to increase in percentage of nanocellulose in PLA matrix, surface roughness of the films was found to be increased. Therefore, low surface roughness in Figs. 2b and c was seen as compared to Figs. 2d and e owing to presence of lower concentrations of nanocellulose.

Tensile properties:

The tensile properties in terms of strength, modulus and elongation (%) of the prepared nanocellulose films were performed. Size of the reinforcement highly affected the tensile properties of the composites, whereas nano size of cellulose offers a high surface to volume ratio providing better reinforcing capacity results in improved tensile strength. Tensile strength and elongation percentages of neat PLA and PLA/BNC films are given in Table 1. From the experimental results, it was observed that each film offers better tensile strength than neat PLA; shows a positive effect of reinforcement of nanocellulose into PLA. Further, tensile strength of the films was found to be improved with increase in content of nanocellulose up to 2 wt.%, and on further loading it was found to decrease. The highest value tensile strength was seen for the film NC(2), which was 63.33% more than that of film P(0). The highest tensile strength of 25.06 MPa for PLA film incorporated with 3 wt.% of nanofibrillated cellulose was reported by Mao *et al.*[12]. In another study, maximum tensile strength of 17.4 MPa for starch/CNC film was reported[15]. The highest value of tensile strength could be due to uniform dispersion of nanocellulose in PLA matrix and strong interfacial adhesion between them. Uniform dispersion of nanocellulose was already analyzed in surface morphology by FESEM (Fig. 2c). Beyond 2 wt.% loading of nanocellulose, tensile strength of the films was decreased due to agglom-

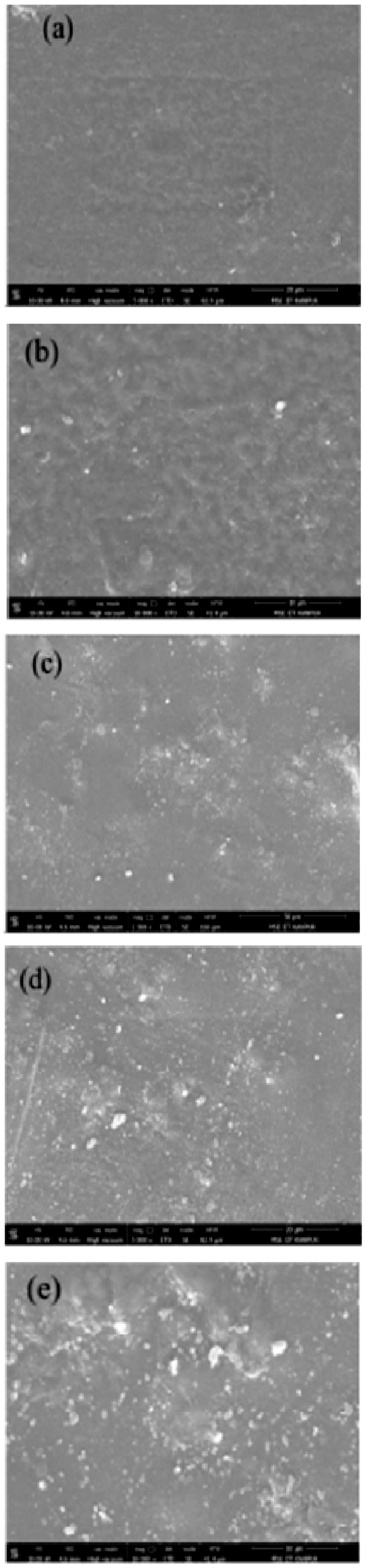

Fig. 2. FESEM micrographs of neat PLA and nanocellulose films: (a) P(0), (b) NC(1), (c) NC(2), (d) NC(3) and (e) NC(4).

eration of nanocellulose in matrix which results in improper stress transfer at interface. In the case of elongation at break, the highest value was observed for PLA, whereas lower values were offered by the films because of less ductility owing to incorporated of nanocellulose. An enhanced mechanical property of PLA/BNC films offers an enhanced load carrying capacity.

Table 1. Tensile properties of nanocellulose films

Film	Tensile strength (MPa)	Tensile strength increase (%)	Elongation at break (%)
P(0)	15.00	–	180.00
NC(1)	19.24	28.26	59.00
NC(2)	24.50	63.33	27.50
NC(3)	20.00	33.33	24.10
NC(4)	22.00	46.66	40.00

Optical property:

Ultraviolet (UV) indicates the region of electromagnetic spectrum between visible light and X-ray. UV light is categorized into three different types by the Scientists i.e. UV-A (320–400 nm), UV-B (290–320 nm), UV-C (100–290 nm). UV- C light is very harmful that kills the microorganisms of food, air and water. Optical properties of prepared neat PLA and PLA/BNC films were measured by UV-Vis spectrometer. The optical transparency of the films was considered in the wavelength range of 200–800 nm. Transmittance as a function of wavelength of the films is presented in Fig. 3. Table 2 represents the light transmittance of the films at wavelength of 280 nm and visible wavelength of 600 nm.

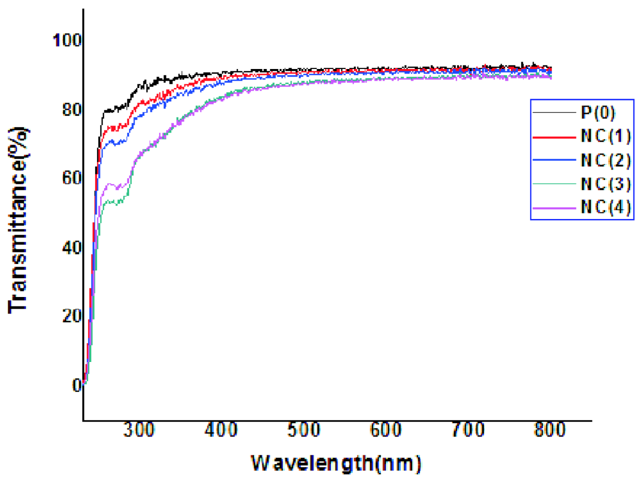

Fig. 3. Transmittance vs wavelength graph of nanocellulose films.

Table 2. Optical properties of nanocellulose films

Film	Transmission (%)	
	Wavelength (280 nm)	Wavelength (600 nm)
P(0)	81.30	92.39
NC(1)	75.58	91.77
NC(2)	71.49	91.32
NC(3)	55.19	89.40
NC(4)	58.49	89.35

It can be noticed that the transmittance of the films was decreased with increasing the nanocellulose content. It was already reported that addition of nano particle in polymer matrix reduces its light transmittance[13]. The PLA/BNC films showed the lower light transmission than PLA film. Above to 600 nm wavelength, transmittance became almost constant for all the films. Similar type of result was also reported for PVA/NFC nanocomposite films[14]. In the visible region, 91.32% of light was transmitted and approximate 8% of light was absorbed by the film with incorporation of 2 wt.% of nanocellulose. In polymer matrix, reinforcement of nanoparticle resists the harmful UV radiation.

Conclusions

The major findings from the present experimental work are as follows:

(i) A uniform dispersion of nanocellulose in PLA matrix was confirmed by FESEM analysis. However, some agglomeration was present in the films contain 3–4 wt.% of nanocellulose.

(ii) Tensile strength of PLA film was significantly improved by incorporation of nanocellulose. The best tensile strength was revealed by the film contains 2 wt.% of nanocellulose.

(iii) Light transmission of PLA and PLA/BNC films was found to decrease by reinforcement of nanocellulose.

(iv) On the basis of the present experimental outcomes, the prepared film can be proposed for the packaging applications.

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