science, engineering and health studies

Research Article

Science, Engineering and Health Studies
https://lio1.tci-thaijo.org/index.php/sehs
ISSN (Online): 2630-0087

Biobased plastic films from cogon grass cellulose

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ABSTRACT

The study of using natural resources such as cellulose as an alternative to synthetic polymers for manufacturing biobased plastic packaging is currently on going amongst numerous researchers to achieve sustainable goals as well as the need to replace petroleum feedstock that are getting depleted by years. Cogon grass is one of the most problematic grasses globally, and it has been observed that cogon grass grows abundantly along the roadside and open areas in Malaysia. Since the said grass is high in cellulose content (64%) it is a suitable candidate for raw material in manufacturing biobased plastics. Hence, the objective of this study was to produce a thin biobased film from cogon grass cellulose. In this study, cogon grass cellulose was extracted using sodium hydroxide before being acetylated to yield cellulose acetate (CA). Solvent casting with dichloromethane (DCM) was performed on the synthesized CA to produce a biobased thin film with different CA concentrations (1%–5%) to study the properties of the thin films. The film with 5% CA had the highest tensile strength (10 MPa). The surface morphology of the thin film was smooth, even and homogenous indicating that the acetylation process during cellulose acetate formation successfully occurred and that the DCM used can penetrate the cellulosic fibre very well.

Keywords: cogon grass; cellulose; biobased film; plastic packaging

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***Received**: 28 April 2023*
***Revised**: 8 July 2023*
***Accepted**: 26 July 2023*
***Published**: 15 December 2023*

***Citation**:
Soloi, S., Teng, O. W., Sarjadi, M. S., and Majid. M. H. A. (2023). Biobased plastic films from cogon grass cellulose. Science, Engineering and Health Studies, 17, 23020005.

1. INTRODUCTION

Biobased packaging is defined as packaging made from primary, annually renewable resources, which are classified into three generations based on their historical development. (Robertson, 2008). Biobased packaging that is fully derived from natural materials such as cellulose is known as third generation biobased packaging. Cellulose $(C_6H_{10}O_5)_n$ is one of the most abundant organic materials on earth, with an attractive combination of different properties existing in the cell walls of all plants (Tajeddin, 2014). The derivative forms of cellulose are usually used in biobased packaging which possess better applicability than cellulose. Among the cellulosic derivatives, cellulose acetate (CA) has gained greater interest because it has excellent optical properties, it is biodegradable, it has a high toughness, and it can produce films either by melting or solvent-casting techniques. It also has low moisture barrier properties and can be modified for film production (Rajeswari et al., 2020).

Cogon grass is one of the most problematic grasses globally, and it has been observed that cogon grass grows abundantly along roadside and open areas in Malaysia. Due to its high cellulose content (64%) (Mohd Kassim et al., 2015), it can be utilized as the raw material for biobased plastic packaging. To date, there has been no research on the preparation of cogon grass as a biobased plastic, hence, this research was presented to study the suitability of cogon grass as a cellulose resource for biobased plastic packaging.

In this study, cogon grass cellulose was extracted using sodium hydroxide since it is a simple extraction method that requires suitable parameters for maximum cellulose yield (Karim et al., 2022). The cell wall of the fibers is disrupted during the treatment process through the hydrolyzation of acetic acid and uronic acid esters, resulting in swollen

cellulose and dissolved lignin, hemicellulose, and silica (Dinh Vu et al., 2017). In addition, the soda pulping method is less reactive towards cellulose than other methods such as sulfate pulping.

2. MATERIALS AND METHODS

2.1 Materials and chemicals

Cogon grass was collected from the UMS roadside that was growing wildly. Sodium hydroxide (NaOH), hydrogen peroxide (H_2O_2), glacial acetic acid (CH_3COOH), sulfuric acid (H_2SO_4), acetic anhydride ($C_4H_6O_3$), dichloromethane (CH_2Cl_2), hydrochloric acid (HCl), and glycerol were all produced by Sigma-Aldrich and purchased from a local supplier.

2.2 Cellulose extraction

The wild cogon grass was collected from the roadside of Jalan UMS, Sabah, Malaysia. The whole plant of the cogon grass, except the rhizome was collected and used in this study. The collected grass was washed thoroughly to remove sand and other contaminants. It was then cut to about 1 cm in length and dried in an oven at 60 °C for 24 h. This was added to a blender to obtain a fine powder for cellulose extraction (Azmin et al., 2020).

2.3 Cellulose acetylation

The cellulose extraction method was adopted from (Azmin et al., 2020), which uses alkaline treatment followed by a bleaching process. With continuous stirring, 100 g of fine cogon grass powder was soaked in 2,000 mL of 4.5 M NaOH solution for 3 h at 100 °C. The solution was then rinsed off using distilled water until a neutral pH was achieved. The sample was, then immersed in hydrogen peroxide (H_2O_2) for 45 min at 70 °C for bleaching. After that, the H_2O_2 was similarly rinsed off using distilled water until neutral pH was obtained. The obtained cellulose fibres were then subjected to acetylation according to the method reported by (Homem and Amorim, 2020). Thus, 10 g of cellulose was added to 200 mL of glacial acetic acid and stirred for 30 min. This was followed by the addition of a 1.5 mL sulphuric acid (H_2SO_4) and 87.5 mL glacial acetic acid solution, with 15 min further stirring time. Subsequently, 100 mL of acetic anhydride was added and stirred for another 30 min, then finally left to stand for 24 h. After this, 500 mL of distilled water was slowly added into the mixture and stirred for another 1 h. The mixture was filtered and rinsed until neutral pH was obtained. The CA was dried at room temperature for 3 days and then the degree of acetylation was determined using titration. The properties of the obtained cellulose were characterized using Fourier transform infra-red (FT-IR) spectroscopy.

2.4 Preparation of biobased plastic

The cellulose thin film production was carried out based on the study of (Candido et al., 2017) with some modifications. Using the solvent casting method, the synthesized CA was solubilized in CH_2Cl_2 (1:50 w/v) and stirred for 2.5 h. Then, 20% (w/v) glycerol was added to the solution and further sonicated for 10 min. The cast solution was poured into a petri dish using a syringe to control the thickness of the film and the solution was left to evaporate at room temperature. The obtained film was then characterized by FT-IR, SEM, and tensile strength analysis to examine the properties of the film. Five formulations of the biobased plastic were prepared and tabulated in as Table 1.

Table 1. Compost characteristics

Sample	Cellulose acetate to dichloromethane ratio (w/v)	CA concentration (%)	Glycerol content (g/mL)
A	0.5:50	1	20
B	1:50	2	20
C	1.5:50	3	20
D	2:50	4	20
E	2.5:50	5	20

2.5 Fourier transform infra-red spectroscopy

An attenuated total reflectance FT-IR spectrometer (Perkin Elmer Spectrum 100) was used to detect the functional groups present in cellulose as well as to monitor the lignin removal. The plastic film was put directly on the sample probe upon scanning. The FT-IR spectra was obtained in the range of 3000 cm^{-1} to 450 cm^{-1} with 16 scans per sample.

2.6 Tensile test

The tensile strength of the synthesized thin film was tested using a universal testing machine (GOTECH Al-7000M) with ASTM D882 used as the reference. The thin film samples were cut into the standard size of 7 cm × 1.5 cm. Finally, the test was carried out with the maximum head speed of 1 mm per min with a load of 50 N. Each sample was in run triplicate.

3. RESULTS AND DISCUSSION

3.1 Cellulose extraction and acetylation

The yield of cellulose fiber obtained in this study was 37.91%, comparable to the cellulose obtained by a previous study (Mohd Kassim et al., 2015) that used the soda extraction method. The FT-IR spectra shown in Figure 1 confirms that the lignin was successfully removed, as the peak of C=C stretching of aromatic lignin at 1627 cm^{-1} had shifted after the treatment. Meanwhile, the hydroxyl group (-OH) stretching of the cellulose resonated at around 3334 cm^{-1} becoming sharp and intense after the treatment. This proves that that the alkaline treatment using NaOH followed by bleaching with H_2O_2 had successfully broken the lignin bonds in the cogon grass (Sayakulu and Soloi, 2022).

Meanwhile, for the synthesis of CA, the degree of acetylation was found to be 2.76 which indicates that cellulose triacetate was formed in this study (Daud and Djuned, 2015). This high degree of substitution was attributed to the elimination of large amounts of lignin during the cellulose extraction. The presence of lignin will compete with cellulose during acetylation. Hence, the removal of lignin increases the activation of cellulose, which allows better accessibility of acetic anhydride towards the free hydroxyl group during the reaction (Candido et al., 2017).

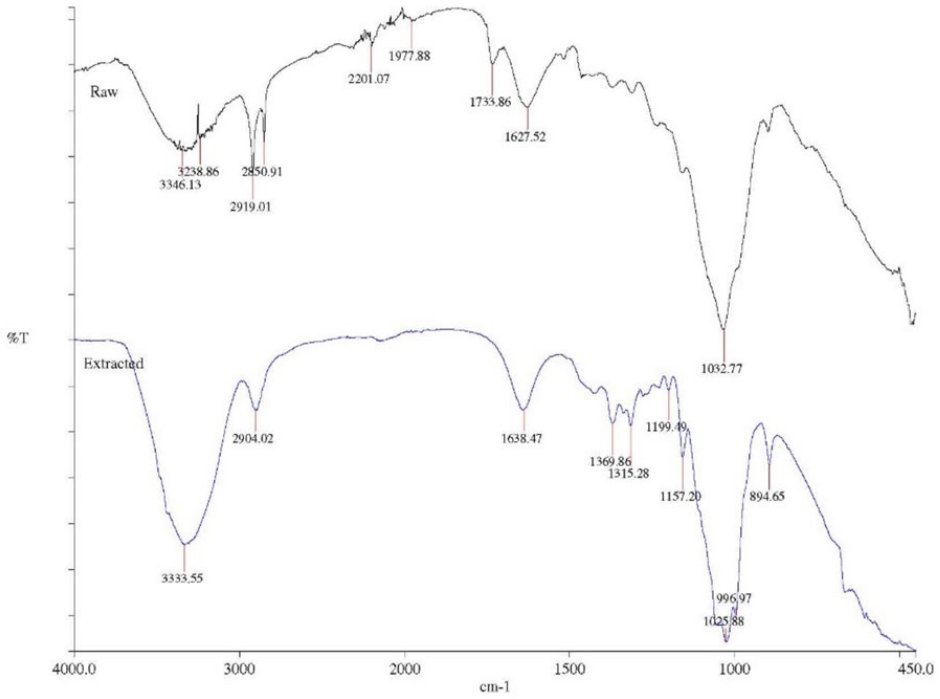

Figure 1. FTIR spectra of cogon grass cellulose

3.2 Cellulose acetate film

The obtained cellulose acetate films were successfully prepared with the characteristics seen in as shown in Table 1. The films were transparent, as shown in Figure 2.

3.3 Surface morphology of thin film

The SEM results showed that the surface of the thin film samples was smooth, even, and homogenous. The surface morphology of the samples remained unchanged with an increase in CA concentration. The smooth surface indicates that the acetylation process during CA formation successfully occurred, and that the reagent used can penetrates the cellulosic fiber very well (Araújo et al., 2020).

3.4 Tensile strength

Figure 3 highlights that the sample with the lowest CA concentration (1%) had the lowest tensile strength of 0.03 MPa, while the thin film sample with the highest CA concentration (5%) shows had a higher tensile strength of 10.46 MPa. The tensile strength results show the higher the CA concentration, the higher the viscosity of the casting solution. The increase in viscosity causes an increase in surface tension of solutions (Bai et al., 2012; Rodríguez et al., 2012), and the surface tension of the casting solution is related to the tensile strength. The increase in surface tension of the casting solution eventually increases the tensile strength of thin film samples (Jindal et al., 2016). The tensile strength of the obtained films ranged between 2–10 MPa, which is comparable to commercial low density polyethylene (LDPE) that is widely used as packaging of non-load goods (Szlachetka et al., 2021).

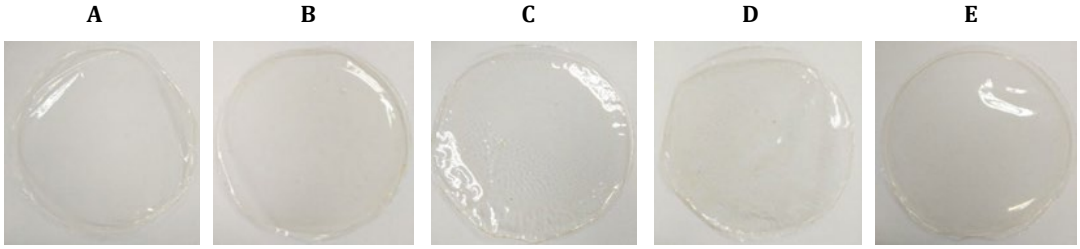

Figure 2. Physical appearance of cellulose acetate films by sample

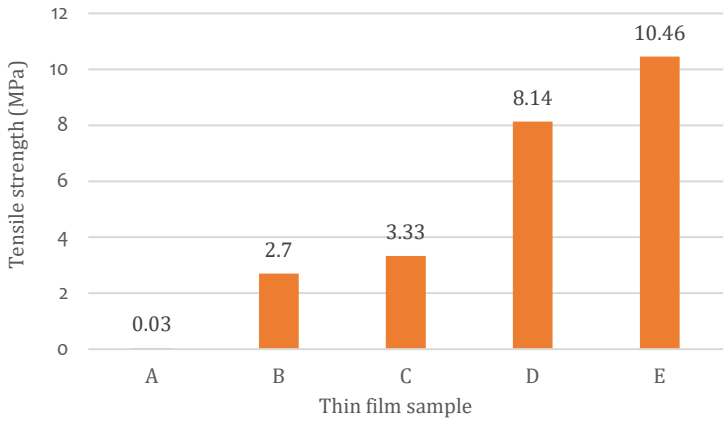

Figure 3. Tensile strength of cogon grass cellulose thin film

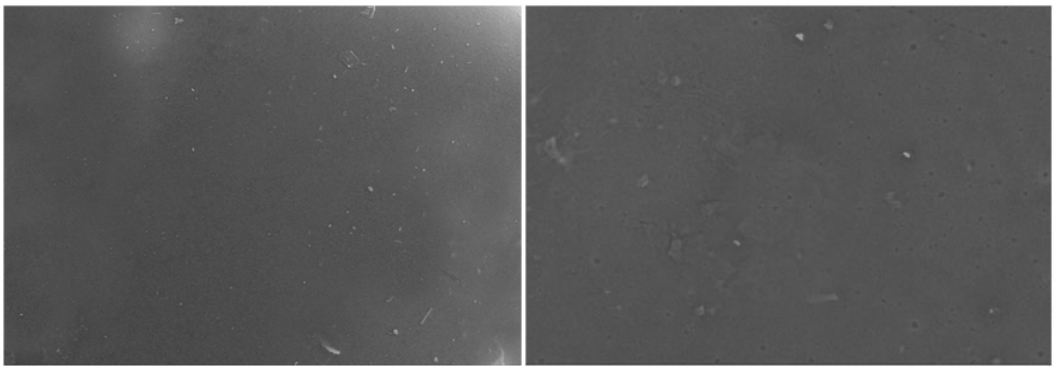

Figure 4. SEM image of cellulose acetate film at 100× (left), and 500× (right) magnification

4. CONCLUSION

The FT-IR reveals that the soda extraction followed by bleaching successfully removes lignin. The SEM images in Figure 4 show that the biobased plastic has a smooth and even surface. The thin film produced at 5% CA had the highest tensile strength comparable to the tensile strength of LDPE from petroleum-based plastic. This concludes that the cogon grass cellulose has the potential to be developed as a biobased plastic for non-load goods.

ACKNOWLEDGMENT

This research was carried out in the University Malaysia Sabah and financially supported by the Fundamental Research Grant Scheme under the grant code RACER/1/2019/STG01/UMS/2.

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