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Investigating the characteristics of carboxymethyl cellulose film as a possible material for green packaging

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Abstract

The primary objective of the current work was to create a value-added biodegradable product for sustainable packaging. The primary goal of using carboxymethyl cellulose (CMC) made from agricultural waste is to lower the cost of making the film, because currently available commercial CMS is expensive. The main aim of the research is to convert the CMC obtained from agricultural waste into a usable biodegradable polymer that can be used as packaging material. Investigations were done into how well hemicellulose-based carboxymethyl cellulose (H-CMC) films performed. Oil palm empty fruit bunches (OPEFB) were used to extract hemicellulose using an alkali-ethanol extraction method. Following that, distilled water was added to the newly generated hemicellulose, and it was embedded with carboxymethyl cellulose (CMC) at various hemicellulose loadings (30, 50, 70, and 90 wt%). H- CMC films were made using the solution casting technique, and they ranged in thickness from 0.07 to 0.12 mm. The tensile test, fourier transform infrared spectroscopy (FTIR), and differential scanning calorimetry (DSC) and SEM analysis were used to characterize the produced films. The hemicellulose content of H-CMC films has raised the glass transition temperature (Tg). The optimal loading of hemicellulose into CMC as a possible material for green packaging applications, according to the overall results, can be determined to be 70 percent weight. By integrating functional elements for upcoming applications including electrically conductive and photocatalytic thin films, the research further attempted to develop the film structure.

Keywords: Extraction; Hemicellulose; Analysis; Colorimetry; SEM

1. Introduction

Environmental effects and pollution caused by plastic trash are currently top priorities in the context of sustainable development. Production of biopolymers, or polymers obtained from biological sources, is one strategy [1]. Plant cell walls contain polysaccharides known as cellulose, hemicellulose, and pectin, which are classified as dietary fibres [2]. A biopolymer called hemicellulose is found in nature and is made from renewable resources like protein and polysaccharides. Due to its low cost and biodegradability, it has a significant potential to replace petroleum-based polymers that cannot degrade [3]. During the pulping and defibration of lignocellulose biomasses, hemicelluloses are frequently discarded. Amorphous and highly branched short sugar chains are found in hemicellulose, the second most prevalent plant polysaccharide after cellulose [4]. This biopolymer is characterized as being biodegradable, non-toxic, and having a lower molecular weight than cellulose [5, 6]. Literature praises the fact that if plastic waste is not properly burned, it will again end up as litter in the environment[7]. Approximately 300 million metric tons of conventional plastic will be released into the environment without being sorted in 2018, according to a report by Olunivi et al. [9].

Recycling plastic is not always simple; there can be an issue with mixed plastic pollutants, and the end products from these wastes might not be appropriate for making post-consumer goods. By taking into account their nontoxicity, biocompatibility, renewability, and biodegradability features, the demand for bioplastic as an alternative to

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conventional plastics has increased globally[9]. Due to its strong water resistance, low gas permeability, and exceptional flexibility, hemicellulose is one of the most promising materials for use in the manufacture of films. Nevertheless, because to its extreme brittleness, poor mechanical characteristics, and hygroscopic nature[10]. The limitation described above may be solved by combining or mixing hemicellulose with other naturally occurring commercially available polymers or biopolymers, according to Coma in 2013. Carboxyl methyl cellulose (CMC) is one of the best instances of biopolymer that draws interest from researchers. It is a cellulose derivative and a significant commercial biopolymer that doesn't impair human health and is very successful at enhancing product quality. The mechanical characteristics, transparency, flexibility, and moisture absorption are all positively impacted by CMC[11].

The use of lignocellulosic biomass was given a lot of attention as a result of the large increase in oil prices as a result of their scarcity. One of the main agricultural lignocellulosic by-products of the palm oil industry is oil palm empty fruit bunch (OPEFB). 4 kg of dry biomass can be saved for every kilogram of palm oil produced. One-third of the biomass in this waste is OPEFB, with the remaining two-thirds consisting of oil palm trunks and fronds [12]. Malaysia releases 19.8 million tons of OPEFB annually. As a result, it is easily accessible and inexpensive [13]. OPEFB includes 25% lignin, 23% hemicellulose, and 33% cellulose [14]. Although determining the biopolymers' permeability to fragrance and flavour components is important, little research has been done in this area[15].

2. Methods and materials

2.1. Materials

From padma oil industry in Bangladesh, OPEFB (oil palm empty fruit bunch) was acquired. After extracting the oil from the oil palm, it was soaked in water for 24 hours and then rinsed with tap water to get rid of any remaining oil and debris. OPEFB was then allowed to air dry for 24 hours at room temperature. Prior to the extraction process, the dried residue were sieved and sieved with a 60 m particle size. Making the particle was done by proximate analysis.

2.2. Extraction of hemicellulose

Alkaline extraction was used to extract hemicellulose from OPEFB in accordance with a method previously described by Ariffin et al. in 2006[16]. Before delignification, which was done before isolating hemicellulose, the extractive material (organic) in the OPEFB was extracted using the soxhlet extraction procedure. This powder is known as extractive free OPEFB. Sodium hypochlorite (NaClO2) was used to delignify the extractive free OPEFB powder for two hours at 70°C. After the delignification process, the holocellulose was refluxed with 1.0 M sodium hydroxide (NaOH) at 45 C for 4 h while being vigorously stirred to create hemicellulose. Following the extraction, 90 ml of ethanol solution was added to the supernatant in order to separate the solid hemicellulose from the solution. The combination was then permitted to stand for 24 hours. Hemicellulose is precipitated from the solution formed during the separation of cellulose and hemicellulose with the use of ethanol. After that, a table top Laboratory Centrifuge automated Rotolavit II (manufactured in China) was used to centrifuge the hemicellulose solution for 15 minutes at 3500 rpm. Before making the films, the precipitate was dried at 40°C and maintained in a desiccator.

2.3. H-CMC film preparation

According to the mixing ratio, various weights of the hemicelluloses were first dissolved in distilled water before being combined with CMC for 10 hours while being stirred at 80 rpm using a magnetic stirrer. The total mass of the solution was kept constant at 20 g, and Bransonic ultrasonic baths (Danbury, CT, USA) were used to further sonicate the resulting solution.

Sample code	Weight (%)	hemicellulose
СМС	100	0
Н-СМС-В	90	10
Н-СМС-С	70	30
H-CMC-D	50	50
Н-СМС-Е	30	70

 Table 1
 Hemicellulose films formulation

Using a knife blade, the prepared solution was thrown onto a glass plate, where coagulation was allowed to occur for 24 hours. The film was then taken out of the distilled water and let to dry at room temperature. Then, until analysis, those were conditioned at 23°C and 50% RH.

2.4. Color and thickness measurement

A colorimeter (CM-3500D Minolta spectrophotometer, Minolta, Japan) calibrated with CM-A124 zero calibration box and CM-A124 white calibration plate was used to determine the colour of the film. The Hunter lab colour scale was used, with chromaticity parameters (A) denoting red direction, (-A) denoting green direction, (B) denoting yellow direction, and (-B) denoting blue direction. Each sample category received five measurements. The thickness of the film was measured using a thickness gauge (Mitutoyo Kawasaki, Japan) with a resolution of 0.0001 mm. For each film style, the average of four separate spots was calculated.

2.5. H-CMC's mechanical definition

The Texture Analyzer was used to test the films' tensile strength. In order to determine the effective cross-sectional area, the thickness of each sample was first measured using a micrometer screw gauge at three different locations along its length. The mean value was then multiplied by the sample's 15 mm width. Initial load cells and grip separations were 25 mm and 30 kg, respectively. Each formulation's H-CMC film samples were uniformly sliced and laid out on the Texture Analyzer. A 0.8 mm/s loading speed and a 5 mm/min strain rate were used. According to ASTM D882 [17], tensile strength (TS) and elongation at break (EAB) were computed. Every film sample received an average of five measurements.

2.6. Thermal characterization by differential scanning colorimetry(DSC)

A hermetic aluminium pan containing around 10 mg of H-CMC films was filled, sealed, and put inside the differential scanning colorimeter (PerkinElmer, Massachusetts, USA). With a heating rate of 10 C/min, the sample was heated under nitrogen purge from 30 to 400 °C. Continuous data collection was done for temperature and time intervals, with an empty hermetic aluminium pan serving as the reference.

2.7. H-CMC's morphological description

SEM imaging was used to examine the surface morphology of H-CMC films in cross-section (Leo Supra, 50 VP, Germany). To avoid surface charge on the specimen when it is exposed to the electron beam, the tensile tested broken cross sections of each sample were fixed onto the SEM holder using double-sided electrically conductive carbon adhesive tapes. Prior to their morphological detection, gold was sputtered because the coatings were non-conductive. The SEM micrograph was created using a 5 kV accelerated voltage under standard secondary electron imaging settings.

2.8. (FTIR) analysis

Thermo Scientific's Nicolet iS10 FTIR spectrometer with a Smart iTR ATR ZnSe with HP clamp was used to conduct the FTIR analysis of the H-CMC films. ATR (Attenuated Total Reflectance) mode was used to record the spectra of H-CMC films, and the software Origin Pro 2016 was used to analyze the peaks.

Table 2 Composition of sample

Component	Sample wt % from literature	Present study wt % approx.	
Free extract	4.1	3.93	
Hemicellulose	28	23.4	
Cellulose	62.9	59.86	
Holocellulose	82.4	78.65	
Lignin	22.8	14.7	
Ash	5.19	3.6	

Hemicellulose film	Color analysis			Thickness (mm)
	L value (lightness)	A value (redness)	B value (yellowness)	average value
СМС	97.2	0.08	1.60	0.128
Н-СМС-В	75.5	0.13	1.59	0.098
Н-СМС-С	87.1	0.11	1.62	0.093
H-CMC-D	93.7	0.28	5.23	0.161
Н-СМС-Е	69.2	0.39	8.10	0.087

Table 3 Physical analysis of different hemicellulose loadings in CMC and H-CMC films.

3. Result and discussion

3.1. Direct analysis of sample

The polysaccharide portion of holocellulose is created when cellulose and hemicellulose are present [18]. The principal OPEFB components detected in the current work are nearly identical to earlier observations published by Birnin-Yauri et al. in 2016 [19], according to the results. In actuality, the general qualities of the raw materials used will vary depending on where the raw materials come from. However, the findings demonstrate that the holocellulose content of OPEFB is notably high. Oil palm empty fruit bunch can be considered a raw material with a high yield for the creation of green packaging as a result.

3.2. Physical analysis

Table 3 provides a summary of the findings from the physical characterization of the CMC and the pre-pared H-CMC films. The findings demonstrate that, when compared to films obtained from other sources, the thickness of the H-CMC films made by the solution casting procedure (0.09-0.161 mm) was within the acceptable range. Other hemicellulose-based films had previously reported film thicknesses in the ranges of (0.29-0.38 mm) for stalk xylane, (0.13-0.14 mm) for hemicellulose from oil palm found, (0.048-0.062 mm) for hemicelluloses from bamboo, (0.13-0.15 mm) for hemicelluloses from sugarcane bagasse[20]. In the meantime, the findings of the colour study showed that CMC film has very light colours and might be the best material for uses like packaging where transparency is a prerequisite. The lack of hemicellulose contributes to the higher transparency of CMC films. H-CMC-E, on the other hand, is regarded as the least transparent film and can be the perfect choice when light-sensitive packaging products are needed.

3.3. (FTIR) Fourier transform infrared spectroscopic analysis

The spectrums obtained for H-CMC films are nearly identical to the spectrum of pure CMC, according to FTIR analysis. Thus, it is proven that CMC's chemical composition is unaffected by the addition of hemicellulose. The strong hydrogen bond may be seen by the O-H spectra with the stretching absorption in the broad spectra of all films between (3647-3581) cm—1. Additionally, at (2918-2841) cm—1 areas, strong C-H stretching vibration absorptions may be seen. The high absorbance peak at (1618–1586) cm–1 in the fingerprint area is ascribed to C-H deformation. According to the findings reported by Xue et al. in 2012 [21], the peak at 1750 cm—1 is attributed to symmetric stretching from COO groups. In the pure CMC spectrum, the peak at 1587 cm—1 denotes the carbonyl absorption signals. This is a result of the carboxylic acid group's presence in the CMC and it signifies the insolubility of water [22]. C-O-C vibration is represented by the peak at 1253 cm—1 in all CMC and H-CMC films. The addition of hemicellulose to CMC can maintain a chemically stable structure of CMC, as shown by the FTIR spectroscopy study for the prepared films[23].



Figure 1 FTIR spectra for a) H-CMC-B b) H-CMC-C, c) H-CMC-D and d) H-CMC-E films.

3.4. Analysis using differential scanning colorimetry (DSC)

The DSC curves of each produced film are shown in Fig. 2. Table 4 shows the glass transition temperature (Tg) and melting temperature (Tm) of CMC and H-CMC films. The findings showed that the removal of moisture was associated with a minor endothermic peak in the CMC and H-CMC films at 100 °C. The peak at 100 °C, according to Zohuriaan & Shokrolahi in 2004, demonstrates the hydrophilic nature of films [24]. Tg of H-CMC films ranges from 133 to 187 degrees Celsius, indicating a high degree of compatibility and favourable molecular interactions between the two polymers (hemicellulose and CMC).



Figure 2 DSC curve for a) CMC; b) H-CMC-B; c) H-CMC-C; d) H-CMC-D and e) H-CMC-E films.

Figure-2 showed that as hemicellulose loading went beyond 20% of hemicellulose weight, the melting temperature (Tm) of H-CMC increased[25]. CMC functions as a lubricant at lower loading levels, resulting in increased hemicellulose

loading in this investigation. The hemicellulose film matrix's association with CMC and intermolecular interactions can be reduced by this lubricating function. As a result of the unordered chain arrangement, the structure's degree of crystallinity decreases. As a result, the amount of heat energy needed to completely melt the spherulite increases, which is what causes the broad peak to form in the melting zone for large hemicellulose loadings (low CMC level). That could be as a result of the drop in CMC content leading to an increase in less crystalline or amorphous structure[26].

Sample	Tg (Degree Celsius)	Tm (Degree Celsius)	
СМС	185.65	338	
Н-СМС-В	131.1	336	
Н-СМС-С	187.2	258.7	
H-CMC-D	187.6	260.6	
Н-СМС-Е	180.7	259	

Table 4 The melting and glass transition temperatures for CMC and H-CMC films.

3.5. SEM, or scanning electron microscopy

Due to the addition of hemicellulose into CMC, H-CMC morphology has a rounded shape while pure CMC exhibits a smooth micro-structure. The addition of hemicellulose increases this rounded shape. However, the significant amount of branch chains in the hemicellulose backbone in the 80 weight percent of hemicellulose loading causes the microstructure of H-CMC-E to be rough. Hemicellulose aggregation occurs at large concentrations and is the cause of the distortion of cross sectional morphology, according to Fahl'en and Salm'en in 2005. Even with large hemicellulose loadings, this effect aids in the reduction of TS[23,27]. The cracked cross-sectional surfaces of the CMC and H-CMC films were examined using scanning electron microscopy (SEM), and those pictures are shown in Fig-3.



Figure 3 Surface morphology for a) CMC, b) H-CMC-B, c) H-CMC-C, d) H-CMC-D and e) H-CMC-E films

3.6. Future prospective

The most important cellulose derivative, CMC, has been employed as a great stabilizer in the food sector and is potentially the best raw material for packaging materials. The CMC identified its strong barrier capabilities against carbon dioxide, oxygen, and lipids as its primary distinguishing trait. It can therefore be viewed as a crucial ingredient in creating a more lasting film. According to numerous researchers' descriptions[28–30], CMC does have remarkable mechanical and barrier qualities that provide for greater compatibility. Additionally, biobased packaging materials could reduce the environmental pollution caused by the disposal of plastic waste, particularly in the packaging industry. Another area that needs a lot of attention from researchers is the recovery of CMC because it is expensive by its own price and requires economical extraction methods. Better availability for creating substantially less expensive polymerbased products would result from that. The current effort has the potential to draw additional researchers to the topic of waste management in agriculture, wealth creation utilizing trash, and has the potential to be a breakthrough in the packaging industry.



Figure 4 Chemical synthesis of CMC from different sources (source: google)

4. Conclusion

In the current study, we reliably retrieved the targeted CMC, a valuable macromolecule contained in the agricultural waste, throughout the studies. By using FTIR, DSC, SEM, swelling, and biodegradation investigations, all the samples were characterized and the intricate analysis of CMC emerged. In this study, blended CMC and hemicellulose films were created by adding hemicellulose at intervals of 20% and ranging from 0 to 70 percent weight. The prepared films' average thickness ranged from 0.08 to 0.16 mm. The features of native CMC are still present in the produced films, according to the FTIR spectra, despite the inclusion of hemicellulose in a variety of amounts. DSC tests show that the addition of hemicellulose to CMC has improved the thermal stability of pure CMC. Therefore, it can be identified that a 70 weight percent hemicellulose content is the ideal level to add to CMC in order to improve the mechanical and thermal durability of pure CMC for the production of biodegradable packaging materials. The recent study provides a solid foundation for scientists developing efficient waste conversion technology. The current study focuses on the manufacturing of biodegradable plastic utilizing CMC mixes that are made from agricultural waste.

Compliance with ethical standards

Disclosure of conflict of interest

There is no conflict of interest regarding this paper.

Availability of data and materials

The data and materials used to support the findings of this study are publicly available.

Author contribution

All author contributed significantly to design and development of this work.

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