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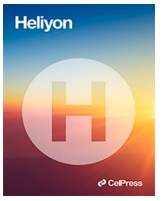

Research article

Development of eco-friendly biofilms by utilizing microcrystalline cellulose extract from banana pseudo-stem

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ABSTRACT

Banana pseudo-stem, often considered as an underutilized plant part was explored as a potential reinforced material to develop an eco-friendly biofilm for food packaging applications. In this study, Microcrystalline cellulose (MCC) was extracted from banana pseudo-stem by alkali and acid hydrolysis treatment. The extracted MCC was used as a reinforced material in different concentrated polyvinyl alcohol (PVA) matrix alone as well as both PVA and Carboxymethyl Cellulose (CMC) matrix to develop biofilm by solvent casting method. The synthesized MCC powder was characterized by scanning electron microscope to ensure its microcrystalline structure and to observe surface morphology. The biofilms composed of MCC, PVA, and CMC were assessed through Fourier-transform infrared spectroscopy (FTIR), mechanical properties, water content, solubility, swelling degree, moisture barrier property (Water Vapor Permeability - WVP), and light barrier property (Light Transmission and Transparency). The FTIR analysis showed the rich bonding between the materials of the biofilms. The film incorporating a combination of PVA, CMC, and MCC (S6) exhibited the highest tensile strength at 26.67 $\pm$ 0.152 MPa, making it particularly noteworthy for applications in food packaging. MCC incorporation increased the tensile strength. The WVP content of the films was observed low among the MCC-induced films which is parallel to other findings. The lowest WVP content was showed by 1% concentrated PVA with MCC (S4) (0.223 $\pm$ 0.020 10^{-9} g/Pahm). The WVP content of S6 film was also considerably low. MCC-incorporated films also acted as a good UV barrier. Transmittance of the MCC induced films at UV range were observed on average 38% (S2), 36% (S4) and 6% (S6) which were almost 6% lower than the control films. The S6 film demonstrated the lowest swelling capacity (1.42%) and water content, indicating a significantly low solubility of the film. The film formulated with mixing of PVA, CMC and MCC (S6) was ahead in terms of food packaging characteristics than other films. Also, the outcomes of this study point out that MCC can be a great natural resource for

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packaging applications and in that regard, banana pseudo-stem proves to be an excellent source for waste utilization.

1. Introduction

Banana (*Musa paradisiaca*) is one of the most common trees in the tropical and subtropical areas of the world. Banana plants, which are members of the *Musaceae* family, are indigenous to the Malaysia-Indonesian region of South East Asia [1]. Bangladesh produces about 840,362 tones of banana per year [2]. Various kinds of banana species are harvested each year (Genre Musa) but only the edible portion (banana fruit) of the plant is utilized. The residue is discarded, and they are generally treated as wastes to the environment. The remains of the banana plant, for instance, leaves; pseudo stems are burned when the plant produces a large quantity of greenhouse gas in the air after collecting banana fruits. Almost all the parts of the banana plants are useable, like fruits are nutritious for our health [3], peels are used as antioxidant [4], leaves are used for wrapping [5], and pseudo stems are used for making specialized and high-quality sanitary products such as baby pampers [6], stalks are used as prevents diabetes [7].

The term "banana pseudo stem" refers to a unique feature of banana plants. The banana pseudo stem, often referred to simply as the banana stem, is a distinctive and essential part of the banana plant (*Musa species*). It is not a true stem but rather a thick, cylindrical structure that emerges from the underground corm of the plant. The banana pseudo stem is used for sanitary products such as baby pampers, pulp, paper raw material, fiber for textiles, and filler or structural reinforcement in composite materials [6]. Other uses of this fiber are for making coffee and tea bags, filter cloths, as reinforcement fibers for plaster, disposable fabrics, and light-density woven fabrics. The pseudo-stem fiber of the banana plant is similar to pineapple leaf, sisal, and other stiff fibers, but it is slightly more elastic [1].

The banana plant residue contains cellulose, the world's most prevalent molecule and a key structural component of plant cell walls. As cellulose is widespread in plants, derived from materials like wood and cotton, it is a renewable resource, contributing to its sustainability. Cellulose, being biodegradable, can be broken down by natural processes, making it environmentally friendly compared to synthetic alternatives. Its strength and rigidity properties, crucial for plant structure, also make cellulose valuable for industrial applications, such as paper, textiles, films, and composites. In the food industry, cellulose-based packaging materials offer complete biodegradability, biocompatibility, and organic derivation, presenting a promising alternative to polyethylene-based environment friendly packaging materials [8,9]. Recent research interest in cellulose stems from its exceptional capacity as a reinforcing material, favourable mechanical qualities, low density, and positive environmental effects, driving applications in composite material production [9]. Polyvinyl alcohol (PVA) is a synthetic, flexible, and biodegradable polymer that is utilized in the packaging industry due to its film-forming characteristics, biodegradability, crystallinity, and mechanical properties [10].

Carboxymethyl Cellulose (CMC) is a cellulose derivative with carboxymethyl groups (–CH_2–COOH) bound to some of the hydroxyl groups of the glucopyranose monomers that make up the cellulose backbone. A cellulose-based biopolymer called carboxymethyl cellulose (CMC) was developed. It is made up of β-D-glucose and β-D glucopyranose 2-O-(carboxymethyl)-monosodium salt that are connected by β-1,4-glycosidic bonds. It is commonly used as a viscosity modifier or thickener, and to stabilize emulsions in various products, both food and non-food but not limited to the use in preparing nanostructured bio composites, hydrogels, paper and films [11]. Recent advancements have come to the light on plasticized poly bio composites embedded with nano fibrillated cellulose to emerge as an alternative to existing polyethene based packaging materials but are limited to use due to the unpredictable ductility and impact toughness [12].

Plastic packaging materials pose a significant environmental problem due to their widespread use and persistence in the environment. Additionally, they have negative impacts on ecosystems and the health of all living forms, similar to the environmental pollution caused by heavy industrialization and urbanization [13,14]. The impact of today's plastics packaging and industrial wastes of all forms are leaving a significant threat to the aquatic life due to presence of heavy metals [15–17]; toxic dyes [18] in it. Nowadays, the use of synthetic polymers in everyday life causes a slew of environmental issues and worries for human society [19]. Most plastic packaging materials are non-biodegradable, which means this can persist in the environment for hundreds of years [20]. This long lifespan intensifies their environmental impact. Because such plastic materials are difficult to degrade, they generate massive amounts of garbage, resulting in serious environmental difficulties [21]. Plastic containers and films used in commercial applications that are manufactured from synthetic polymers derived from petrochemical sources have long been recognized to harm the environment owing to their non-biodegradable and non-recyclable composition. On the other hand, microplastics derived from plastic packaging materials are a significant threat in current times for both human and marine life [22].

Biodegradable packaging refers to materials those are broken down and degraded naturally over time, typically through the action of microorganisms such as bacteria and fungi, as well as other environmental variables such as sunshine and moisture [23] and this type of packaging has become a novel interest for preserving fresh fruits and vegetables [24,25]. On the other hand, traditional packaging materials such as plastics can take hundreds of years to disintegrate and frequently contribute to environmental contamination. Biodegradable packaging is designed to be more environment friendly and sustainable compared to traditional packaging options. It can help reduce the amount of waste and decrease the negative impact of packaging on ecosystems and wildlife. Heavy metals those are left as footprints from industrial wastes and dyes from different packaging materials are usually non-bio-degradable [26] and that is why the current world is aggressively approaching towards the sustainable and green processes [27] in place of the passive clean up processes like photocatalytic process, coagulation, extraction processes etc. [19,28].

Eco-friendly packaging produced from renewable resources and natural materials is becoming more popular [29]. In recent years,

the use of eco-friendly, renewable, and sustainable materials has become more important for producing a variety of high-value products with minimum environmental effects. This area of research has gained the attention of many academics and industrial leaders because these materials seem to offer a feasible solution to decline the use of non-renewable resources. Moreover, they aim to minimize environmental pollution, thereby decreasing global warming and the utilization of less energy. Additionally, due to the concern regarding environmental pollution, polyvinyl alcohol-based biodegradable packaging film has led to amplified in packaging research. In this regard, cellulosic nanocrystals and microcrystals emerge as optimal substitutes for synthetic materials and bio-fibers in the context of bio-film formation, owing to their inherent structural characteristics and advantageous properties. The utilization of microcrystalline cellulose fillers in polymer composites results in significant improvements in measures like tensile strength, tensile modulus, and water vapor barrier properties. These findings highlight the effectiveness of MCC in the construction of biofilms [30]. MCC can be extracted from various sources. *Phormium tenax* and *flax* of the Belinka variety were utilized in the extraction of CNC for the preparation of PVA-CNC nanocomposites with enhanced mechanical and thermal properties [31]. However, biofilm preparation, matrix incorporation of MCC and CMC, and film properties were not addressed in the study. The use of cellulose nanocrystals obtained from water hyacinth fiber (WHF) as a reinforcing agent in a polyvinyl alcohol (PVA)-gelatin nanocomposite resulted in the formation of a film with enhanced thermal and mechanical characteristics, but reduced water absorption and WVP [32]. CMC was not incorporated into the film matrix in the investigation. Characterization was performed on microcrystalline celluloses extracted from four agricultural byproducts: sweet sorghum stalk, Jerusalem artichoke stalk, grain stillage, and Chinese herb residue [33]. To investigate the thermal and mechanical properties of the film, the obtained microcrystalline celluloses were composited with polylactic acid as a packaging film. However, neither CMC nor PVA were incorporated into the film matrix.

Based on current understanding, there exists a notable gap in the investigation of the extraction of MCC from banana pseudo-stems for incorporation into a PVA and CMC matrix for the purpose of biofilm development and characterization. It is imperative to acknowledge and rectify this gap to conduct a thorough evaluation of biodegradable packaging obtained from fresh agricultural goods, thereby offering valuable insights into prevailing patterns and future possibilities. In general, the objective of this study is to extract microcrystalline cellulose from banana pseudo-stems, employ it as a reinforcing agent in PVA and CMC matrices, and conduct a comprehensive evaluation of the properties exhibited by the developed biofilm. The intended outcome of this study is the development of eco-friendly and ecologically favourable MCC-based food packaging materials sourced from natural origins.

2. Materials and methods

The study was conducted in the laboratory of Food Engineering and Technology (FET), which is a part of the engineering faculty of Hajee Mohammad Danesh Science and Technology University, Dinajpur. Some of the analyses were conducted at the central laboratory of Jashore University of Science and Technology (JUST) and Sonali Bag Research Laboratory, Bangladesh Jute Mills Corporation Dhaka.

2.1. Materials

Banana pseudo stem from the local market, Ethanol absolute (Merck KGaA), Toluene (Merck KGaA), Sodium hydro-oxide pellets (or pearls) 98% (LOBA CHEMIE PVT. LTD.), Hydrogen Peroxide 30% (perhydrol) (Merck KGaA), Acetic acid (glacial) 100% Food grade (Merck KGaA), Polyvinyl alcohol (LOBA CHEMIE PVT. LTD.), Glycerol (Merck KGaA), Carboxymethyl cellulose (LOBA CHEMIE PVT. LTD.), Distilled water.

2.2. Method of micro cellulose extraction

2.2.1. Raw material preparation

Banana pseudo stems, which were collected from the local market, Basherhat, Dinajpur. The stems were cut into pieces about 3 cm in length and dried under the sun (30 ± 5 °C) until it was ready for further processing. Brown, atrophied pieces were the indication of prepared raw materials.

2.2.2. Purification of the raw materials

The prepared sample (about 4.5 g) was purified by Soxhlet apparatus for 24 h, using the solvent, mixture of ethyl alcohol (135 mL), and toluene (270 mL), at 90 °C temperatures. Hemicellulose, Lignin, and other unwanted materials were removed after purification. The treated sample was dried for 24 h at ambient temperature and pressure.

2.2.3. Extraction process

Cellulose microcrystalline was derived from banana pseudo stem through a dual-stage process. Initially, the pseudo stems were subjected to alkali treatment and bleaching. Subsequently, cellulose pulp underwent hydrolysis. These sequential steps culminated in the acquisition of Cellulose Microcrystalline.

2.2.4. Alkali treatment and bleaching of pseudo stems

The Soxhlet purified material was transferred into an aqueous NaOH (1 M) solution to remove hemicellulose and ash. Then, it was washed with distilled water until it reached the pH of 7. It was stored in the refrigerator for several hours. After that, this solid material was subsequently bleached in aqueous solution containing H_2O_2 (1.3% w/w) and acetic acid (0.1% v/v). These materials were filtered

off and washed again with H_2O until the pH level reached 7. The solids were dried again at ambient temperature and finally subjected to another Soxhlet extraction using ethyl alcohol and toluene mixture at same proportion as mentioned earlier. This material was ready to go for the next steps after drying for 24 h.

2.2.5. Acid hydrolysis of cellulose pulp

The extracted and bleached solids from the previous step were soaked in water and disintegrated in a blender. After blending, the pulp was transferred into a beaker which was placed in an ice-bath, and the pulp was stirred with magnetic stirring until the temperature reached 4 °C. Concentrated sulfuric acid (150 mL) was slowly added while the mixture was being stirred maintaining the mixture temperature below 20 °C. When the addition of sulfuric acid was completed, the mixture was heated at 50 °C. After the hydrolysis of the mixture, the solid was separated by centrifugation at 869*g* force (RCF) for 15 min. This procedure was repeated for several times. The solid remaining after the last centrifugation was dialyzed against H_2O for 7 days. The cellulose dispersion underwent sonication for a duration of 12 h, followed by the removal of H_2O through a 48 h lyophilization process. After the processes, fluffy solids were prevailed (Fig. 1) which was confirmed microcrystalline cellulose later through SEM analysis. The microcrystalline cellulose yield ranged between 16 and 18% in terms of dried banana pseudo-stem across all the runs which reflects the repeatability of the extraction process. A 10 % yield of nano-cellulose extracted from banana pseudo-stem was found in research which is quite comparable with our finding [34].

We encountered a constraint in the extraction process because of the weather conditions in Bangladesh. It was challenging to maintain the ice bath temperature at 4 °C and later at 20 °C during the process before concentrated acid addition, resulting in a temperature fluctuation of ±3 °C. It was difficult and time-consuming to keep the pH at 7 by washing with water, resulting in some losses in microcrystalline cellulose yield.

2.3. Making of composite film

Composite films preparation underwent different steps. Control samples consisted of PVA alone or a combination of PVA and CMC (Table 1). For the PVA control sample, 200 mL of water, 0.75 g of PVA, and 1 mL of glycerol were mixed in a beaker, stirred at 90 °C for 30 min, and then dried in an oven at 45 °C for approximately 16 h. The PVA + MCC film was produced by blending 200 mL of water, 0.75 g of PVA, 0.5 g of microcrystalline cellulose, and 1 mL of glycerol, followed by stirring at 90 °C for 30 min and drying in an oven at 50 °C for about 16 h. PVA 1% samples were prepared using 225 mL of water, 2.25 g of PVA, and 1 mL of glycerol, stirred at 70 °C for 30 min, and then dried in an oven at 50 °C for approximately 16 h. The PVA 1% + MCC film was prepared by combining 225 mL of water, 2.25 g of PVA, 0.5 g of microcrystalline cellulose, and 1.15 mL of glycerol, stirred at 70 °C for 30 min, and dried in an oven at 50 °C for about 16 h. The PVA + CMC control sample included 225 mL of water, 2.25 g of PVA, 1.125 g of carboxymethyl cellulose, and 1.15 mL of glycerol, stirred at 70 °C for 30 min, and dried in an oven at 50 °C for approximately 16 h. Lastly, the PVA + CMC + MCC film was prepared by combining 225 mL of water, 2.25 g of PVA, 1.13 g of carboxymethyl cellulose, 0.5 g of microcrystalline cellulose, and 1.15 mL of glycerol in a beaker, stirring at 70 °C for 30 min, and drying in an oven at 50 °C for about 16 h. All the composite films are shown in Fig. 2.

Bubble formation was prominent at higher mixing temperatures for more concentrated samples compared to S1 and S2, attributed to vortex formation. Therefore, mixing procedures were carried out at 70 °C for 30 min for samples S3 to S6, followed by drying at 50 °C for 16 h, which was believed to be the optimal condition for our research. There was a limitation at our facility with respect to this film deposition process that we didn't have a vacuum chamber to avoid the mentioned bubble formation during the ingredients

Fig. 1. Microcrystalline Cellulose Powder obtained after 12 h of sonication and 48 h of lyophilization.

Table 1
Formulation of Composite film.

Formulated Composite Films	Film's Code	PVA (g)	CMC (g)	MCC (g)
PVA 0.3% Control	S1	0.75		
PVA 0.3% + MCC	S2	0.75		0.5
PVA 1% Control	S3	2.25		
PVA 1% + MCC	S4	2.25		0.5
PVA + CMC Control	S5	2.25	1.125	
PVA + CMC + MCC	S6	2.25	1.125	0.5

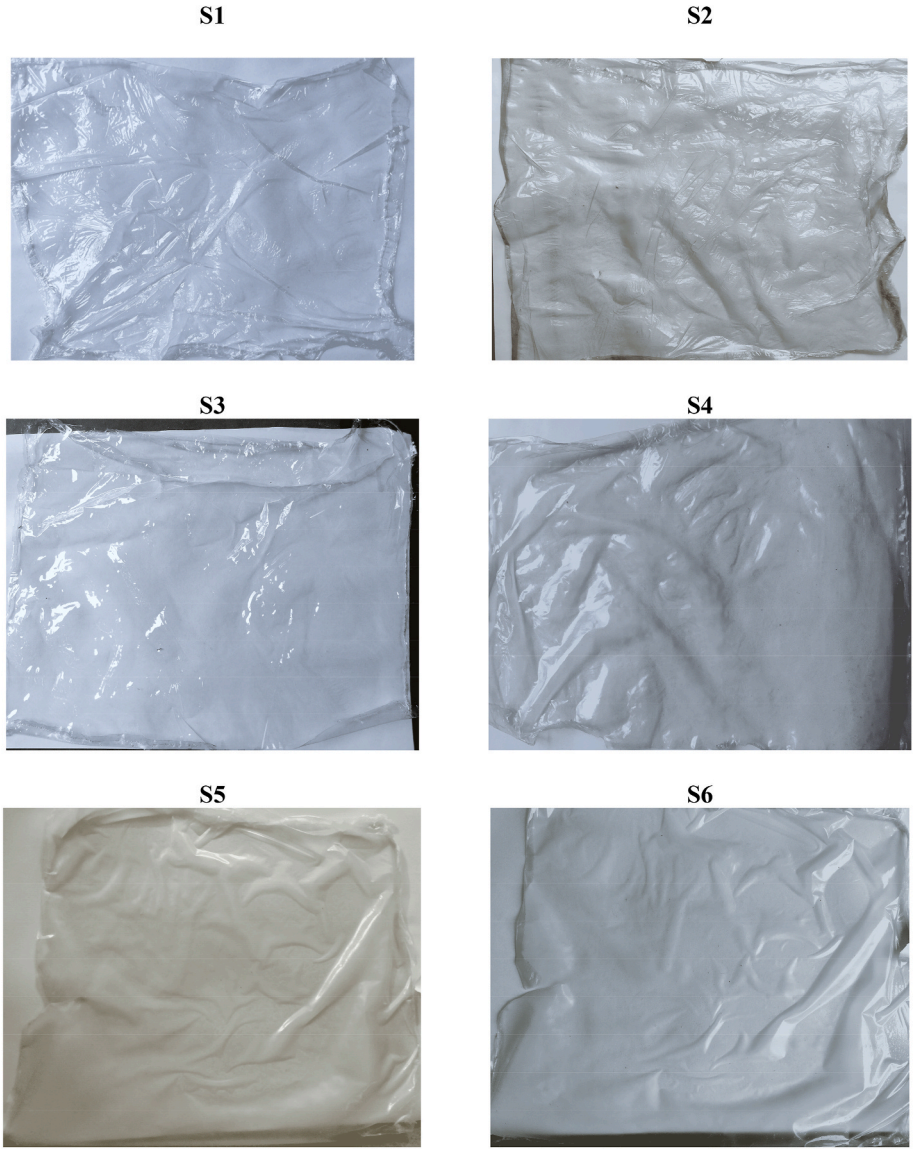

Fig. 2. Formulated composite films after final drying for 16 h at varied temperatures (S1 at 45 °C and S2 – S6 at 50 °C).

(PVA, CMC and MCC) mixing process.

2.4. Characterization of the extracted MCC powder and formulated composite films

The extracted MCC powder and formulated biofilms were characterized by different sophisticated analyses such as SEM, FTIR, WVP content, Light-Transparency and Transmittance, Water Content, Water Solubility, and Swelling Degree.

2.4.1. Scanning electron microscope (SEM) analysis of the MCC powder

A SEM (TESCAN MIRA3, Shanghai, China) was used to examine the MCC powder. On a double-sided piece of conductive tape, MCC powder was spread out. The MCC powder was coated for 60 s with a gold sprayer using a 10-mA spraying current to disperse static electricity. At an accelerating voltage of 5 kV, SEM pictures were captured.

2.4.2. Fourier transform infrared spectroscopy (FTIR) analysis of the composite films

Utilizing the FTIR (PerkinElmer, Spectrum Two, Germany) instrument, the bonding characterization of the functional group of the films was performed. To follow the development of the polymer bond, the results were examined throughout a scanning range of 4000 to 400 cm^{-1}.

2.4.3. Mechanical properties of the composite films

By inserting around 50 mm of each film between the grips of a Zwick/Roell (model Z010) testing device, the tensile strength and percent elongation of the prepared films were measured. The tensile modulus speed during the test was 12.5 mm/min, with a continuous test speed of 125 mm/min.

2.4.4. Water content, water solubility, and swelling degree of the composite films

The formulated biofilms' water content, solubility, and swelling level were assessed using the accepted methods [35]. Each film was trimmed and precisely weighed (M_1) at 0.001 g. To determine the initial dry matter content and record the weight (M_2) of the dried films, the films were then dried in a cabinet drier for 24 h at 105 °C. Each film was then placed in distilled water (50 mL) and left to soak for 24 h at room temperature. With the use of a spatula, the undissolved films were collected and spread out on the filter to dry, and their weights were noted (M_3). The undissolved films were then dried for 24 h at 105 °C to determine the sample's ultimate dry weight (M_4). Using the following equations (1)–(3), the water content, solubility, and swelling level were computed.

$$Water\ content\ (\%) = \frac{M_1 - M_2}{M_1} \times 100 \tag{1}$$

$$Solubility\ (\%) = \frac{M_2 - M_4}{M_2} \times 100 \tag{2}$$

$$Swelling\ degree\ (\%) = \frac{M_3 - M_2}{M_2} \times 100 \tag{3}$$

2.4.5. Water vapor permeability (WVP) of the composite films

The water vapor permeability was assessed according to JIS Z0208, using a gravimetric method. To obtain 0% relative humidity, 7 g of anhydrous calcium chloride was added to the compact-size vials. The films, which were cut into circles with a diameter of 5 mm, were used to hermetically seal the vials. The vials were put in a desiccator that was soaked with a magnesium nitrate solution to keep the relative humidity (RH) 51% at 25 °C. The interior conditions of the desiccator were observed using a digital temperature and humidity meter. The slope up to three days was calculated to track the beakers' weight gain at 24 h intervals. The water vapor transmission rate (WVTR) was calculated using equation (4):

$$WVTR = \frac{W/T}{A} \tag{4}$$

Where W/t represents slope (weight change (g) versus time (hour) graph), and A represents the area of vapor transmission of the films (78.54 mm^2). Finally, water vapor permeability was measured by following equation (5):

$$WVP = \frac{WVTR/h}{p_1 - p_2} \tag{5}$$

Where, h represents the thickness of the films (mm), P_1 represents the partial pressure of water vapor in the desiccator at 51% RH (21.64 $\times 10^5$ Pa) and P_2 represents the vials' partial pressure of water vapor at 0% RH (0 Pa).

2.4.6. Light transmittance and transparency analysis of the composite films

Film samples were sliced into 10 × 40 mm strips and inserted into the cuvette cell of the Shimadzu UV-1800 UV/VIS spectrophotometer (Japan). At wavelengths spanning from 200 to 700 nm, the barrier qualities of the films were evaluated. A process for measuring the transparency and light transmission of film samples following equation (6) below [36]:

$$\%Transmittance = Antilog\ (2 - Absorbance) \tag{6}$$

$$Transparency = \frac{A_{600}}{x} \tag{7}$$

Where A_{600} stands for the film sample's absorbance at 600 nm and x represents the average thickness of the composite films measured in mm. According to this equation, there is a direct correlation between transparency and degree of opacity.

2.5. Statistical analysis

The statistical analysis was done using SPSS software (IBM SPSS Statistics 25). Three replications of each analysis were performed. Each analysis was calculated using means and standard deviations. Analysis of variance (ANOVA) and DMRT were used to compare the variance between mean values, with an acceptable threshold for significance of 5%.

3. Results and discussion

3.1. Scanning electron microscope (SEM) analysis of extracted MCC

The intricate details of the microcrystalline cellulose (MCC) extracted are revealed in Figs. 3–6. The MCC exhibits a particle character that is clearly visible. Despite having non-isometric forms, SEM analysis shows a flattened morphology with a convoluted ribbon-like structure, comparable to the observations made by Das [37]. The particle dimensions range from 14 μm to 350 μm, aligning partly with the studies conducted by Ventura-Cruz [38], Perumal [39], Shah [40], and Mohammadinejad [41]. It is to be mentioned that the particles did not enter the nano region due to the limited breakdown of cellulose chains, as reported by Awual & Hasan [42], Hasan [15] and Salman [43] where the molecular chain lengths ranged between 100 and 200 nm, 50–200 nm and 50–100 nm respectively. The aspect ratio (length/diameter) of the particles spans from 1.5 to 7.5. Following acid hydrolysis, the amorphous portion of MCCs is pierced, disrupting intermolecular clumping and revealing a more loosely bundled arrangement of tiny fibers, as elucidated by Ren [33]. The accompanying figure underscores distinctive and uniform fibers, consistent with spectroscopic evidence indicating the removal of lignin and hemicellulose, as observed by Ventura-Cruz [38] and Mohamad Haafiz [44]. The inherent roughness of MCC facilitates hydrolysis, contributing to the production of microcrystals with diverse shapes and overall characteristics, as highlighted by Ren [45] for the films with 5% MCC inclusions.

3.2. FT-IR analysis of the composite films

Fourier transform infrared spectroscopy (FTIR) analysis was conducted to analyse the chemical structural interactions within the composite films composed of Microcrystalline Cellulose (MCC) reinforcing the control films made with Polyvinyl Alcohol (PVA) and Carboxymethyl Cellulose (CMC) [46–48]. The broad spectrum observed at 3280 cm^{-1} signifies terminal (vinyl) C–H stretch and hydroxy group H-bonded OH stretch, confirming the presence of PVOH and OH functional groups of micro cellulose. The presence of hydroxyl (-OH) groups within cellulose facilitates reinforcement mechanisms through interactions with the polymer matrix [21]. The absorption band between 2917 cm^{-1} and 2941 cm^{-1} represents methylene C–H asymmetric/symmetric stretch (Fig. 7). In the composite films, broad-spectrum bands within the range of 1200 cm^{-1} to 800 cm^{-1} indicate the carbohydrate fingerprint region which is comparable with the findings of a study on liquid phase exfoliation of metal–organic framework to reduce CO_2 in quaternary NH_4OH solution where the spectrum ranged between 1300 cm^{-1} to 1500 cm^{-1} [49], with concurrent observations of cyclohexane ring vibrations between 1030 cm^{-1} and 1042 cm^{-1} [50–54]. Given the similarities in the chemical structure of Carboxymethyl Cellulose (CMC) and Cellulose Nanocrystals (CNC) and the relatively low content of CNC in each composite film, the FTIR spectra of PVA + CMC

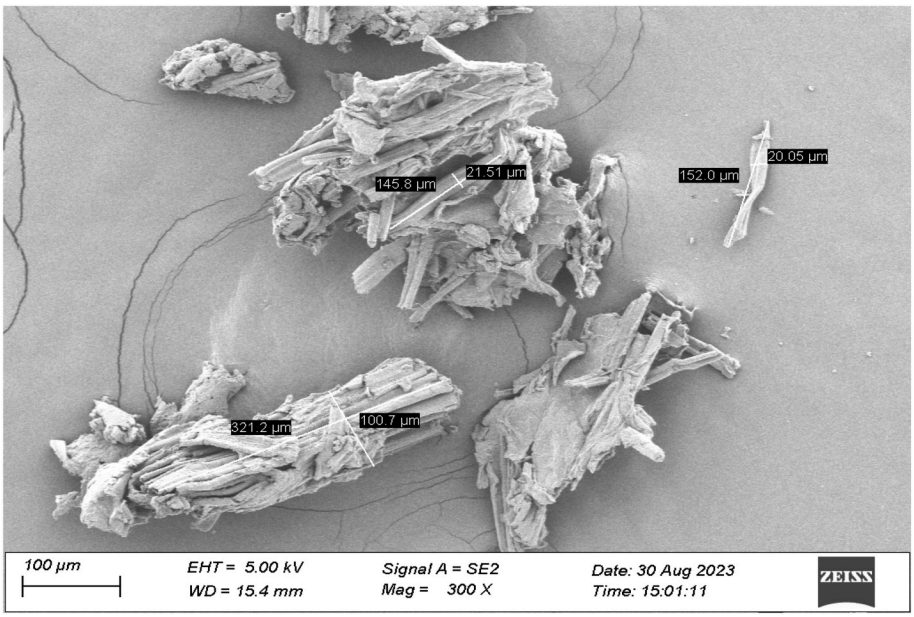

Fig. 3. Surface sectional view of micro cellulose fibers at 300× magnification.

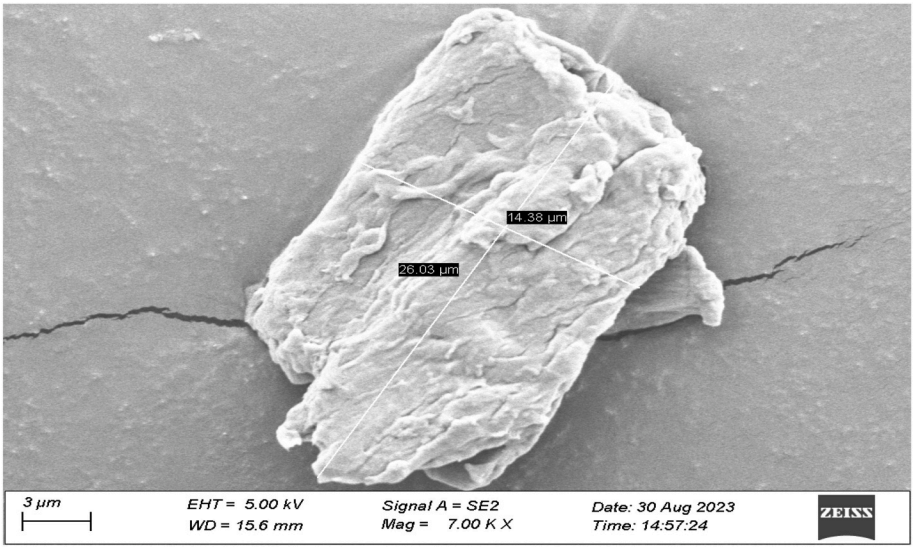

Fig. 4. Particle size measurement of cellulose fiber (length - 26.08 µm and diameter - 14.38 µm) by surface sectional view at 7000× magnification.

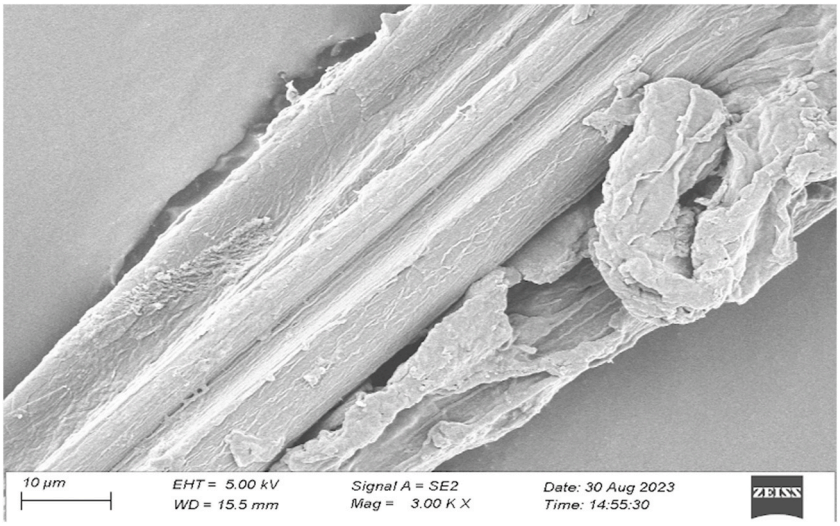

Fig. 5. Surface sectional view of single particle cellulose at 3000× magnification.

control films and PVA + CMC + CNC films exhibit nearly identical characteristics [55]. However, any discernible changes in absorption bands within the FTIR spectrum with the introduction of specific components suggest interactions between those components [35]. A noteworthy observation is the slight reduction in the broad spectrum at 3280 cm^{-1} in PVA control films compared to CNC-incorporated PVA films. This discrepancy may arise from interactions between the hydroxyl groups of CNC and those of the PVA matrix. The reduction in polyvinyl alcohol (PVA) band intensity that is observed upon the addition of microcrystalline cellulose (MCC) suggests that PVA's propensity for crystallization has been lessened. This could be due to the formation of intermolecular complexes that are aided by hydrogen bonding interactions between MCC and PVA [56]. Comparable finding has been observed where the C–H asymmetric/symmetric stretch spectrum ranged between 3273 and 330 cm^{-1} in a chitosan coated toxic dye encapsulation study [17]. The similar findings have been reported in previous studies [57–59].

3.3. Mechanical properties of the formulated composite films

The tensile strength and % elongation of the formulated composite films was tested to examine the effect of mechanical properties after the addition of MCC on PVA as well as PVA and CMC matrix. The tensile strength of the control films was significantly lower than the MCC-added films. Fig. 8 showed that sample S6 had the highest tensile strength properties of the other films as it was formulated with the addition of both CMC and MCC on the PVA matrix. The tensile strength of the MCC-added films was stepped up from 2.51

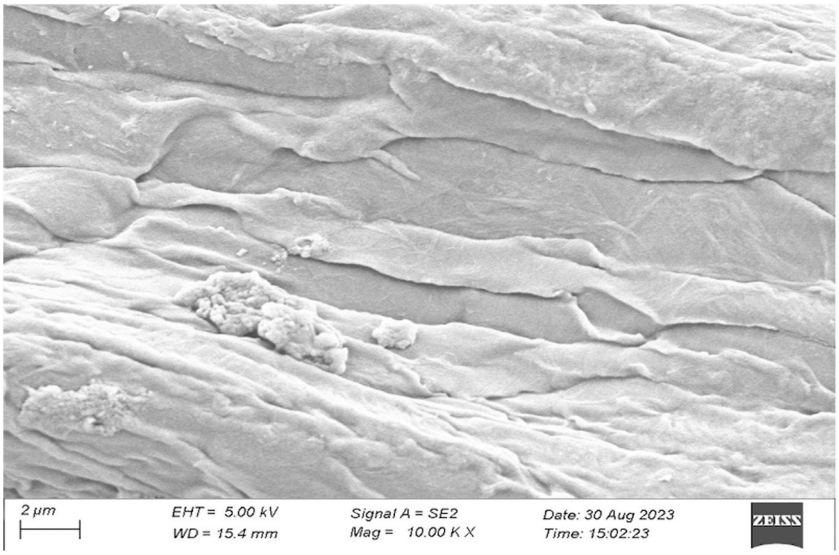

Fig. 6. Surface sectional view of single particle cellulose at 10,000× magnification.

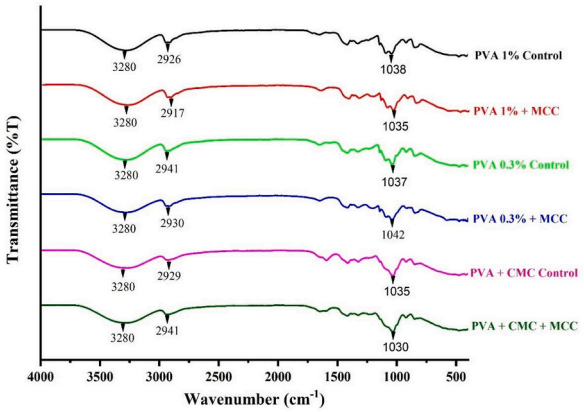

Fig. 7. Fourier Transform Infrared Spectrogram of the prepared composite biopolymer films.

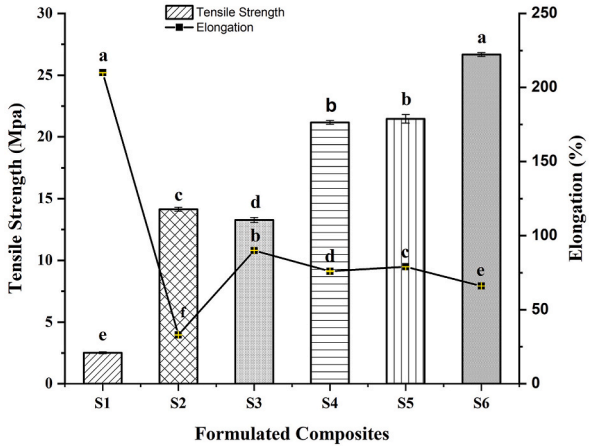

Fig. 8. Mechanical properties (Tensile Strength, Mpa and Elongation, %) of the formulated composite films.

MPa, 13.27 MPa, and 21.46 MPa–14.13 MPa, 21.16 MPa, and 26.66 MPa respectively. The dispersion of natural polymers in films ensures inter-molecular bonding of biopolymer with the other substances in the film, which is the main reason for higher tensile strength. Hydrogen bonding, hydrophobic bonding, and other bond formation in biopolymer films improved compound compatibility [60]. Similar phenomenon was observed in the study of 5% sweet sorghum stalk and Distiller's grain MCC incorporation where the tensile strength was measured about 37 MPa for both the scenarios [45]. The reason behind the accelerated tensile strength could be the complex bonding of the molecules and absence of CMC in composition matrix. On the other hand, percent (%) elongation showed different results than usual. S3 displayed significantly higher % elongation than other composite films (Fig. 8). The higher the % elongation the higher the deformation quality of the films. Elongation of the S4 was 76% whereas S5 and S6 were 79% and 66%, respectively. The presence of CMC abridged the % elongation of the CMC-added films. CMC has some limitations including low elongation at break, exceptionally stiff behaviour, and loss of electrochemical consistency [61]. In the current study, it was found that % elongation was drastically reduced after the addition of MCC at S2 film, but it increased when the percent ratio of PVA was introduced at S4 as shown in (Fig. 8). The reduction in elongation at break caused by the insertion of fibers in the polymer is a usual pattern observed in thermoplastic composites when the incorporation of stiff reinforcements generates stress concentrations [62,63]. The same phenomenon was observed by the author Calvino [64] and Fortunati [59].

3.4. Water content, solubility, and swelling degree of the formulated composites films

Water content, solubility, and swelling degree of the formulated composite films were carried out to identify the films' ability to absorb and retain water, as well as their stability and durability. Fig. 9 depicts that the water content of the MCC-added films was higher except for the one that had CMC in it. The water content of the S1 sample was 43%, in contrast, the water content of S2 was 93%. These drastic changes in water content after the addition of MCC could be because of an increase in a hydrophilic compound in the PVA matrix or it could be the reason for poor interaction of MCC with the small amount of PVA in S2 film. But for the case of the higher amount of PVA in S3 and S4 films, the change of water content was not so high which is quite comparable with the work of Kanatt & Makwana [65], Riaz [66] and Akhtar [67] where the films were dried at higher temperatures. It indicates that there is a proper interaction of MCC with a higher amount of PVA. When CMC was added to the PVA-MCC matrix, a slight decrease in water content was observed. This could be due to the hydrophobic nature of CMC.

The solubility of the films was observed to decrease when MCC was added (Fig. 10). The solubility of the S1 control sample was 16% whereas the solubility of MCC incorporated S2 sample was 9%. In the case of the higher percentage of PVA, the same phenomenon was observed. PVA is hydrophilic in nature but soluble in water [46,47]. PVOH is extremely hydrophilic and dissolves in water, facilitating hydrolysis. Furthermore, the hydroxyl groups in PVA will likely produce hydrogen bonds and acetyl connections with other materials like cellulose and aldehydes. The hydrogen bonds between OH groups are critical in influencing the water solubility of PVA and PVA films [68]. The decrease in the solubility in water of the films with increasing E-MCC concentration suggests that E-MCC enhanced the crystallinity of the films [21]. The same phenomenon was observed by Debnath [21], Patel & Joshi [68] for the case of the incorporation of cellulose nanofiber in the PVA matrix and the addition of MCC in starch respectively.

S3 formulated film swelled at 67%, which is the highest among all (Fig. 11). It is also visible that the swelling degree of S4 film drastically reduced when MCC was introduced in the S3 formulation. The lowest swelling degree belonged to S6 film, which swelled at 2%. Chemical interaction and stability made the S6 film swell at its lowest. In another study, CMC-PVA and ZnO Nanoparticles, the CMC-PVA-ZnO-Nps film showed less swelling, and the films were compact and tighter because of the intermolecular interaction [69]. In the case of S1 and S2 formulated films, the interconnection between PVA and MCC might not be well established. Therefore, it showed contradictory results from the rest. Biopolymer films were kept from swelling due to the network of connected biopolymers and other substances [70]. Akhter [71] also observed compatible results in the case of bacterial nanocellulose-induced biopolymer

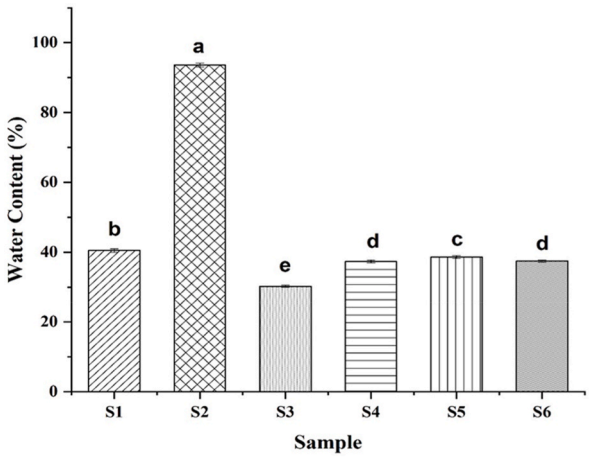

Fig. 9. Water content of the formulated composite films.

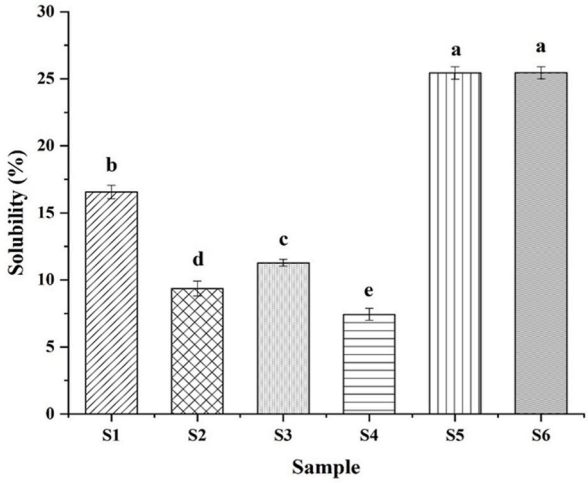

Fig. 10. Solubility behaviour of the formulated composite films.

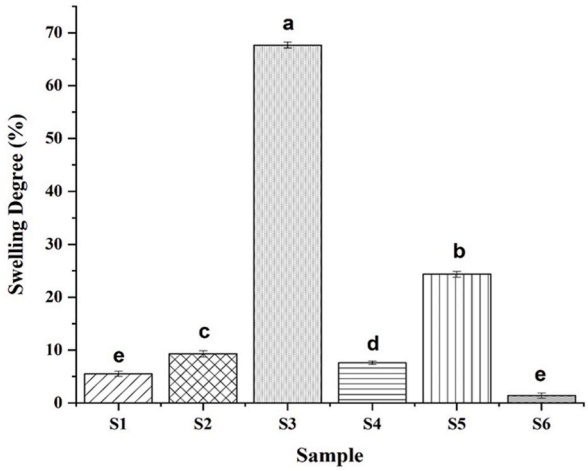

Fig. 11. Swelling degree of the formulated composite films.

film.

3.5. *Water vapor permeability (WVP) of the formulated composite films*

Water vapor permeability is important for evaluating the barrier properties of the formulated composite films. The barrier property of the biopolymer packaging film increases with a decrease in WVP content. Table 2 depicts that S4 (PVA 1% + MCC) had the lowest and S3 (PVA 1% control film) had the highest WVP content which means S4 film has great barrier properties as a biopolymer film rather than other films. There is a decline in WVP with the presence of MCC content inside the films. The permeability of the moisture decreased as the presence of MCC filler was introduced because the permeation action was impeded by the presence of impervious MCC, which caused rigidity in the film matrices [58,64,66]. An increase in filler material PVA increased the film's WVP content. This could be the reason for the amount of hydroxyl group of PVA in the film matrix. Besides, filler at low concentrations is usually effectively disseminated in the film matrix, providing a tortuous path that minimizes WVP, whereas filler at high concentrations usually results in the formation of agglomeration, which favours water permeability [72]. A similar pattern of results in terms of WVP was observed by Riaz [66] with the incorporation of Chinese chives to a CMC based food packaging film, Hermawan [73] with the incorporation of MCC in seaweed-based film, the addition of MCC in Tapioca starch film by Othman [72].

3.6. *Light-Transparency and Transmittance analysis of the formulated composite films*

Table 3 shows that MCC-incorporated films had lower transparency than the control films. Transparency values of 3.546 for S2, 1.500 for S4, and 5.178 for S6 at 600 nm were noted. This demonstrates that biopolymer film were less transparent and were difficult to

Table 2
Water vapor permeability of the composite films.

Film Code	Formulated Composites	Water Vapor Permeability (10^{-9} g/Pa h m)
S1	PVA 0.3% Control	1.100 ± 0.001^{c}
S2	PVA 0.3% + MCC	0.927 ± 0.015^{c}
S3	PVA 1% Control	3.290 ± 0.100^{a}
S4	PVA 1% + MCC	0.223 ± 0.020^{d}
S5	PVA + CMC Control	1.326 ± 0.152^{b}
S6	PVA + CMC + MCC	1.320 ± 0.200^{b}

Results are expressed as mean ± standard deviation.
Differences in letters (a-e) in the same column indicate significant differences at $p < 0.05$ among all the films.

Table 3
Light-transparency of the composite films.

Film's Code	Formulated Composites	Light-Transparency at 600 nm
S1	PVA 0.3% Control	5.333 ± 0.152^{b}
S2	PVA 0.3% + MCC	3.546 ± 0.205^{c}
S3	PVA 1% Control	2.050 ± 0.035^{d}
S4	PVA 1% + MCC	1.500 ± 0.251^{e}
S5	PVA + CMC Control	13.514 ± 0.335^{a}
S6	PVA + CMC + MCC	5.178 ± 0.033^{b}

Results are expressed as mean ± standard deviation.
Differences in letters (a-e) in the same column indicate significant differences at $p < 0.05$ among all the films.

see. The transparency of the films not only offers insight into the particle size but also the degree of distribution of the fillers inside the matrix [74]. Opacity (contrast to transparency) measurement is important in film applications, particularly in food packaging, because it controls the absorption of sunlight, fluorescent light along candent light through the films; such absorption can cause degradation, resulting in discoloration and nutrient loss due to photo degradation of the food [75]. The decreased transparency caused by the inclusion of MCC particles was most likely caused by the broad reflection of light by the MCC particles. Because the size of the particles of MCC is larger beyond the wavelength of light, one would anticipate MCC to scatter light [73].

The films also showed UV screening properties. The films had higher transparency percent at the visible region (400 nm–700 nm) but lower at the UV region (200 nm–400 nm). It is seen from Fig. 12 that S6, which contains CMC in it, had the lowest transmittance among the films. Other MCC-induced films also had more depressed transmittance than control films. Cano [76] observed that, in the visible wavelength spectrum, the control films had transmittance values above 90%, while in the UV range, the values were substantially lower when induced CNC in Starch-PVA blend. Both agar/NC and agar/MCC composite films showed a similar trend of decreasing transmittance at 280 nm (T280), which suggests transmission or blocking UV-light. The loss in the transparency of the composite film is mostly due to the crystalline cellulose limiting light passage through the film [77]. A similar trend was observed by Naduparambath [78] when introducing MCC in the PVA matrix, the author also observed a decreasing effect of transmittance while

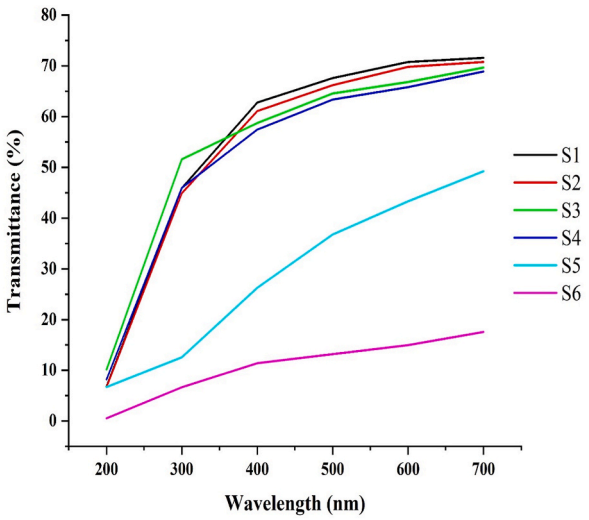

Fig. 12. Light transmittance property at a wavelength of 200–700 nm of the formulated composite films.

increasing MCC concentration in the film's matrix.

4. Conclusions

This research has demonstrated the feasibility and potential of utilizing banana pseudo-stem as an economical and readily available source to produce biodegradable microcrystalline cellulose crystals films. The scanning electron microscopy analysis provided valuable insights into the abundant MCC fiber content, highlighting the efficacy of the extraction process from banana pseudo-stem. Incorporating MCC into both PVA and PVA-CMC matrices was the novel part in this research which exhibited promising outcomes for biofilm preparation. Notably, the addition of MCC led to reduced solubility and swelling of the films, while enhancing tensile strength and decreasing water vapor permeability content. Moreover, the films demonstrated improved light barrier properties with MCC inclusion. Additionally, higher concentrations of polyvinyl alcohol positively influenced the material properties, highlighting a pivotal parameter for optimization. The interaction observed between the binding material and MCC, as revealed by FTIR analysis, further underscores the compatibility and potential synergistic effects in enhancing the biofilm properties. Particularly, the incorporation of carboxymethyl cellulose in the biofilm matrix emerged as a significant advancement, especially in the context of food packaging applications. In culmination, the biopolymer films incorporated with MCC and CMC present a compelling case as a sustainable alternative to conventional non-biodegradable packaging materials. This study strongly suggests for the adoption of MCC as a reinforcement material in environment friendly packaging, anticipating outstanding results. Looking ahead, future investigations should focus on essential aspects such as the shelf-life assessment of perishable products and the evaluation of food-grade biodegradability for the prepared composite film and commercialization. These critical endeavours will contribute invaluable knowledge towards realizing the full potential of this innovative sustainable packaging material in the near future.

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Data availability

The data associated with this study has not been deposited into a publicly available repository. Most of the data are embedded in the manuscript and if any additional data is required, it will be made available on request.

CRediT authorship contribution statement

Ishmam Haque Sachcha: Writing – original draft, Methodology, Investigation, Formal analysis, Software. **Kushal Paddar:** Resources, Methodology, Investigation. **Minhajul Matin Minar:** Visualization, Validation, Resources. **Latifur Rahman:** Investigation, Data curation. **S.M. Kamrul Hasan:** Writing – review & editing, Visualization, Validation. **Md Akhtaruzzaman:** Formal analysis, Data curation. **Mir Tuhin Billah:** Project administration, Conceptualization, Writing – review & editing, Validation, Software, Supervision. **Sabina Yasmin:** Project administration, Conceptualization, Writing – review & editing, Validation, Software, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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