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Sustainable Polyvinyl Alcohol/Chitosan Biocomposite Films Reinforced With Crystalline Microcellulose Extracted From Waste Notebook Paper

Vu Viet Linh Nguyen[1], **Minh Khanh Hao Phung**[1], **Thai Hoa Nguyen**[2], **Vinh Tien Nguyen**[1], **Van Quy Nguyen**[1*]

[1]*Ho Chi Minh City University of Technology and Engineering, Vietnam*
[2]*Ho Chi Minh City University of Technology, VNU-HCM, Vietnam*

[*]Corresponding author. *Email: quynv@hcmute.edu.vn. Phone number:+84-374910094*

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ABSTRACT

The micro composite film based on crystalline cellulose and biodegradable polymers has attracted tremendous attention because of its numerous applications in food preservation, medicine, and water purification. This study develops sustainable biocomposite films composed of polyvinyl alcohol (PVA) and chitosan (CS), reinforced with crystalline microcellulose (CMCs) extracted from used notebook paper. After bleaching and acid hydrolysis, CMCs were obtained with a yield of 35.3%, an average size of 615.15 ± 6.7 nm, and the crystallinity index (CrI) of 53.1% was determined by dynamic light scattering (DLS) and X-ray diffraction (XRD), respectively. The extracted CMCs were incorporated into the PVA and CS matrix with different concentrations of 0, 3, and 6 wt.% using a solution casting-evaporating technique to fabricate the PVA/CS@CMC composites. Photographs, SEM images, and transmittance measurements show that the transparency of these biocomposite films decreases when increasing the CMC content. The FTIR spectra indicate that these biocomposite films simultaneously present the characteristic peaks of CMCs and the PVA/CS matrix polymer. In addition, these biocomposite films display a typical crystalline peak of PVA at 2θ of 19.76° and two characteristic cellulose I peaks of CMCs at 2θ of approximately 14° and 16–17°. Moreover, the composite film with 3 wt.% CMCs exhibits the highest value in the tensile strength. This study suggests that the CMCs derived from used notebook paper can be used as an effective reinforcing material for the PVA/CS blends, offering promising applications in packaging and related fields.

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1. Introduction

The growing world population has led to an increase in the demand for daily paper products in printing, copying, packaging, decoration, and hygiene [1]. After use, waste paper, primarily office paper (from printers and photocopiers), old magazines, and sludge paper (a by-product of the paper production process) will be either landfilled or recycled. In 2020, only 59.7% of total consumer paper was recycled worldwide, mostly in Europe (73.3%) and North America (68%) [2]. Notably, waste paper ranks first among the most recycled packaging materials in Europe (82%), far surpassing other materials such as metal (77.4%), glass (75.4%), and plastic (40.6%) [2]. This data further demonstrates the great recyclability and reuse potential of waste paper. About 40.3% of total consumer paper is called waste paper (WP) and is hardly used or disposed of inappropriately, leading to global environmental problems [3]. This poses an urgent need to find alternative solutions to increase the reuse or recycling of waste paper. Several studies have been conducted to utilize waste paper for the production of valuable compounds such as bioethanol [4] and activated carbon, as well as fillers in polymeric composites [5].

One of the emerging approaches is to isolate fibers, including crystalline microcelluloses (CMCs), from waste paper [5]. In fact, CMCs extracted from waste paper can contribute to the development of biomaterials, thereby promoting cost-effective and sustainable production methods. Sustainable biomaterials are created using environmentally sustainable technology or derived from diverse sources of life [6]. Several biomaterials have been discovered and fabricated as potential substitutes for traditional substances, and many of them have been successfully utilized in several biomedical domains, including infilter and membrane technology, energy-related products, packaging components, and fabrics [7]. Biocomposite films are a potential alternative to traditional plastic materials, with characteristics such as sustainability and biodegradability.

Chitosan (CS) and polyvinyl alcohol (PVA) are popular in biocomposite films due to their biodegradability and non-toxicity [8]. Chitosan is a natural biopolymer that possesses antimicrobial and antioxidant characteristics and a bifunctional nature, regarded as one of the most essential and cost-effective biopolymers used in various fields, including food preservation, medicine, and water purification [9]. However, compared to conventional plastics, chitosan still has many limitations, such as high water swelling, low water vapor resistance, elongation, and toughness [10], [11]. For these drawbacks, chitosan was usually combined with other biodegradable polymers, such as poly(vinyl pyrrolidone), poly(lactic-co-glycolic acid), poly(lactic acid), poly(caprolactone), poly(allylamine hydrochloride), and poly(vinyl alcohol) (PVA). Among them, PVA is the most used because it is a non-toxic polymer with chemical resistance, good flexibility, and foldable film-making properties [12], [13]. All these characteristics make it a suitable polymer for the preparation of flexible membranes for food packages and controlled drug delivery systems [14]. The combination of CS and PVA could partially enhance the high water swelling, elongation, toughness, and film-forming characteristics. In addition, these properties can be further improved by adding reinforced agents such as crystalline microcellulose (CMC).

CMC is a renewable, biodegradable micro material derived from cellulose, offering exceptional mechanical strength, such as a high elastic modulus (110–220 GPa), tensile strength (7.5–7.7 GPa), customizable aspect ratios, a large specific surface area, easy surface performance, adaptable crystallization, an essential quantity of polymerization, and excellent chemical resistance [15]. Many studies have indicated the usefulness of CMCs in enhancing biocomposites' tensile and thermal properties [16]. Therefore, CMCs extracted from paper and various other raw materials are useful in micro-biocomposites [17], [18]. Micro-biocomposites represent a fascinating group of hybrid materials consisting of a bio-based matrix combined with micro-sized fillers [10]. Combining micromaterials and environmentally friendly biopolymers is a creative approach that helps enhance their properties, including being lightweight, environmentally friendly, and recyclable [19].

Herein, we focused on extracting and isolating CMCs from a low-cost cellulose source in waste notebook paper, which is then incorporated into a biodegradable PVA/CS polymer blend to develop sustainable PVA/CS@CMC biocomposite films through a simple and green fabrication method. The properties of prepared CMCs and biocomposite films, such as morphological, transmittance, swelling rate, and mechanical characteristics, were determined.

2. Materials and Methods

2.1. Materials

The used notebook paper was collected at Ho Chi Minh City University of Technology and Engineering (HCM-UTE), Ho Chi Minh City, Vietnam. CMCs were provided by Dr. Vu Viet Linh Nguyen and prepared according to the scientific publication accepted for publication in the Industry and Trade journal (ISSN: 0866 – 7756) on October 7, 2025. The properties of CMCs, such as isolation efficiency of 35.3%, average particle size of 615.15 ± 6.7 nm, and crystallinity index (CrI) of 53.1% were determined by dynamic light scattering (DLS) and X-ray diffraction (XRD) methods, respectively. Polyvinyl alcohol (PVA, M_w = 60–125 kDa) and chitosan (CS, degree of deacetylation of 75%) were purchased from HiMedia Laboratories Pvt. Ltd. (India).

2.2. Preparation of PVA/CS@CMC biocomposite films

PVA/CS composite films were prepared by mixing PVA and CS in a weight ratio of 8:2. A 15 wt.% PVA solution and 2 wt.%. CS solution were prepared separately, then combined and stirred at 80 °C for 1 h to achieve a homogeneous blend. CMCs were weighed with an appropriate amount of 3 and 6 wt.% of matrix polymer weight and dispersed in distilled water (100 mg/mL) and stirred at room temperature for 1 h, followed by sonication to ensure a uniform dispersion. The CMC dispersion was then added to the PVA/CS mixture under continuous stirring to create PVA/CS@CMC solutions. After mixing, the PVA/CS@CMC solutions were filtered to remove residual impurities or aggregates. Finally, the filtered solutions were poured into the molds and dried at 25 °C for 48 h to form the composite films. The prepared PVA/CS@CMC composite films were obtained by the solution casting-evaporation method with different CMC concentrations of 0, 3, and 6 wt.%.

2.3. Characterizations of the materials

Surface morphology of PVA/CS@CMC biocomposite films was analyzed using scanning electron microscopy (SEM, JEOL JCM-7000, Japan) after sputter-coating with platinum. Imaging was conducted at an accelerating voltage of 5 kV.

The crystallinity of used notebook paper was characterized using an X-ray diffractometer (D8 Advance, Bruker, USA). Radial scans of intensity were captured at an ambient condition over a diffraction 2θ angles from 5º to 60º (step size = 0.02º) using a Ni-filtered Cu Kα radiation source (λ = 1.54 Å), an operating voltage of 45 kV.

The chemical composition of the PVA/CS@CMC film was identified and analyzed by using an FTIR spectrometer (FTIR-4700, Jasco Inc., Japan). The spectra were recorded in the wavenumber range of 4000–400 cm^{-1} under the attenuated total reflection (ATR) mode with a spectral resolution of 2 cm^{-1} and a scanning speed of 2 mm/sec at 25 ºC.

The tensile properties of films were measured using an MTC-500 testing machine (PTA Group, Spain) following the ASTM D882-18 standard [20]. The tensile specimens were cut in rectangular shapes with dimensions of 100 mm in length and 15 mm in width at a crosshead speed of 50 mm/min. The thickness of each specimen was determined as the average of ten random measurements, and at least five specimens were measured for each sample.

The optical transmittance of the biocomposite films was evaluated using a UV–Vis spectrophotometer (Jasco V–73, Japan) in the wavelength range of 700–300 nm. Film samples were placed directly in the test cell, with air used as the reference.

The film specimens (1 cm × 1 cm) were weighed (m_0) using an analytical balance, immersed in distilled water for 20 min, blotted with filter paper, and reweighed accurately (m_1). The swelling index (X) of the film was calculated using equation (1) [21]:

$$\mathrm{X} = \frac{\mathrm{m_1 - m_0}}{m_0} \times 100\% \tag{1}$$

3. Results and Discussion

3.1. Preparation of PVA/CS@CMC composite film

The isolated CMCs have a yield of 35.3%, a hydrodynamic diameter of 615.15 ± 6.7 nm, and the crystallinity index (CrI) of 53.1% was sequentially examined by dynamic light scattering (DLS) and X-ray diffraction (XRD). Subsequently, these CMCs were dispersed into the PVA/CS mixture with varying CMC contents of 0, 3, and 6 wt.% to form PVA/CS@CMC composite films. Firstly, the optical transparency of PVA/CS@CMC composite films varies with CMCs content, as shown in Figure 1a–c. The 0 wt.% CMC film (Figure 1a) had the highest transparency, making the logo clear and recognizable. The increment up to 3 wt.% CMCs (Figure 1b) slightly reduced transparency, while at 6 wt.% CMCs (Figure 1c), transparency declined significantly, resulting in blurred images. This reduction is due to increased light scattering from the CMC particles, leading to a more heterogeneous structure. Thus, while CMCs can improve certain functional properties, they negatively affect optical transparency. Figure 1d–f present scanning electron microscopy images of PVA/CS@CMC biocomposite films containing 0 wt.%, 3 wt.%, and 6 wt.% CMCs, all viewed at a magnification of 500×. Each film exhibits smooth, flat, and uniform surfaces, suggesting consistent film formation and an even distribution of CMCs throughout the matrix. The absence of visible phase separation indicates good mixing between CMCs and the PVA/CS matrix, which helps maintain the structural integrity of the biocomposite films.

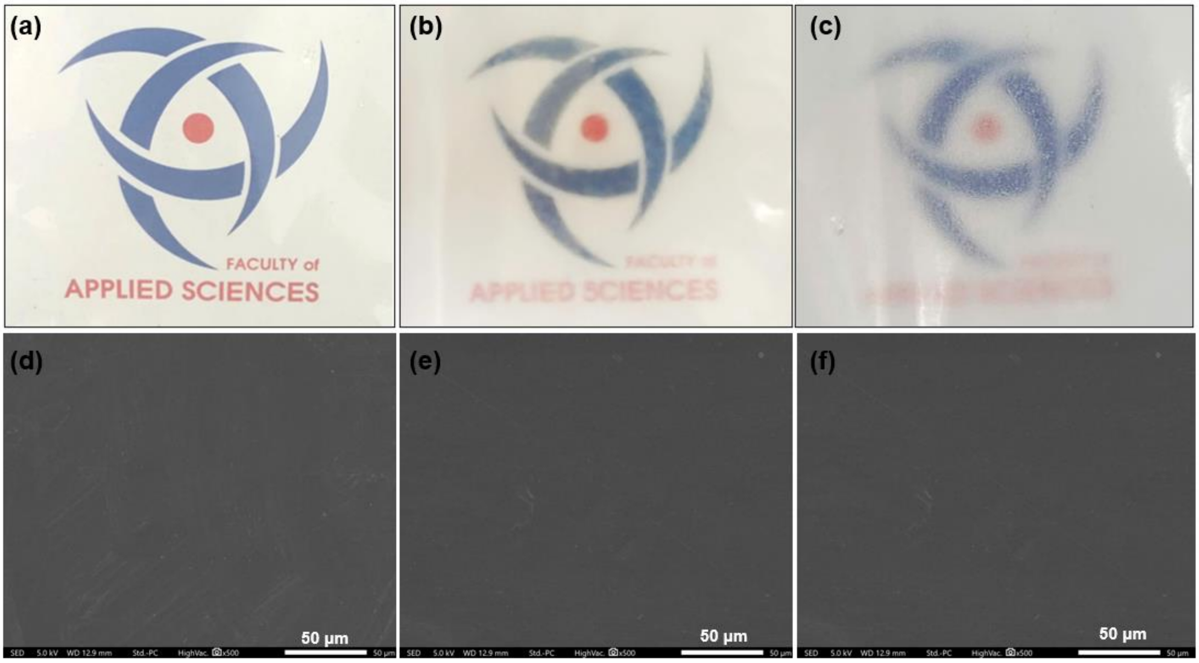

Figure 1. *Photographs (a-c) and SEM images (d-f) of PVA/CS@CMC composite films with different CMC contents: 0 wt.% CMC (a, d), 3 wt.% CMC (b, e), and 6 wt.% CMC (c, f).*

As shown in Figure 2a, the broad absorption band at 3250 cm^{-1} is characteristic of the O–H stretching of hydrogen bonds between PVA, CS, and CMCs molecules. The C=O and C–O stretching vibrations from the acetate groups are characterized by the 1710 cm^{-1} peak, indicating that the acetate groups are not completely hydrolyzed in the PVA structure [22]. The stretching of the C–H bonds from the alkyl groups was observed at 2920 cm^{-1}. The absorption band at 1645 cm^{-1} is related to the amide I region, associated with the C=O stretching of the chitosan. The peak at 1555 cm^{-1} corresponds to the amide II bond, linked to the N–H bending in chitosan [23], [24]. The 1645 cm^{-1} peak indicates C=O stretching or hydrogen bonding, which slightly decreases with CMCs addition, suggesting that CMCs replace some hydrogen bonding interactions. In the 1375–1327 cm^{-1} region, the peaks are related to C–H bending vibrations in PVA, CS, and CMC [25]. Finally, the increased intensity at 1082 cm^{-1} is attributed to the stretching vibration of the C–O–C ether bond in the cyclic pyranose structure of cellulose, confirming the presence and strong interaction of CMC in the PVA/CS matrix.

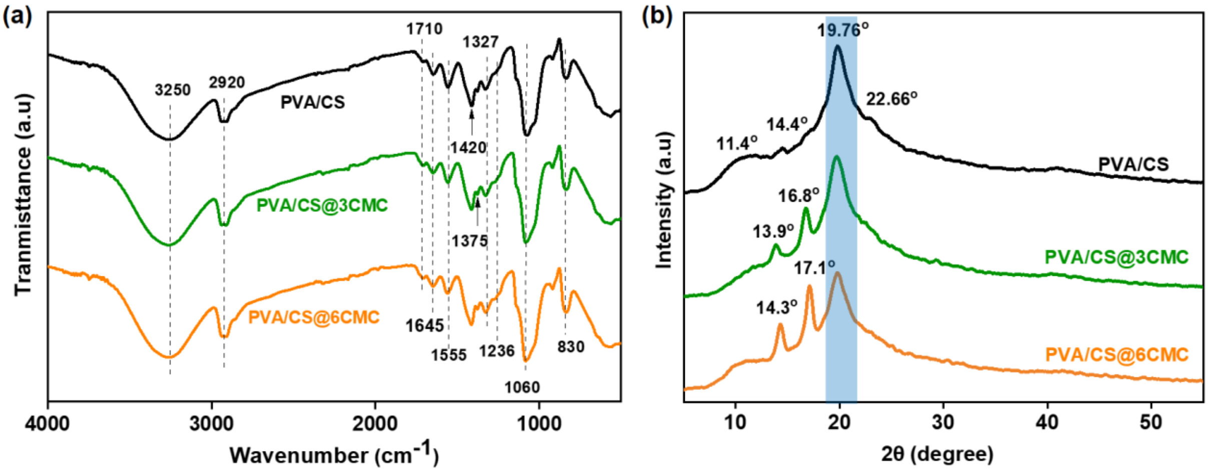

Figure 2. *FTIR spectra (a) and XRD pattern (b) of PVA/CS film and PVA/CS@CMC composite films.*

The XRD pattern of PVA/CS@CMC composite film is depicted in Figure 2b. The pure PVA/CS film exhibited the typical semicrystalline peak at (101) lattice of PVA visible at 2θ = 19.76° [26]. The

PVA/CS@3CMC and PVA/CS@6CMC composite films displayed the appearance of a secondary peak at approximately 2θ = 16–17° and 14°, which in turn corresponds to (001) and (110) lattices of pristine cellulose I material. When the CMC content increased to 3 wt.% and 6 wt.%, the peak intensity at 2θ of 19.76° decreased significantly and became broader, indicating a decrease in the crystallinity of the film. This is presumably due to the influence of CMC, which is present under the amorphous phase or small crystalline phase. Additionally, CMC has many hydroxyl groups (–OH) similar to those found in PVA. When mixed, the –OH groups of CMC form hydrogen bonds with PVA, disrupting the intramolecular bonds of PVA, thereby reducing the ability to arrange the PVA chains, leading to a decrease in crystallinity [27], [28]. Moreover, given that CMC is not dispersed uniformly, it may create microscopic regions that disrupt the PVA crystal structure [27], [28].

The UV–Vis spectra of the films are presented in Figure 3a. The UV-Vis spectra indicated that PVA/CS film, PVA/CS@3CMC and PVA/CS@6CMC composite films maintained high transparency in the visible range (380–700 nm). However, higher concentrations of CMCs slightly reduced the transmittance, particularly in the UV range (300–380 nm) [29], [30]. This decrease in transmittance may be attributed to the increased CMC content, which can lead to greater light scattering. The film containing 6 wt.% CMC exhibited the lowest transmittance, suggesting an enhanced ability to block UV light and potential effectiveness as a UV shielding material.

The swelling index of the PVA/CS@CMC films, shown in Figure 3b and Table 1, decreases progressively as the CMCs content increases. Specifically, the swelling index drops from 479.12% for the film without CMCs to 286.09 % with 3 wt.% CMCs and further reduces to 166.27 % with 6 wt.% CMCs. This decline can be attributed to enhanced hydrogen bonding interactions between the hydroxyl groups of CMCs and the functional groups of PVA and CS. These interactions create a denser and more compact polymer network structure, which restricts the material's ability to absorb and retain water [31].

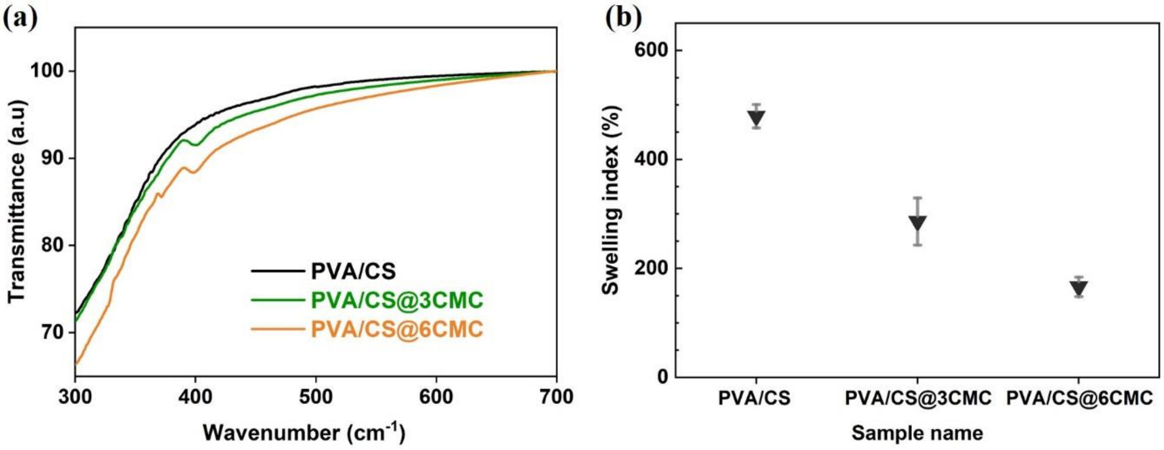

Figure 3. *UV-Vis spectra (a) and swelling index (b) of PVA/CS film and PVA/CS@CMC biocomposite films.*

Table 1. *Swelling and mechanical properties of PVA/CS film and PVA/CS@CMC biocomposite films.*

Sample	Swelling index	Tensile strength at break (MPa)	Elongation at break (%)	Young's modulus (MPa)
PVA/CS	479.12 ± 21.57	50.63 ± 5.45	121.4 ± 22.21	4.71 ± 0.15
PVA/CS@3CMC	286.09 ± 43.09	59.07 ± 2.03	106.6 ± 6.82	10.89 ± 1.56
PVA/CS@6CMC	166.27 ± 17.56	41.54 ± 3.63	80.57 ± 5.81	4.98 ± 2.46

The tensile strength and elongation at break of the PVA/CS@CMC films are shown in Table 1 and Figure 4a. The pure PVA/CS films, PVA/CS@3CMC and PVA/CS@6CMC composite films exhibited tensile strengths of 50.63 MPa, 59.07 MPa, and 41.54 MPa, respectively. The PVA/CS@3CMC sample significantly improved tensile strength due to hydrogen bond formation with the PVA/CS matrix, enhancing stress transfer and cohesion. However, increasing CMC to 6 wt.% resulted in a decrease in tensile strength, likely due to phase separation or agglomeration that compromised the film's mechanical integrity. The elongation at break decreased from 121.40 % at pure PVA/CS film to 106.60 % and further to 80.57 % at PVA/CS@3CMC and PVA/CS@6CMC biocomposite film, respectively. This reduction indicates that CMCs restrict polymer chain mobility, lowering the elongation of films. Previous studies have also shown that increasing filler content beyond an optimal level reduces the extensibility of biopolymer films [32], [33]. Adding 3 wt.% CMCs to the PVA/CS matrix improved the tensile strength but reduced the elongation at break, showing that the film became stiffer and that Young’s modulus increased (from 4.71 MPa to 10.89 MPa), as shown in Table 1 and Figure 4b. This improvement is mainly due to stronger hydrogen bonding and better stress transfer between the polymer and CMC. However, when the CMC content was increased to 6 wt.%, the tensile strength dropped, and the elongation continued to decrease, indicating that the film became more brittle [10]. At PVA/CS@6CMC, Young’s modulus decreases (4.98 MPa) due to poor dispersion of CMC and weaker bonding between the filler and the polymer matrix. Similar observations were also recorded in the tensile stress-strain curves of the material samples (Figure 4c). It can be seen that the PVA/CS@3CMC sample exhibited the highest tensile stress at fracture, followed by the PVA/CS and PVA/CS@6CMC films. In addition, the PVA/CS film showed the highest fracture strain, followed by the PVA/CS@3CMC and PVA/CS@6CMC samples. On the other hand, the PVA/CS@3CMC sample exhibited a maximum slope, followed by the PVA/CS@6CMC and PVA/CS films. These results are likely due to the increase in reinforcing filler content, which increases the interfacial surface bonds between CMCs and the matrix polymer chains (especially at 3% by weight of CMCs), increasing tensile stress at fracture and Young's modulus, as well as reducing fracture strain due to the limitation of relative motion between CMCs and the matrix polymer chains. When the CMC content is increased to 6%, the reinforcing material does not disperse evenly but aggregates, resulting in an insufficient polymer matrix that fails to cover and bond well with the CMC's surface. This results in loose bonding between the reinforcing filler and the matrix, thereby reducing the ability to transfer force from the polymer to the filler and ultimately reducing the tensile stress at break. At the same time, the high filler content occupies a significant portion of the film volume, restricting the slippage of the polymer chains and leading to reduced elongation at break.

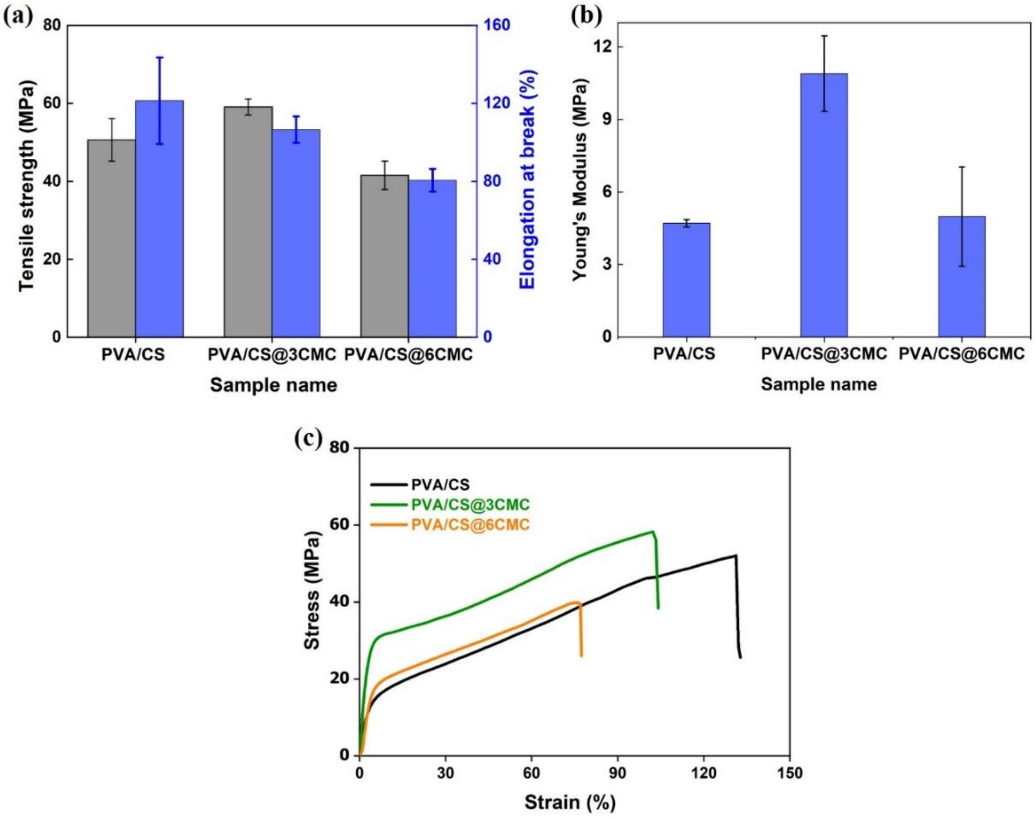

Figure 4. *Tensile strength and elongation at break (a), Young's modulus (b), and stress-strain curve (c) of PVA/CS and PVA/CS@CMC biocomposite films.*

4. Conclusions

The study successfully extracted crystalline microcellulose from low-cost waste notebooks and used it as a reinforcement in bio-composite films composed of polyvinyl alcohol and chitosan. The average particle size was 615.15 nm, and the crystallinity index was high at 35.1 %. The use of CMCs to reinforce PVA/CS@CMC films significantly improved the mechanical properties of the films with a maximum tensile strength of 59.07 MPa and an elongation at break of 106.60 % at 3 wt.% CMCs content. The films had excellent UV blocking properties, absorbing a high percentage of UV light, while maintaining high transparency. Furthermore, the swelling index decreased with increasing CMCs content, indicating a tighter polymer network that reduced water absorption. The improvements achieved by using CMC to reinforce PVA/CS films. The study proposes an alternative solution to enhance the reuse or recycling of waste notebooks, partially reducing worldwide environmental issues, producing a valuable reinforcing agent in eco-friendly biocomposite film, and potentially in food packaging.

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Conflict of Interest

The authors have no conflicts of interest to declare that are relevant to the content of this article.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Vu Viet Linh Nguyen has been a lecturer in the Department of Materials Technology, Faculty of Applied Sciences, at Ho Chi Minh City University of Technology and Engineering (formerly Ho Chi Minh City University of Technology and Education) since 2020. She earned her engineering, master's, and doctoral degrees from Ho Chi Minh City University of Technology – Vietnam National University in 2008, 2011, and 2020. She has published over 20 research papers on polymer materials, hydrogels, and drug delivery systems.
Email: linhnvv@hcmute.edu.vn. ORCID: https://orcid.org/0000-0002-3247-6754

Minh Khanh Hao Phung is a final-year undergraduate student at the Faculty of Applied Sciences, Ho Chi Minh City University of Technology and Engineering (formerly Ho Chi Minh City University of Technology and Education), Vietnam. He is researching the development of nanomaterials and and polymer composite films.
Email: 21130065@student.hcmute.edu.vn. ORCID: https://orcid.org/0009-0002-7671-0183

Thai Hoa Nguyen is an assistant teaching and researcher in the Laboratory of Materials Technology (MT lab) and Polymer Research Center, Ho Chi Minh City University of Technology – Vietnam National University. His background is Materials Science and Technology, with undergraduate and master's theses at the same University. Furthermore, he has published more than 18 research papers on polymer biomaterials, polymer blends, and rubber technology.
Email: nthhoa@hcmut.edu.vn. ORCID: https://orcid.org/0000-0002-5751-610X

Vinh Tien Nguyen received the specialist degree in chemistry from Tula State University, Tula, Russia, in 2009 and the Ph.D degree in chemistry also from Tula State University in 2014. From 12/2013 to 12/2014, he was a probationary lecturer at the Department of Chemical Technology, Faculty of Chemical and Food Technology, Ho Chi Minh City University of Technology and Education (currently Ho Chi Minh City University of Technology and Engineering), Vietnam. From 12/2014 till now, he has worked as a full-time lecturer in the same faculty. His research interests include nanomaterials and polymeric materials applied in food technology.
Email: tiennv@hcmute.edu.vn. ORCID: https://orcid.org/0000-0002-1863-4138

Van Quy Nguyen is currently a Lecturer at Ho Chi Minh City University of Technology and Engineering. He received a PhD degree from the School of Chemical Engineering, Sungkyunkwan University (SKKU), South Korea in 2022. He has published about 20 research articles. His research interests are in composites and biomaterials.
Email: quynv@hcmute.edu.vn. ORCID: https://orcid.org/0000-0002-5952-0700