SCIENCE LETTERS

ISSN: 1675-7785
eISSN: 2682-8626

Available online at
http://myjms.mohe.gov.my/index.php/sl

Science Letters 18(3) 2024, 34-42.

Cellulose Agro-Waste for Potential Bio-packaging

Noor Saadiah Mohd Ali[1], Nurhafizah Saidin[2]*, Rosmaya Jamaudin[1], Siti Fathimah Putery Jemain[1], Nur Farrah Syazwanie Ismail[3], Wan Ahmad Khairi Wan Ahmad[4]

[1]*Department of Chemistry, Centre for Foundation Studies, International Islamic University Malaysia, Gambang Campus, 26300, Pahang, Malaysia*
[2]*Department of Mathematics, Centre for Foundation Studies, International Islamic University Malaysia, Gambang Campus, 26300, Pahang, Malaysia*
[3]*Department of Islamic Revealed Knowledge, Centre for Foundation Studies, International Islamic University Malaysia, Gambang Campus, 26300, Pahang, Malaysia*
[4]*Department of Human Science, Centre for Foundation Studies, International Islamic University Malaysia, Gambang Campus, 26300, Pahang, Malaysia*

ARTICLE INFO

Article history:
Received 19 July 2024
Revised 1 August 2024
Accepted 28 August 2024
Online first
Published 10 October 2024

Keywords:
Biofilm,
Cellulose,
Antimicrobial,
Food Packaging

DOI:
10.24191/scl.v18i4.6785

*Corresponding author.
E-mail address:
nurhafizah@iium.edu.my

ABSTRACT

Millions of tons of food and materials get spoiled daily worldwide because of storage and transportation issues, pointing out inequalities in packaging systems. Packaging remained an important factor in ensuring the quality of packed food products. It had been refined from time to time to provide a standard. Parallel to the introduction of the National Green Technology Policy by the government of Malaysia on alternatives to solve energy and environmental issues through green technology, it is our interest to develop a new class of materials from agro-waste sources that can contribute towards the advancement of eco-friendly and sustainable materials for packaging applications with enhanced barrier, thermo-mechanical, rheological, and antimicrobial properties. The bio-packaging was designed using cellulose-based biodegradable films. The biofilm was evaluated based on the mechanical, structural, surface, and chemical properties to confirm the suitability of the biofilm as food packaging. Different fruits and vegetables were packed in a container in three different environments: 1) control, 2) covered with PE film, and 3) covered with biofilm. It was observed that the samples packed with the biofilm demonstrated better freshness and lower moisture content than that of the control and container with PE. Naturally, renewable materials used in this work can replace current synthetic plastic and can be commercialized in the environmentally friendly food packaging market.

INTRODUCTION

The intensification of agricultural activities throughout the year increases the volume of agricultural waste. According to Ugwouke et al. [1], agricultural wastes are residual materials from the production, processing, and marketing of crops and animals that can be found in the form of liquid and solid. The increased amounts of waste mentioned are becoming a concern as they might cause environmental issues. The vast number of residues from agricultural-based materials that fail to follow proper disposal procedures may cause environmental pollution that might harm humans and animals [2]. Thus, the researcher discovered that it is important to devise an initiative to manage those wastes.

Bio packaging can be one of the alternatives to plastic packaging, and it has been used widely worldwide. Shaikh, Yaqoob, and Aggarwal [3] mentioned that the risk caused by non-biodegradable plastic can be controlled by using biodegradable polymers as an alternative. Millions of tons of food and materials get spoiled due to storage and transportation issues caused by unequal packaging systems. Based on [4], optimizing packaging may be one of the solutions for 30% of food produced worldwide that is wasted along the food supply chain. The packaging industry has evolved using modern technology from the conventional methods of new-generation packaging, including glass, wood, paper, and other biodegradable materials, to extend the shelf life of food products. The right selection of packaging materials and technology can play a significant role in determining the shelf life of a certain food product [5]. All in all, it is necessary to develop an innovation to improve the current packaging that has been used widely.

The bio packaging is in line with one of the areas under Malaysia's National Green Technology Policy (NGTP) that focuses on waste management. NGPT is an initial step by the government to embrace Green Technology to accelerate the national economy as well as promote sustainable development through four pillars, namely energy, environment, economy, and social [6]. However, the government is still new to the policy compared to developed countries. According to [7], a lack of local expertise and initiative from the government leads to insufficient market demand for sustainable production and consumption of green products and services. This research can be one of the initiatives to uphold the NGTP in Malaysia by producing new types of packaging, as the research on it is relatively low. This is in line with research on food loss and waste due to the relatively low types of packaging [4]. Thus, researchers found it crucial to develop a new type of bio packaging with a new material that can advance eco-friendly and sustainable materials for packaging applications with enhanced barrier, thermo-mechanical, improved rheological, and antibacterial properties.

The world's population has increased rapidly and is estimated to reach 9 billion by 2050 [8]. This fast-growing pollution led to the significant production of livestock and crop production, producing abundant agro waste. Agro wastes are made from various sources, including grape pomace, tomato pomace, pineapple, orange, lemon peels, sugarcane bagasse, rice husks, wheat straw, and palm oil fibers. Fruit-processing waste has abundant bioactive compounds such as phenolics, alkaloids, flavonoids, carotenoids, glycosides, tannins, saponins, terpenoids, and vitamins [9]. These wastes generally have natural fibers, which are reusable as raw materials to obtain lignocellulose, which consists of cellulose, hemicellulose, and lignin. Globally, cellulose is considered a potent material for microbial transformation as it makes up the majority of total biomass, which is approximately 30-50 % [10]. The agriculture sector can be sustainable by converting agro-waste into valuable resources. Previous studies have revealed that cellulose agro-waste has been used for diverse applications, including paper manufacturing, the food industry, pharmaceuticals, and food packaging.

Over time, the packaging industry has evolved using modern technology, from conventional methods of new-generation packaging, including glass, wood, and paper, to most new biodegradable materials. Thus, we are interested in developing a new class of materials that can contribute towards advancing eco-friendly and sustainable materials for packaging applications with enhanced barrier, thermo-mechanical, improved rheological, and antibacterial properties. The present study aims to prepare biodegradable film based on carboxymethyl cellulose (CMC) and polyvinyl alcohol (PVA) using economic techniques. Secondly, this study aimed to evaluate the suitability and compatibility of the biodegradable film for food packaging application.

https://doi.org/10.24191/scl.v18i4.6785

EXPERIMENTAL

Sample Preparation

The overview of sample preparation for this study is summarized in Figure 1. Carboxymethyl cellulose (CMC) (MW: 90000; Acros Organic Co.) and partially hydrolyzed polyvinyl alcohol (PVA) (MW: 70000; Merck Schuchardt) were used to produce the polymer blend. The polymer blend was prepared via solution casting. A ratio of 80:20, CMC/PVA, was stirred in 98.0 mL of distilled water for 4 hours. After a homogenous solution was produced, the solution was cast into glass petri dishes and left in the oven. The solution was oven-dried for approximately 6 h at 60 °C and followed by additional drying in a desiccator to verify that all the solvent was evaporated [11].

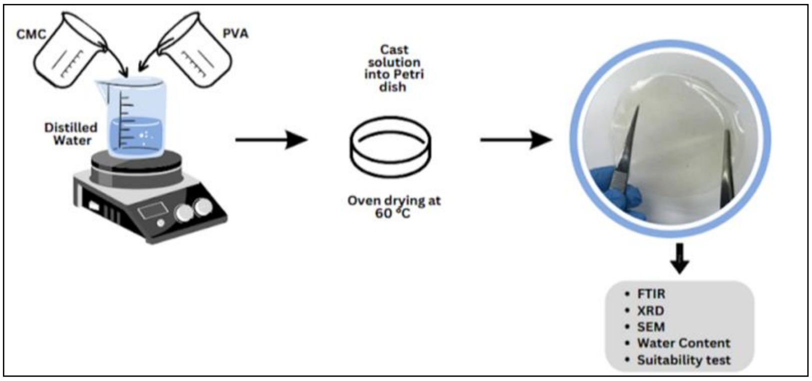

Figure 1. Sample preparation of the CMC/PVA and characterization.

Attenuated Total Reflection-Fourier Transform Infrared Spectroscopy (ATR-FTIR)

A Perkin Elmer FTIR spectrophotometer with an attenuated total reflection (ATR) in the frequency range of 700 cm^{-1} to 4000 cm^{-1} was used to investigate the complexation of CMC/PVA. All ATR-FTIR spectra were collected with a resolution of 2 cm^{-1} after 10 scans.

X-ray Diffraction (XRD)

XRD analysis is a valuable method for determining the degree of crystallinity and amorphousness. The XRD measurements were taken with an XRD-Rigaku MiniFlex II equipped with nickel-filtered Cu Kα (λ = 0.154 nm) radiation (30 kV, 15 mA) and scanned at angle 2θ between 5° and 80°.

Scanning Electron Microscopy (SEM)

The surface morphology of the CMC/PVA was examined using a Hitachi High Technologies Tabletop Miniscope TM3030Plus equipped with an energy-dispersive X-ray spectrometer (EDS). For SEM and EDX, accelerating voltages of 10 and 15 kV were utilized, respectively. The charge-up reduction mode was employed [12].

https://doi.org/10.24191/scl.v18i4.6785

Water Content

Samples of tomatoes are placed in three of the same type of plastic containers. The first sample was exposed to an open environment without cover, which acted as the control. The other two samples were covered using polyethylene (PE) and CMC/PVA biofilm. Analytical balance weighed each sample on days 1, 7, and 14. The moisture content of tomatoes is calculated based on the initial (day 1) and final weights (days 7 and 14) of the sample, which assumes that all weight loss is due to the removal of water and ignores the loss of other volatiles. Moisture content was calculated using the following formula:

$$\text{Moisture Content} = \frac{\text{IW} - \text{FW}}{\text{IW}} \times 100 \qquad \text{Equation 1}$$

Where IW is the initial weight (day 1) of fruit + container (in g), and FW is the final weight (days 7 and 14) of fruits + container (in g) [13].

Suitability Test

Different fruits and vegetables were packed in a container in three different environments; the first was the control, where we exposed the food to an open atmospheric environment. The second environment is where the food is covered with PE film, and the third is covered with CMC/PVA biofilm. All of them were left at room temperature.

RESULTS AND DISCUSSION

Evaluation of Surface Morphology, Chemical and Structural Properties

The resulting CMC/PVA biofilm was characterized via FTIR, XRD, and SEM analysis to evaluate chemical properties, structural properties, and surface morphology. Further testing was done to study the suitability of CMC/PVA biofilm as food packaging. Utilizing infrared (IR) spectroscopy is of paramount significance within this investigation, owing to its aptitude in delineating polymers' intricate structural configuration and conformational aspects. This is pivotal in unraveling the complexities inherent in the amalgamation of biopolymers within the blend system. Illustrated in Figure 2, the distinct spectrum corresponding to pure PVA showcased the predominant characteristics of the PVA polymer chain. This was discernible through the manifestation of vibrational carbon skeleton motions spanning the spectral range of 1089 to 1376 cm^{-1}.

In contrast, the IR spectrum of pure CMC unveiled the presence of five distinct absorption bands, each aligning with the signature peaks of CMC. Remarkably, within the IR spectrum of the CMC/PVA biofilm, two novel shoulder peaks emerged at 844 cm^{-1} and 1737 cm^{-1}. These correspond to the vibrational stretching of C-C and C=O within PVA, respectively.

This phenomenon robustly indicates the successful integration of CMC and PVA within the blend. The method of choice for crafting these blended systems is the solution-casting approach. This method confers enhanced mastery over the composition of CMC-PVA blends. The mechanical integrity, thermal resilience, and barrier characteristics of the resultant films can be judiciously optimized through meticulous adjustments in the concentration of CMC, PVA, and the solvent milieu. This, in turn, paves the way for creating biodegradable materials tailored to diverse packaging requisites, exemplifying the potential of these blends in fostering sustainable solutions for modern packaging demands [12].

https://doi.org/10.24191/scl.v18i4.6785

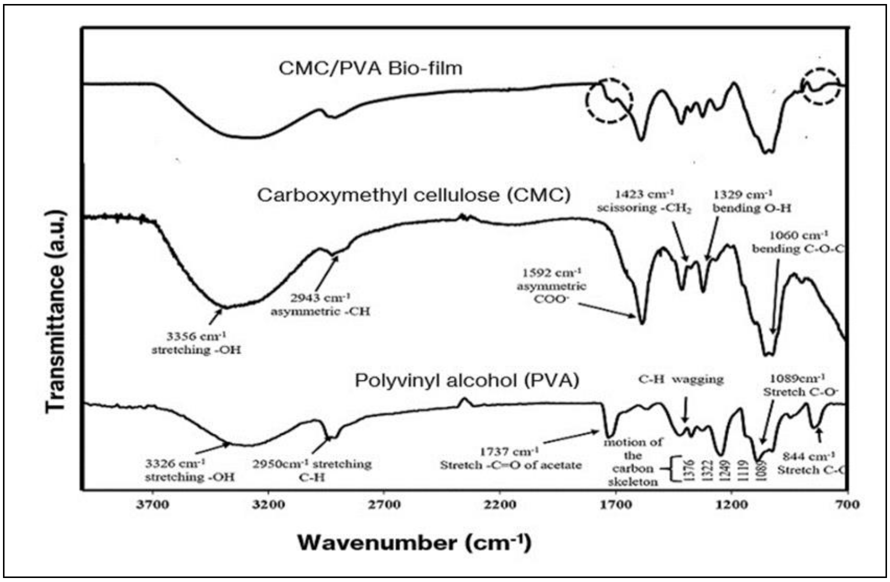

Figure 2. FTIR spectra of the CMC/PVA Biofilm compared with that of CMC and PVA.

The significance of combining two polymers has been documented [14]. In this research, samples of polymer nanocomposites were produced using the solution casting technique. This method employed a polymer mixture consisting of polyvinyl alcohol and carboxymethyl cellulose in a ratio of 70/30 wt.% as the organic host matrix. The matrix was then infused with varying concentrations of biosynthesized gold nanoparticles (Au NPs) obtained from the leaf extract of the green mint plant (Mentha Spicata L.) to serve as the inorganic nanofiller. Scientists explored various blending methods to attain optimal structural integrity of films within the CMC-PVA blends. A study conducted by Rajan and Joseph [15] scrutinized the effects of two blending approaches, solution casting and melt blending, on the formation of the films.

The findings of this study revealed that the solution-casting technique outperformed melt blending in terms of enhancing film integrity. This was attributed to its ability to uniformly disperse CMC and PVA within the matrix, resulting in fewer flaws and improved mechanical characteristics of the films. Conversely, Zhang et al. [16] opted for an in-situ polymerization method to examine how initiator concentration and reaction time influenced the molecular weight and thermal properties of CMC-PVA blends. The outcomes of their investigation demonstrated that the in-situ polymerization process yielded blends with tailored properties that were well-suited for specific applications in food packaging. A pivotal aspect in enhancing the flexibility of polymer blends for food packaging involves inducing an amorphous structure through molecular modification.

Meanwhile, Figure 3(a) above depicts the XRD spectra corresponding to pure CMC, pure PVA, and the composite CMC/PVA biofilm. The XRD methodology ascertains the atomic arrangement within materials, whether crystalline or amorphous. In the context of solid electrolytes, these encompass two categories: amorphous and crystalline. An optimal polymer electrolyte should prominently exhibit an elevated amorphous composition alongside diminished crystalline content. It is important to note that the spectra of unadulterated CMC and pure PVA display attributes of semi-crystallinity owing to the coexistence of both crystalline and amorphous domains. However, after the blending process, the peaks exhibit a broadened and flattened profile, indicative of an increased prevalence of amorphous characteristics. This property holds significant promise, particularly for applications in food packaging. Meanwhile, Figure 3(b) showcases a high-resolution SEM image (25 μm) of the CMC/PVA biofilm

https://doi.org/10.24191/scl.v18i4.6785

surface. The image distinctly portrays a consistent microstructure within the polymer blend system, devoid of any observable phase segregation, thus strongly implying its amorphous nature.

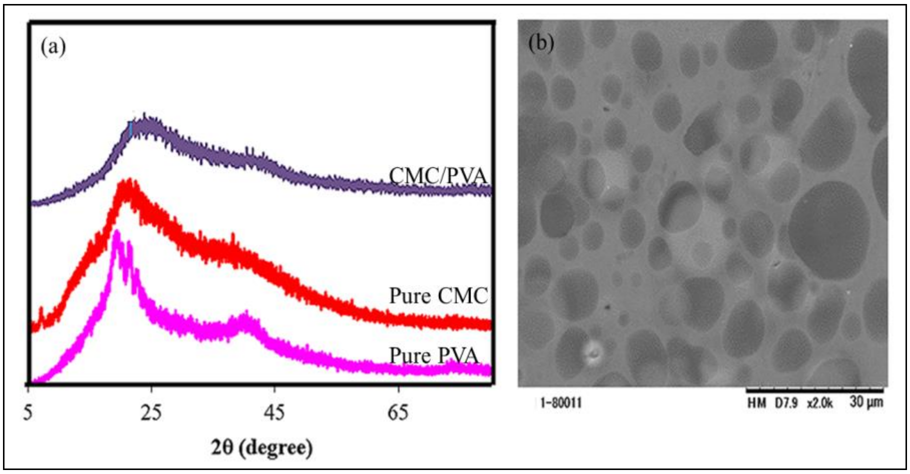

Figure 3. (a) XRD spectra of the CMC/PVA Bio-film compared with that of CMC and PVA; (b) SEM image of the CMC/PVA.

Water resistance is a critical requirement for food packaging to prevent moisture ingress and maintain the freshness of food products. The solution casting method can tailor the hydrophobic-hydrophilic balance in CMC-PVA blends, resulting in films with enhanced water resistance and reduced water uptake [17]. Figure 4 below demonstrates the water content analysis for tomatoes in a controlled environment, in polyethylene (PE) plastic, and in CMC/PVA biofilm, respectively.

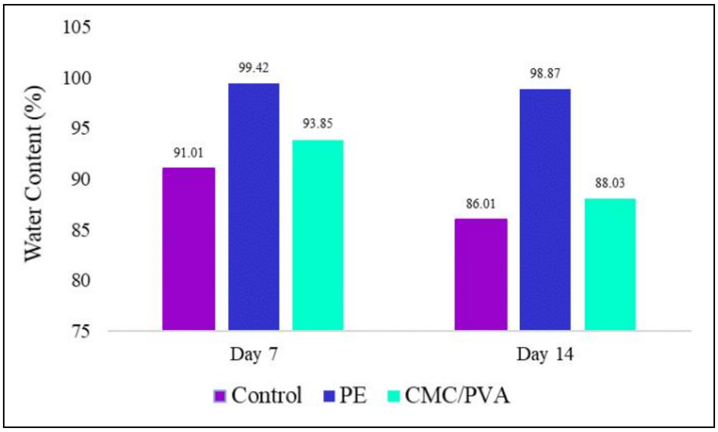

Figure 4. The water content analysis for tomatoes in a controlled environment in PE plastic and in CMC/PVA biofilm, respectively.

Based on Figure 4 above, the bar chart shows that the water content of tomatoes packed with PE is the highest and becomes unfavorable because moisture could affect food quality. The literature shows that CMC/PVA is a moisture absorber [18]. Consequently, the water content in containers packed with CMC/biofilm is relatively lower than PE. A study by [19] revealed that lower water content could reduce the risk of microbial contamination while preserving the sensorial characteristic of packed food.

https://doi.org/10.24191/scl.v18i4.6785

CMC /PVA act as moisture absorbers. Thus, CMC/PVA biofilm can reduce the risk of microbial contamination while preserving the sensorial characteristics of packed food. As evident in Figure 5 below, different fruits and vegetables were packed in a container in three different environments; the first one was the control, where the sample was exposed to an open atmospheric environment; second, the sample covered with PE film; and the third was covered with CMC/PVA biofilm. All of them were left at room temperature. It was observed that the samples packed with the biofilm demonstrated better freshness after 10 weeks.

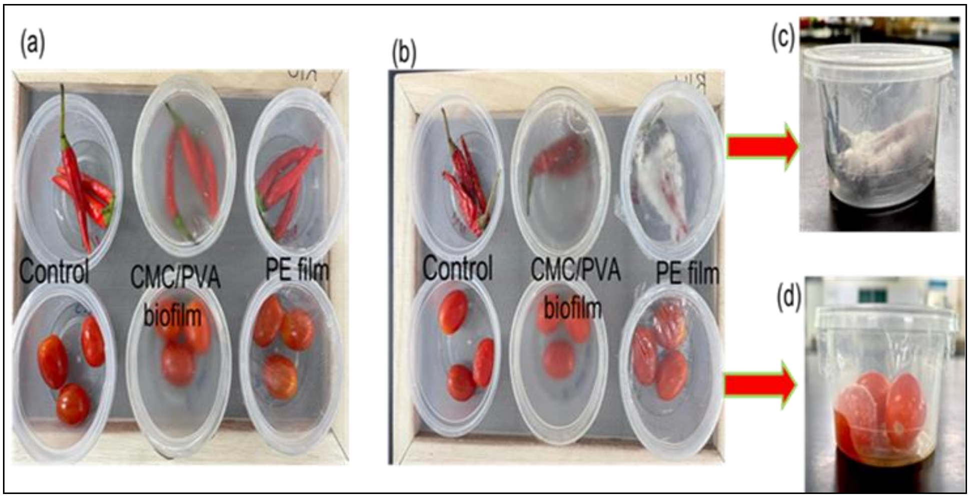

Figure 5. Suitability test of the CMC/PVA and PE film (a) Day 1; (b) After 10 weeks; (c) and (d) food preserved using PE synthetic film

Based on the results observed in Figure 5, these results might be due to the characteristics of the CMC/PVA biofilm that have excellent air permeability and breathability. It was confirmed by the water content analysis previously that food packed with CMC/PVA has lower water content than those packed with PE plastic. On top of that, the presence of water vapor and mold formation in the container wrap with PE supports our claim. This discovery is reminiscent of research conducted by reference [20], in which a biofilm composed of CMC and sugar cane bagasse was found to provide effective air permeability and breathability, thereby contributing to the preservation of food freshness [21]. In a separate investigation, the amalgamation of chitosan ascorbate with CMC was identified to enhance its mechanical robustness, a crucial factor in materials utilized for food packaging purposes [17]. Based on the findings, CMC/PVA biofilms demonstrated good physicochemical and antioxidant properties, showing their potential for food packaging applications. In addition, CMC and PVA are biodegradable materials that comply with the Food Safety Act 1990 and GRAS (generally known as safe materials) and have the potential to be used as safe and sustainable food packaging.

CONCLUSION

This study focused on biofilm preparation using CMC and PVA for food packaging applications. CMC and PVA are regarded as potent materials in food packaging applications due to their biocompatibility, biodegradability, and sustainability. The FTIR, XRD, and SEM analysis indicates the success of blending the two polymers of CMC and PVA with enhanced properties suitable for food packaging applications. Moreover, the suitability test reveals that the samples packed with the biofilm demonstrate better freshness and lower moisture content, as compared to that of the control and container

https://doi.org/10.24191/scl.v18i4.6785

with a synthetic polymer, PE. Thus, CMC and PVA used in this work can replace current synthetic plastic and can be commercialized in the environmentally friendly food packaging market.

ACKNOWLEDGEMENTS

The acknowledgement is extended to the International Islamic University Malaysia (IIUM) for their generous provision of research grant support under reference (SOTL23-004-0004). Furthermore, appreciation is also directed towards the Centre for Foundation Studies (CFS IIUM) for their invaluable technical assistance throughout this endeavor.

CONFLICT OF INTEREST STATEMENT

The authors agree that this research was conducted without any self-benefits or commercial or financial conflicts and declare the absence of conflicting interests with the funders.

AUTHORS' CONTRIBUTIONS

Nurhafizah supervised the progress of editing of the draft. Noor Saadiah developed the method and analysis and also anchored the reviewing progress. Rosmaya, Siti Fatimah Putery, Nur Farrah Syazwanie, and Wan Ahmad Khairi assisted with the sample preparation and analysis. All authors were involved in writing the manuscript and editing the subsequent drafts.

REFERENCES

[1] Ugwuoke, C. U., Monwuba, N., Onu, F. M., Shimave, A. G., Okonkwo, E. N., & Opurum, C. C. (2018). Impact of agricultural waste on sustainable environment and health of rural women. *Civil and environmental research*, *10*(9), 1-9.

[2] Sadh, P. K., Duhan, S., & Duhan, J. S. (2018). Agro-industrial wastes and their utilization using solid-state fermentation: a review. *Bioresources and bioprocessing*, *5*(1), 1-15.

[3] Shaikh, S., Yaqoob, M., & Aggarwal, P. (2021). An overview of biodegradable packaging in the food industry. *Current Research in Food Science*, *4*, 503-520.

[4] Wohner, B., Pauer, E., Heinrich, V., & Tacker, M. (2019). Packaging-related food losses and waste: an overview of drivers and issues. *Sustainability*, *11*(1), 264.

[5] Marsh, K., & Bugusu, B. (2007). Food packaging—roles, materials, and environmental issues. *Journal of Food Science*, *72*(3), 39-55.

[6] Ministry of Energy, Green Technology and Water (KeTTHA). (2017). *Green technology master plan Malaysia 2017–2030*. Prime Minister's Office of Malaysia. https://www.pmo.gov.my/wp-content/uploads/2019/07/Green-Technology-Master-Plan-Malaysia-2017-2030.pdf

[7] Borhanordin, A. H., Hussin, H., Samah, I. H. A., Rashid, I. M. A., Shafiai, S., Roni, M., & Zainudin, A. D. (2023, June). Green technology policy towards sustainable consumption and production in Malaysia. In *AIP Conference Proceedings* (Vol. 2571, No. 1). AIP Publishing.

[8] Koop, S. H., & van Leeuwen, C. J. (2017). The challenges of water, waste and climate change in cities. *Environment, development and sustainability*, *19*(2), 385-418.

[9] Ayala-Zavala, J. F., Rosas-Domínguez, C., Vega-Vega, V., & González-Aguilar, G. A. (2010). Antioxidant enrichment and antimicrobial protection of fresh-cut fruits using their own byproducts: Looking for integral exploitation. *Journal of Food Science*, *75*(8), 175-181.

[10] Koul, B., Yakoob, M., & Shah, M. P. (2022). Agricultural waste management strategies for environmental sustainability. *Environmental Research*, *206*, 112285.

https://doi.org/10.24191/scl.v18i4.6785

[11] Saadiah, M. A., Nagao, Y., & Samsudin, A. S. (2020). Proton (H^+) transport properties of CMC–PVA blended polymer solid electrolyte doped with NH_4NO_3. *International Journal of Hydrogen Energy*, *45*(29), 14880-14896.

[12] Saadiah, M. A., Zhang, D., Nagao, Y., Muzakir, S. K., & Samsudin, A. S. (2019). Reducing crystallinity on thin film-based CMC/PVA hybrid polymer for application as a host in polymer electrolytes. *Journal of Non-Crystalline Solids*, *511*, 201-211.

[13] Sinha, S. R., Singha, A., Faruquee, M., Jiku, M. A. S., Rahaman, M. A., Alam, M. A., & Kader, M. A. (2019). Post-harvest assessment of fruit quality and shelf life of two elite tomato varieties cultivated in Bangladesh. *Bulletin of the National Research Centre*, *43*, 1-12.

[14] Morsi, M. A., Oraby, A. H., Elshahawy, A. G., & Abd El-Hady, R. M. (2019). Preparation, structural analysis, morphological investigation and electrical properties of gold nanoparticles filled polyvinyl alcohol/carboxymethyl cellulose blend. *Journal of materials research and technology*, *8*(6), 5996-6010.

[15] Rajan, R., & Joseph, K.T. (2018). Biodegradable Polyvinyl Alcohol (PVA)-Carboxymethyl Cellulose (CMC) Composite Film with Balanced Mechanical and Barrier Properties. *Journal of Polymers and the Environment, 26(1)*, 135-146.

[16] Zhang, L., Wang, Q., Li, T. & Zhang, G. (2019). In-situ polymerization of CMC-PVA blends: Effect of initiator concentration and reaction time on molecular structure and thermal properties. *Journal of Polymer Science Part A: Polymer Chemistry*, *57(10),* 1037-1045.

[17] Lee, Y., Park, J., Lim, G. & Kim, K. (2020). Effects of blending temperature and shear rate on the mechanical properties of CMC-PVA blends prepared by melt blending. *Journal of Applied Polymer Science, 137(5),* 1-14

[18] Arefian, M., Hojjati, M., Tajzad, I., Mokhtarzade, A., Mazhar, M., & Jamavari, A. (2020). A review of Polyvinyl alcohol/Carboxymethyl cellulose (PVA/CMC) composites for various applications. *Journal of Composites and Compounds*, *2*(3), 69-76.

[19] Batista, R. A., Espitia, P. J. P., Quintans, J. D. S. S., Freitas, M. M., Cerqueira, M. Â., Teixeira, J. A., & Cardoso, J. C. (2019). Hydrogel as an alternative structure for food packaging systems. *Carbohydrate polymers*, *205*, 106-116.

[20] Yaradoddi, J. S., Banapurmath, N. R., Ganachari, S. V., Soudagar, M. E. M., Mubarak, N. M., Hallad, S., Hugar, S. & Fayaz, H. (2020). Biodegradable carboxymethyl cellulose-based material for sustainable packaging application. *Scientific reports*, *10*(1), 1-13.

[21] Bi, J., Tian, C., Zhang, G. L., Hao, H., & Hou, H. M. (2021). Novel procyanidins-loaded chitosan-graft-polyvinyl alcohol film with sustained antibacterial activity for food packaging. *Food Chemistry*, *365*, 1-9.

https://doi.org/10.24191/scl.v18i4.6785