

From orange peel to sustainable biocomposite: production and characterization of pectin/CMC films with matcha tea extract

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This study aims to evaluate the production of polysaccharide-based films obtained from orange peel waste, combining pectin (P) and a synthesized carboxymethyl cellulose (C) matrix with matcha tea extract (M). P was recovered from orange peel waste using an acid extraction technique. The extraction yield has been established at 23.37% and 67.38% esterification (DE). Biopolymer films named C/P and C/P/M were produced using the solvent casting method. In the films, C and P solutions were employed at different concentrations (80/20 w/w C:P) to achieve a total biopolymer content of 2% by weight. M was included into the film at concentrations of 0.2%, 0.4%, and 0.6% (w/w relative to total biopolymer weight). This work proposes to develop biocomposite films from C-P and investigate the effect of M on their structural and functional characteristics. Evaluations of opacity, moisture content, thickness, and water vapour transfer rate (WVTR) were performed to describe the films. M in the films, opacity and film thickness increased, but moisture content and WVTR values diminished. The reduction in WVTR, along with diminished moisture content, indicates an improved water vapour barrier resulting from the establishment of a dense network of new hydrogen bonds between M and the C-P matrix. Moreover, the increased film thickness and compact internal structure significantly impeded the transfer of water molecules within the matrix, enhanced its barrier characteristics. These biocomposite films, distinguished by improved water vapour barrier characteristics and heightened opacity, have significant potential as environmentally acceptable, sustainable for synthetic plastics in the food packaging sector.

Keywords: Orange peel, Polysaccharide-based, Pectin, Matcha tea

INTRODUCTION

The food packaging sector is among the largest consumers of plastic. It constitutes over 40% of overall plastic use, with a significant fraction of this packaging becoming waste in a relatively short duration [1]. Food packaging is essential for maintaining product quality, facilitating traceability, and protecting consumer safety. Nevertheless, the majority of conventional packaging materials are derived from petroleum and are non-biodegradable, so presenting an environmental hazard. In formulating sustainable solutions for the food packaging industry, it is essential to account for both the packaging materials and the waste produced by the food sector [2, 3]. In this context, by contributing to the circular economy in line with sustainability principles, high-value-added packaging materials (biopolymers, bioactive materials) can be produced [4].

The utilization of pectin extracted from fruit and vegetable peel waste in the fabrication of polysaccharide-based films is regarded as possessing considerable potential for the advancement of sustainable packaging materials.

Diverse sources of pectin may be utilized, including by-products from juice production, as well as peels and seeds from oranges, mangoes, bananas, limes, and pomegranates [5-7]. The fruit juice sector utilizes approximately 40% of global citrus production. The operation generates substantial quantities of citrus (orange, lemons/limes, mandarines) byproducts, including peels, pulp, and seeds. Peels constitute roughly 20% of an orange's (*Citrus sinensis*) total weight and, if inadequately processed, could result to water contamination and environmental damage. This waste represents an exceptional source for pectin extraction due to its diverse biological properties and significant economic worth. [8, 9]. Acidic extraction using citric acid, nitric acid, or hydrochloric acid is acknowledged as one of the most cost-effective and efficient techniques for separating high-yield and high-quality pectin. Among these methods, acidic extraction utilizing such as citric acid is distinguished as an environmentally sustainable and economically viable approach for obtaining high-quality pectin with a sustainable production [10].

Pectin's linear chain structure provides it with significant film-forming potential, excellent

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biodegradability, and biocompatibility. The main approach employed in the creation of pectin-based films is the casting process. Biocomposite films can be made by assuring optimal compatibility with biopolymers, including proteins, lipids, and other polysaccharides. Films produced from pectin demonstrate low gas permeability barrier characteristics which are significant, especially in food packaging applications. To mitigate the limitations of pure pectin films, including their elevated water vapor permeability and fragile mechanical properties, novel films are being formulated in the literature through the integration of diverse biopolymers (starch, cellulose, alginate, chitosan) and active compounds (organic acids, phenolic compounds) [7, 10, 11].

Furthermore, a robust film matrix can be successfully formed by utilizing various biopolymers such as carboxymethyl cellulose (CMC), offering significant potential to enhance the mechanical and barrier performance of cost-effective, non-toxic, renewable, and biodegradable packaging materials [12, 13]. Pure CMC, an anionic cellulose derivative, often demonstrates limited mechanical strength; however, this property is enhanced when integrated with other biopolymers, such as pectin [14]. Matcha (*Camellia sinensis*) is a natural green tea produced by carefully grinding tea leaves, historically cultivated in Japan. It is highly abundant in natural antioxidants, particularly polyphenols, which constitute almost 30% of its dry mass [15, 16]. The elevated biological activity of matcha, along with its soluble powder form, has significant promise for stabilizing functional food ingredients and incorporating them into biodegradable packaging methods [16, 17].

This work developed films utilizing a polysaccharide-based matrix, utilizing the film-forming, non-toxic and biocompatible characteristics of pectin and carboxymethyl cellulose. Pectin was extracted from orange peel waste, and created by including matcha tea extract into the film matrix. The films were submitted to characterization tests, including thickness, moisture, water vapor transfer rate (WVTR) and opacity, to assess their suitability for food packaging. The incorporation of matcha tea extract contributes to the existing literature by providing further insight into its effects on the physicochemical, optical, and barrier properties of biodegradable films. Furthermore, the use of pectin extracted from orange peel, a juice-industry byproduct, supports the valorization of food waste.

EXPERIMENTAL

- **Pectin extraction.** Pectin was extracted from orange peel with 1 N citric acid (1:25 w/v) at 80°C and pH 1.2 for 60 min. Following extraction, the mixture underwent filtering using Whatman No. 1 filter paper and was permitted to cool to room temperature. Ethanol was added to the cooled solution at a 1:2 (v/v) ratio and allowed to precipitate at +4 °C for 24 h. The precipitate was acquired using centrifugation at 1000 g for 15 min, washed with distilled water, and then dried in an air oven at 40 °C for 24 h to produce pectin. The pectin yield was calculated by the mass difference between the dried pectin and the initial dry raw material.

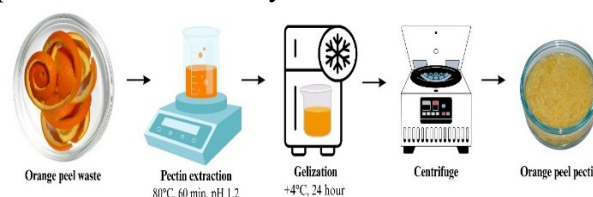


Figure 1. Schematic representation of the carrot pectin extraction

The extent of esterification of orange peel pectin was assessed utilizing a modified approach based on [18]. The dehydrated pectin was solubilized in 20 mL of distilled water and ethanol at a temperature of 40°C and titrated with 0.1 M NaOH. Upon the color shift, the volume of NaOH was noted as V_1 . Subsequently, 10 mL of 0.1 M HCl was introduced, and the solution was vortexed until the pink hue vanished. The titration with NaOH was conducted once more, and the volume was noted as V_2 . The DE was computed by Equation 1.

$$DE (\%) = \frac{V_2}{V_1 + V_2} \times 100 \quad (1)$$

- **Matcha tea extraction.** Matcha extract (ME) was produced by dissolving 3 g of matcha in 150 mL of boiling distilled water, subjecting the mixture to a water bath for 45 min, filtering it, and subsequently evaporating it with a rotary evaporator [19].

- **Production of films.** Film-forming solutions were prepared with a total polymer concentration of 2% (w/v) by adjusting the weight ratio of pectin (P) to carboxymethyl cellulose (C) as detailed below: Formulations of 80:20 (C:P, w/w) were prepared, incorporating matcha tea extract (M) abundant in polyphenols, at concentrations of 0.2%, 0.4%, and 0.6% (w/w based on total biopolymer weight. A and P were utilized in 80:20 (C:P w/w) ratios.

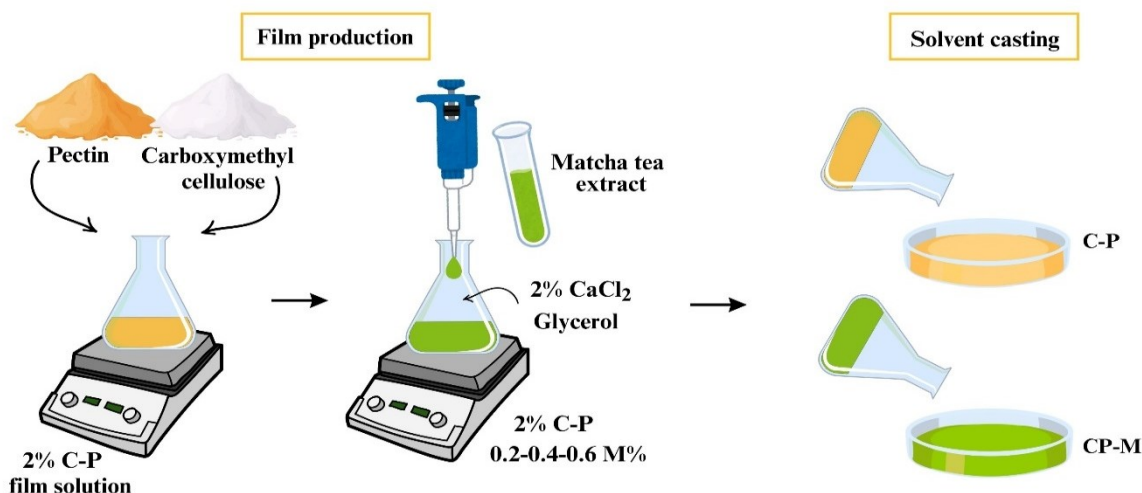


Figure 2. Schematic representation of the orange P, C and M composite films production

The film solution was stirred at 50 °C for 1 h and cross-linked with 2% CaCl₂ [20]. The films were produced using the cast film method. Glycerol is preferred as a plasticiser to impart flexibility and films drying oven at 40 °C. The films produced were assigned as C-P (C8-P2), CP-M2, CP-M4 and CP-M6 accordingly.

- *Film characterization.* The film thickness was measured with the Dasqua 4310 digital micrometre gauge. The thickness of each film has been evaluated at three locations: one at the centre and two next to it. The films' opacity, or light transmittance, was assessed utilizing a UV-Vis spectrophotometer (UV1280, Shimadzu, Japan). The opacity in absorbance at 600 nm was calculated utilizing Equation 1.

$$\text{Opacity} = \frac{Abs_{600}}{x} \quad (2)$$

where: Abs_{600} : absorbance of the film at 600 nm, x : thickness of the film (mm). The colors of the films were determined using a portable colorimeter (Linshang LS173). Before measuring the color parameters of the prepared films, the device was calibrated using a white standard plate.

The moisture content was determined by drying the films in an oven at 105 °C for 24 h until a constant weight was achieved. The moisture content was determined by dividing the difference between the starting weight (W_i) and the weight lost during drying (W_s) by the starting weight (W_i) (Equation 2).

$$\text{Moisture content (\%)} = \frac{W_i - W_s}{W_s} \times 100 \quad (3)$$

The water vapor transfer rate (WVTR) was assessed utilizing the usual gravimetric approach. Film samples were positioned in a Petri dish holding a prescribed volume of water. The films were trimmed to a precise dimension and affixed to the inner surface of the Petri dish lid with a waterproof

hydrophobic glue. The cover was affixed to the Petri dish, which was maintained at 25 °C in a controlled-humidity laboratory; the weights of the films were recorded after 24 h. Equation 4 was used to determine the WVTR.

$$\text{WVTR (\%)} = \frac{\Delta m}{A \times \Delta t} \quad (4)$$

where: Δm (g): weight of the container or film being tested, A (m²): surface area of the film, Δt (s): test duration.

Materials

Carboxymethyl cellulose from Sigma Aldrich, citric acid, ethanol, calcium chloride (CaCl₂) were of analytical grade and were purchased from Merck Chemicals, Türkiye. Fresh oranges (*Citrus sinensis*) and matcha tea powder (*Camellia sinensis*) were purchased at a local market in Çanakkale.

RESULTS AND DISCUSSION

Many factors affect pectin yield. The factors encompass the supply of pectin, pH, temperature, extraction duration, and the type of acid utilized. Literature indicates that pectin yield increases inversely with decreasing pH. This could be attributed to the limited solubility and degradation of pectin at elevated pH levels. The extent of pectin esterification is also affected by pH. Pectin is categorized into low-methoxyl pectin (DE < 50%) and high-methoxyl pectin (DE > 50%) [21, 22]. At the end of the study, the degree of esterification of pectin was calculated to be 67.38%, and the gelling ability of pectin was high (Table 1). Generally, an increase in pectin extraction yield was observed as extraction temperatures rose. At low temperatures, pectin yield decreases because acids cannot hydrolyze the insoluble form of pectin. Additionally, longer extraction times at constant temperature and pH increase pectin yield [9]. Yields and degrees of esterification of pectin derived from orange peels

under different conditions with those reported in the literature are summarized in Table 1.

Table 1. Literature review of pectin extraction parameters and yields from orange peels.

Type	Extraction method	Yield	DE (%)	Ref.
Orange peels	Citric acid 80°C-90 min- pH 2.0	18.27	61.65	[9]
Orange peels	Hydrochloric acid 95°C-60 min- pH 1.5	21.53	74.02	[21]
Orange peels	Citric acid 70°C-60 min- pH 3.2	29.41	-	[22]
Orange peels	Citric acid 80°C-60 min- pH 1.2	23.37	67.38	This study

Compared to the reference film, the thickness values of the CP-M films are gradually increasing (Table 2). The observed increase in film thickness corresponding to the rise in matcha tea extract (M) concentration can be attributed to the increased total

solids integrated into the matrix. The polyphenolic components in M enter the polymer matrix (C-P), modifying the dense arrangement of the chains and resulting in a more expansive internal structure through intermolecular interactions, so directly increasing the film thickness [23].

The addition of matcha to the C-P film resulted in enhanced opacity values, signifying low light transmission, a conclusion that aligns with existing research [16]. The reduction in optical performance is attributable to the colour of the matcha itself. As the amount of matcha added to the films increases, they take on a greener color, and their opacity increases in direct proportion to the amount of matcha (Fig. 3). The decrease in light transmittance can be considered as beneficial for packing materials. By shielding shelf-packaged food from sunlight, it captures consumer attention, enhancing the product's visibility on the shelf [17].

Table 2. Film thickness values

Film code	Thickness (mm)
CP	0.029
CP-M2	0.027
CP-M4	0.031
CP-M6	0.049

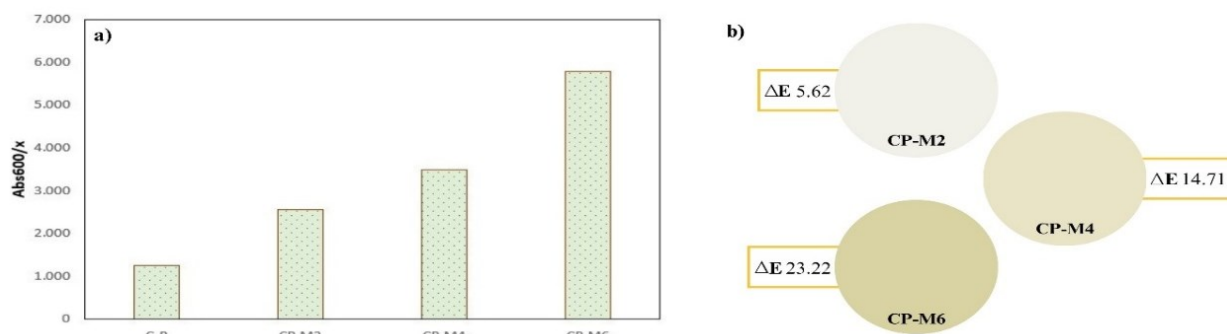


Figure 3. Films' opacity values (a) and color values (b)

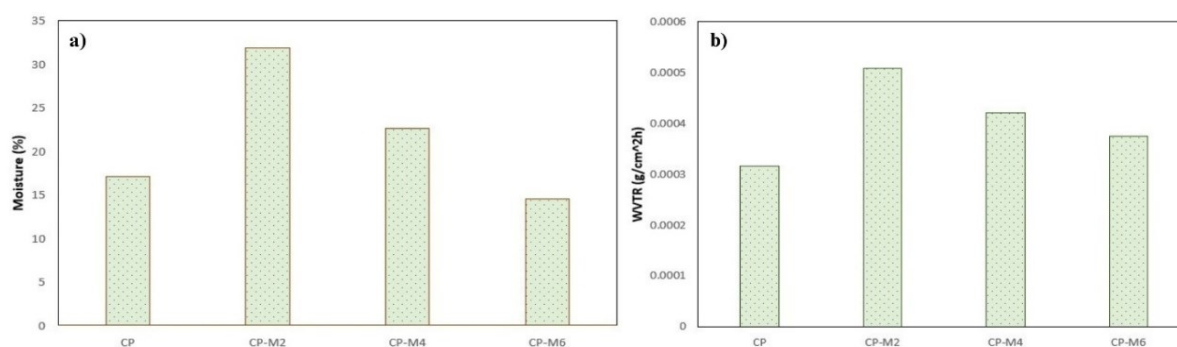


Figure 4. Moisture content (a) and vapor transmission rate (b) of the C-P and CP-M film series

The water vapor transmission rate (WVTR) diminished in accordance with the films' moisture content (Fig. 4b). Matcha is rich in polyphenolic compounds and has enhanced the water barrier qualities of films [24]. This results from the extensive network of hydrogen bonds established between the matcha polyphenols and the C-P matrix, along with the augmented film thickness. The polyphenol-rich M phase, at elevated concentrations, made the matrix more compact, hence inhibiting the transfer of water molecules and enhancing the films' water barrier capabilities. A lower permeability value of a packaging material indicates high resistance to water vapour intrusion [25].

CONCLUSION

This work involves the extraction of pectin from orange peel waste using hot acid extraction with citric acid at 80°C for 60 minutes at a pH of 1.2. The yield of the produced pectin (P) was 23.37%, and the degree of esterification (DE) was 67.38%, categorizing it as high-methoxyl pectin. Biocomposite packaging films were established by integrating pectin into a carboxymethyl cellulose-based film matrix and enhancing it with Matcha tea extract. The incorporation of M has modified the structural and functional characteristics of the C-P film. The M addition resulted in heightened opacity and thickness of the films, concurrently decreasing moisture content and water vapour transmission rate (WVTR). The films' efficacy as a water vapour barrier has been improved.

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