engineering proceedings

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Proceeding Paper

Beeswax and Castor Oil to Improve the Moisture Barrier and Tensile Properties of Pectin-Based Edible Films for Food Packaging Applications †

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† Presented at the 2nd International Electronic Conference on Processes: Process Engineering—Current State and Future Trends (ECP 2023), 17–31 May 2023; Available online: https://ecp2023.sciforum.net/.

Abstract: Biopolymer-based edible films and coatings are vital in making the global food-packaging industry more sustainable. These films/coatings protect and extend the shelf life of food by acting as barriers to moisture, oxygen, microorganisms, and ultraviolet light. Polysaccharides, due to abundant availability from natural plant-based resources and the tendency to form a gel in water, are excellent low-cost choices for packaging films. Additives such as hydrophobic agents, plasticizers, binders, and antimicrobial agents will improve the properties of films and coatings. The present work aims to develop pectin-based packaging film (from 5% *w*/*v* film-forming solution) by adding castor oil as a hydrophobic agent, beeswax as a plasticizer, and clove oil as an antimicrobial agent. Films were developed by using 2^3 (two-level, three-factor) statistical factorial design experiments. The amount of castor oil (5% and 15%), beeswax (5% and 10%), and clove oil (2% and 4%) are taken as the three factors. The developed films were analyzed for physical, moisture barrier, morphological, thermal, and tensile properties, and resistance to microbial growth. The results indicated that clove oil is a good antimicrobial agent. Furthermore, beeswax had a great impact by enhancing antimicrobial activity, elongation, and moisture barrier properties. Castor oil integration remarkably lowered the moisture and oxygen transmission rates relative to pure pectin films and some other additives reported in the literature. The optimized biofilms had a thickness of ~0.10 ± 0.004 mm, pH = 3, and transparency of $\Delta E = 9.15$ to 25. The elongation at break increased at least four times. The films were thermally stable at 400 °C. The detailed statistical analysis and analysis of various studies indicate that the amount of castor oil ($p < 0.05$), a combined effect of castor oil and beeswax ($p < 0.05$) is significant on barrier properties while the effect of beeswax ($p < 0.05$) is also significant on mechanical properties.

Keywords: polysaccharide; pectin; bio-edible films; beeswax; water vapor transmission rate (WVTR); mechanical test

Citation: Dharhshini, A.; Sistla, Y.S. Beeswax and Castor Oil to Improve the Moisture Barrier and Tensile Properties of Pectin-Based Edible Films for Food Packaging Applications. *Eng. Proc.* **2023**, *37*, 33. https://doi.org/10.3390/ECP2023-14670

Academic Editor: Chi-Fai Chau

Published: 17 May 2023

1. Introduction

Biodegradable edible films are gaining significant attention worldwide due to the increasing awareness of environmental challenges associated with synthetic polymeric films [1]. This is because of the fact that some biopolymers are safe to consume, and most of the biopolymers are environmentally friendly in nature and biodegradable. Biopolymer films are renewable, and with edible functional additives, they degrade much more readily than synthetic films. The biopolymers can be used as active green packaging materials to increase the shelf life of food, such as fresh and processed produce [2]. Biopolymer packaging keeps the freshness of food due to green ingredients, and also improves consumer health and safety. The bio-edible films and coatings are usually made from natural polymers like polysaccharides, proteins, and lipids, along with other additives [2,3]. A good film should have good moisture barrier properties, mechanical strength, flexibility, thermal properties, gas barrier properties, etc. The great diversity in the polymeric structures of polysaccharides, proteins, and lipids influences the linkage of polymers, degree of polymerization, etc.; these cause film properties to vary greatly. As a result, the films are tailored by altering the compositions and film-forming conditions like temperature, relative humidity, pH, etc. Polysaccharides that have a gel-forming ability with water include cellulose, carrageenan, chitosan, starch, etc. [4–6]. In general, polysaccharides are hydrophilic and have poor gas barrier properties. However, upon adding naturally occurring additives, their properties are enhanced. The scope of this research is narrowed down to pectin-based bio-edible films, as pectin is abundantly available in the peels of fruits and vegetables, and also because it is economically advantageous. Pectin is an anionic compound having 1-4-D-galacturonic acid residues. The property of pectin varies depending upon the degree of methyl esterification of uronic acid, based on which it is classified as high methoxy pectin (HMP) or low methoxy pectin (LMP) [5]. HMP forms excellent films. Pectin is a long chain of Homogalacturonan (HG), Xylogalacturonan (XGA), and Rhamnogalacturoanan (RGA). However, HG forms 60% of it, and the rest is formed by XGA and RGA [1,5].

2. Materials and Methods

Chemical-grade pectin powder was brought from Loba Chemie Pvt. Ltd. in Mumbai, India. The molecular weight varied from 30,000 to 100,000. The methoxyl concentration in pectin was 6–10%, and the DE (degree of esterification) was 63–66%. Beeswax and castor oil were procured from local vendor. Other chemicals, such as sodium hydroxide buffer tablets, were procured from Sigma-Aldrich. Double-distilled water was made in our lab.

2.1. Film Development

The experiments proceeded with the preparation of 5% weight/volume (w/v) pectin-based control films, where 5 g of pectin powder was agitated in 100 mL of distilled water. Lab scale magnetic stirrer was used to agitate and homogenize the film-forming base solution at a temperature of 45 °C for 30 min and at 500 rpm stirrer speed. During the last 10 min of this process, 0.5 g of emulsifier was added to help the immiscible components combine. The solution was cooled to ambient temperature, and the power of hydrogen (pH) was checked; it was around 2.8, which was because of the acidic nature of pectin. It was adjusted to be around 3 using a diluted sodium hydroxide solution. Furthermore, the solution was mixed in the magnetic stirrer along with a melted mixture of beeswax, castor oil, and clove oil of appropriate composition. The solution was further homogenized using a homogenizer (IKA T25 ULTRA-TURRAX) at speed of 4000–6000 rpm. To achieve proper mixing of the small particles, the solution was ultrasonicated for 45 min. A total of 40 mL of perfectly homogenized solution was poured into lab scale Petri dish of 100 mm diameter by spreading it uniformly. These were then kept in the humidity chamber (NECSTAR NEC-HTC-150) to dry the films at a controlled temperature and relative humidity (40–50%). The dried films were peeled and stored in a desiccator containing silica gel.

2.2. Characterization

2.2.1. Moisture Content and Water Solubility

To analyze the moisture content bound in the film, a 4 cm × 4 cm film was kept in a hot air oven for 3 h at a temperature of 110 ± 5 °C. The percent weight loss against the initial weight of the film was calculated as $((W_{\text{initial}} - W_{\text{final}})/W_{\text{initial}}) \times 100$, where W_{initial} is the weight of the film and W_{final} is the weight of the dry film. To confirm the results, the analysis was performed by taking at least three samples from the same film.

Similarly, to analyze the water solubility of a film, a 3 cm × 3 cm was taken and soaked in 15 mL of water for 30 min. The time taken for the film to completely dissolve was noted.

2.2.2. Thickness

The thickness of the film developed by spreading 40 mL of film solution on a 100 mm diameter Petri dish was measured. To measure the thickness of this film, a micrometer screw gauge with a least count of 0.001 mm was used. The thickness at five random positions on the 100 mm diameter film was measured, and the average was reported as the film thickness.

2.2.3. Optical Property or Transparency

The transparency analysis of the film was performed using a spectrometer (CHN SPEC & CS-580 A). The calibration was performed by keeping the film over a white surface on which the spectrometer gave a color analysis value. Film color was measured by lightness/luminosity (*L**), chromaticity (*a**), and chromaticity (*b**) on the color scale. *L** values gave the range from black (zero) to white (100), *a** ranged from green (negative) to red (positive), and *b** ranged from blue (negative) to yellow (positive). The analysis was repeated at 5 different areas of the film, and the average was considered.

$$\Delta E = \sqrt{(L^* - L)^2 + (a^* - a)^2 + (b^* - b)^2} \tag{1}$$

The standard values are L^* = 86.75, a^* = 0.93, and b^* = -1.03; total transparency or color difference parameter is ΔE.

2.2.4. WVTR

The water vapor transmission rate was analyzed due to the fact that less moisture from the food should be transmitted out so that desirable moisture is present in the food to keep up the freshness of the food. It is usually measured in $g/m^2/day$. The analysis was performed by using the facility at Northern India Textile Research Association, Ghaziabad, India, and Sree Chitra Tirunal Institute For Medical Sciences & Technology, Thiruvananthapuram, India. It was performed according to the ASTM standard ASTM E96/E96M-05 (water method), maintained at a temperature of 32 $\pm$ 2 °C and relative humidity of 50 $\pm$ 2%, with air velocity set between 0.02 and 0.3 m/s.

2.2.5. Water Contact Angle and Mechanical Properties

The water contact angle (WCA) was found using the sessile drop method using a device that measures the contact angle between the film and the water droplet as video data (Data Physics OCA 15 Plus, Germany). The analysis was performed at Sree Chitra Trunal Institute For Medical Sciences & Technology, Thiruvananthapuram, Kerala, India. Mechanical properties of the films were studied at Sree Chitra Trunal Institute For Medical Sciences & Technology, Thiruvananthapuram, Kerala, India. The study was performed based on the ASTM D882 method. The analysis was performed on a film of 10 mm width $\times$ 150 mm length over a crosshead speed of 10 mm/min.

2.2.6. FTIR

FTIR analysis of the films was performed with a Fourier transform infrared spectrophotometer. The spectrum of each sample of the film was found in the range of 4000–500 cm^{-1} with a resolution of 4 cm^{-1}. The results obtained from the FTIR analysis provided insights into the structural properties of the film.

3. Results and Discussion

All the pectin films with additives such as castor oil, beeswax, and clove oil were developed as per the 2^3 (two-level, three-factor) statistical design of experiments. The details of the experiments are given in Table 1. The films were flexible and easy to peel off from the Petri dish. All the films were developed from a 5% *w*/*v* (5 g of pectin per 100 mL of water) concentration film-forming solution. The other process parameters, such as pH (=3), and drying conditions, such as temperature (40 °C) and relative humidity (60%), were kept constant based on our previous studies. The films were kept free from atmospheric moisture and other contaminations by storing them in a vacuum desiccator.

Table 1. Details of low and high levels of functional additives.

Run No	Castor Oil (% *w/w* of Pectin)	Beeswax (% *w/w* of Pectin)	Clove Oil (% *w/w* of Pectin)
1	5 (low)	5 (low)	2 (low)
2	15 (high)	5	2
3	5	10 (high)	2
4	15	10	2
5	5	5	4 (high)
6	15	5	4
7	5	10	4
8	15	10	4

3.1. Water Solubility

All the films were observed to be completely water soluble. When a 5 cm × 5 cm film was dipped in 20 mL of water, within 20 min, almost all the film dissolved. However, a minute amount of beeswax was visible floating on the surface of the water based on the amount of wax added. This is because of the hydrophobic nature of beeswax.

3.2. Thickness

The beeswax, castor oil, and clove oil-integrated pectin films developed were observed to be thin. This can be confirmed by Figure 1, where the thickness of the films is in the range of 0.12 ± 0.004–0.15 ± 0.004 mm.

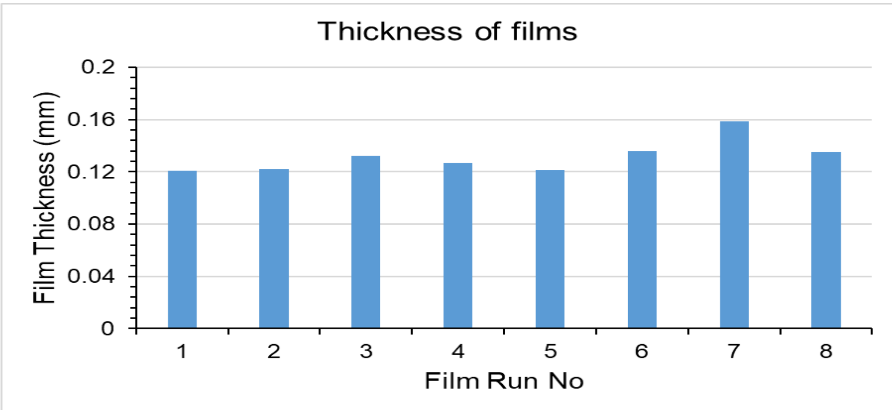

Figure 1. The thickness of films (run number as per Table 1).

3.3. Transparency

Figure 2 shows that, all the films were transparent enough when compared with a transparent plastic sheet. The transparency parameter Δ*E* of the films was observed to be in the range of 15 ± 2–20 ± 2. Δ*E* values of less than 50 are usually referred to as transparent films. Therefore, the beeswax, castor oil, and clove oil-integrated pectin films developed in the present work are transparent.

3.4. WVTR

The moisture transmission to and from the food material will cause drying and fungal growth. Therefore, the packaging film must possess good moisture barrier properties. The moisture barrier property of the films was studied in terms of water vapor transmission rate (WVTR). The WVTR of the beeswax, castor oil, and clove oil-integrated pectin films developed in the present work is in the range of 1017.12 g/m^2/day–1739.73 g/m^2/day. The WVTR of the control pectin is 1815.70 g/m^2/day. A comparison of the WVTR values of the eight composite films and the control pectin film is shown in Figure 3. This shows that the hydrophobic nature of beeswax, castor oil, and clove oil reduced the WVTR to a significant level. It was observed that run no 8, which has a higher percentage of beeswax, castor oil, and clove oil, showed the lowest WVTR. This means that the highest moisture barrier property is exhibited by run no. 8.

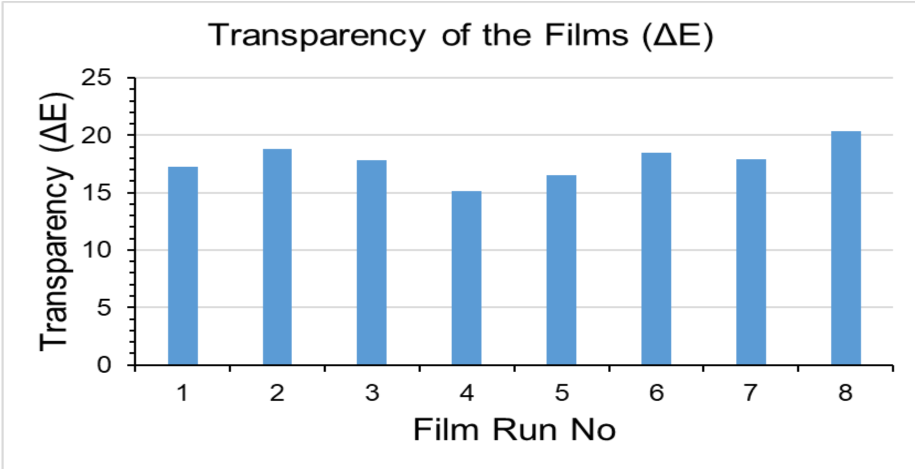

Figure 2. Transparency of the films (run number as per Table 1).

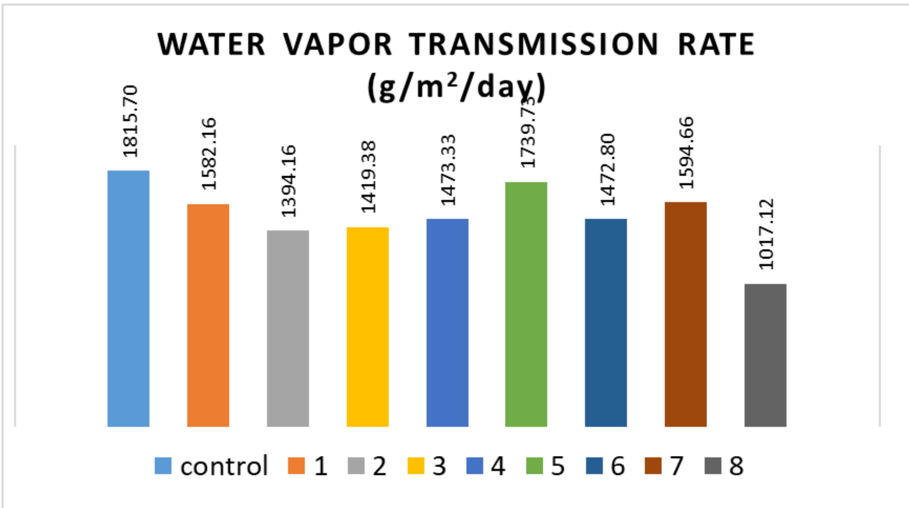

Figure 3. Water vapor transmission rate of the films (run number as per Table 1).

3.5. Water Contact Angle and Mechanical Properties

The water contact angle of the films was observed to be in the range of 75–85 $\pm$ 5° after 12 s of the sessile drop method. This is considerably higher than the control pectin film, for which the water contact angle is 62°. The tensile strength of the film was in the range of 8–12 $\pm$ 3 MPa, with an elongation at break of 15% at a strain rate of 10 mm/min. At similar strain rates, the control pectin film has a tensile strength of 15 MPa and elongation at break of 3%. This indicates that the additives beeswax, castor oil, and clove oil improved the mechanical properties.

3.6. FTIR

The Fourier transform infrared spectroscopy (FTIR) of the four film samples is shown in Figure 4. Figure 4 clearly shows that all the films are of the same nature with only a slight difference in the intensity of transmittance. Figure 4 also shows that no chemical transformations happened during the film-forming process and solvent evaporation.

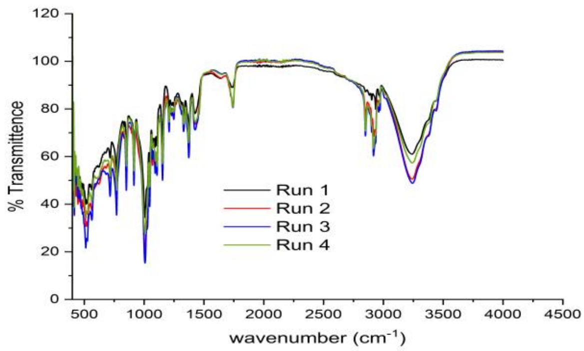

Figure 4. FTIR of the films (run number as per Table 1).

4. Conclusions

The present work reports pectin biopolymer-based films for food packaging applications. Beeswax, castor oil, and clove oil were used as hydrophobic and plasticizer additives. The films developed were thin, transparent, and easily soluble in water. Furthermore, the films showed excellent moisture barrier properties compared to control pectin films. The films were also found to be biodegradable. The results suggest that pectin-based films with beeswax, castor oil, and clove oil have the potential to be used as edible packaging materials with enhanced mechanical, barrier, and antimicrobial properties. This research can contribute to sustainable food packaging practices with biodegradable films.

Author Contributions: A.D. has done the experiments. Y.S.S. is responsible for analyzing the results and manuscript writing. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Acknowledgments: The WVTR analysis was made possible with the support of the Sree Tirunal Institute Chitra for Medical Sciences & Technology, Trivandrum, Kerala, India, and the Northern India Textile Research Association (NITRA), Ghaziabad, India.

Conflicts of Interest: The authors confirm that there is no conflict of interest.

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