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Development of Pectin and Sodium Alginate Composite Films with Improved Barrier and Mechanical Properties for Food-Packaging Applications †

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Abstract: The rate of plastic deterioration is currently significantly outpacing the rate at which plastic waste is being produced, leading to a biome-wide imbalance. Biopolymers derived from sustainable raw materials are widely explored as potential alternative packaging materials to increase the shelf-life of fresh produce and processed food. The present work aims to develop polysaccharide-based composite films. Sodium alginate- and castor oil-blended pectin films were developed as per the 2^3 (two-level three-factor) factorial design of experiments. Sodium alginate was used as a stabilizer and film-forming agent to enhance the mechanical properties of the films. Castor oil was used as an additive to improve the moisture barrier and antimicrobial properties. D-sorbitol was used as a plasticizer to improve the flexibility of the films. The amounts of sodium alginate (25% and 50% *w*/*w*), castor oil (10% and 15% *w*/*w*), and D-sorbitol (15% and 30% *w*/*w*), with respect to pectin, and the sonication time were chosen as the four factors. Based on our prior optimization studies, all other process variables, such as pH (<4), drying temperature (60 °C), and humidity (40%), were maintained constant. The moisture barrier, mechanical, surface hydrophobicity, morphological, thermal stability properties, and biodegradability characteristics of each film were studied. All films were thin (~0.110 ± 0.004 mm) and transparent (ΔE = 3 to 11). The moisture barrier properties improved three-fold compared to pure pectin films. The elongation at break increased at least three times. The films were thermally stable at 400 °C. The melting point of the films increased to 150 °C, compared to 95 °C of the pure pectin film.

Keywords: biopolymers; food safety; polysaccharide; design of experiments; moisture barrier

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1. Introduction

Demand for food packaging is increasing at a significant rate. Food packaging was predicted to have a global market value of around USD 363 billion in 2022. According to the report of the global food-packaging market, it is expected to reach USD 512 billion by 2028 [1]. Most of the available synthetic polymers are used in food packaging, which take years to degrade and are harmful for human life. The environmental concern has led to a need for an alternative to the synthetic polymers, which has directed research towards creating packaging with natural biodegradable and edible products. Biodegradable products do not harm human health and are quick to degrade in the environment. These biodegradable products are based on biopolymers such as carbohydrates, lipids, fats, and proteins, which have the tendency to form homogeneous gels when mixed with water [2].

Pectin covers a broad group of complex polysaccharides found naturally in the primary cell walls and middle lamella of plants. Fruits and vegetables such as apples, citrus fruits, berries, green beans, carrots, tomatoes, and others have high amounts of pectin [3,4].

Pectin is primarily used in the food sector as a gel-forming agent and a stabilizer, but nowadays it is being popularized as a fat replacement option and an ingredient that promotes good health [5–8].

Furthermore, pectin is proving to be an effective ingredient in the food-packaging industry. Though synthetic polymers and plastics have their advantages, in today's world of sustainable development, non-biodegradable materials are a serious issue. Pectin, in the presence of other biopolymers and components, is turning out to be a game changer [9].

In this regard, the current research centers on developing edible biodegradable films for food packaging using pectin and sodium alginate. Various key parameters, such as the concentration of the plasticizer and stabilizer, pH, stirring time, sonication, and the temperature and humidity for drying, had an effect on the final output. The formed films were tested for transparency, thickness, moisture barrier, and mechanical properties, and the thermal stability.

2. Materials and Method

Chemical-grade extra-pure pectin was purchased from Loba Chemie Pvt. Ltd., Mumbai, India, which has a molecular weight range of 30,000 to 100,000 g/mole. The degree of esterification (DE) of the pectin was 63–66%, whereas the methoxyl content was 6–10%. Pure castor oil was procured from a local vendor by cold-pressed extraction. Double-distilled water was from our lab.

2.1. Film Preparation

Films were prepared in a lab-scale (100 mm diameter) petri dish. Initially, the control pectin films (2.5% weight/volume) were made by incorporating 2.5 g of pure pectin powder in 100 mL of distilled water. This was mixed using a magnetic stirrer at 45 °C and 600–700 rpm at a constant rate for 20–25 min. This solution was cooled and further homogenized to completely disperse the remaining larger particles into the solution using a Homogenizer (IKA T25 ULTRA-TURRAX, Panacea Instruments, New Delhi, India) at 3000–4000 rpm. This film-forming solution was then passed through a muslin cloth to filter out any bigger particles. The pH of this solution was measured using a pH meter, which was adjusted to an approximate value of 3, using sodium hydroxide solution. Smaller particles suspended in the solution were further solubilized using an Ultra-Sonicator (E-Chrom Tech make Ultrasonic Processor UP 100, Panacea Instruments, New Delhi, India) to ensure there were no bubbles present in the final solution. Around 40–50 mL of this solution was poured into petri dishes and placed in a humidity chamber (NECSTAR NEC-HTC-150, The Royal Technologies, New Delhi, India) at a constant relative humidity of 60% and a temperature of 40 °C, for a sufficient period of time. Once dried, the films were peeled off and stored in a vacuum desiccator containing silica gel. Sodium alginate (a stabilizing polysaccharide), D-sorbitol (a plasticizer), and pure castor oil were added for a more flexible film that demonstrated better barrier and mechanical properties. These were added in stages with successive heating of the solution at a constant temperature of 45 °C for 20–25 min, before adding the next ingredient. An emulsifier was used to maintain a uniform distribution of the castor oil in the water. A similar procedure of film-forming and drying was implemented to prepare the films based on the statistical design of experiments.

2.2. Characterization of Films

2.2.1. Thickness

Films prepared by pouring 50 mL of film-forming solution into a 100 mm-diameter petri dish were measured by a micrometer screw gauge with at least a count of 0.001 mm. The thickness at five arbitrary points was measured and the average was reported.

2.2.2. Optical Properties (Transparency)

An analysis of the transparency was performed using a CHN SPEC, CS-580A Spectrophotometer. Calibration was conducted using a transparent film to obtain standard values (L* = 89.16, a* = −1.13, b* = 3.6). L, a, and b values of the prepared films were measured by setting the film on a white plate. These values were found at five arbitrary points and the average L, a, and b values were reported. Further, these values were used to calculate the total color difference or transparency parameter (ΔE) using Equation (1):

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

2.2.3. Water Vapor Transmission Rate (WVTR)

WVTR analysis (the water method) was performed at Sree Chitra Tirunal Institute for Medical Sciences and Technology, Thiruvananthapuram, India, as per ASTM E96/E96M–16. The procedure was performed at a temperature of 37 °C and relative humidity of 50 ± 2%. The WVTR for the given samples were reported as $g/h{\cdot}m^2$.

2.2.4. Water Contact Angle

The water contact angle was measured at room temperature (approximately 23 °C) using the sessile drop method by a video-based contact measuring device (Data Physics OCA15 plus, DataPhysics, Filderstadt, Germany) and imagining software (SCA20 software, DataPhysics, Filderstadt, Germany) within 10 s after the introduction of water droplets. It was calculated at six arbitrary points and the average was reported. Analysis was performed by using the facility at Sree Chitra Tirunal Institute for Medical Sciences and Technology, Thiruvananthapuram, India.

2.2.5. Mechanical Properties

The test method was taken from ASTM D882. The sample was fixed between upper and lower pneumatic grips with a gauge length of 100 mm. A crosshead speed of 10 mm/min was used. Strips of 10 mm in width and 150 mm in length were taken from the specimens for analysis. Analysis was performed at the facility at Sree Chitra Tirunal Institute for Medical Sciences and Technology, Thiruvananthapuram, India.

2.2.6. Fourier-Transform Infrared Spectroscopy (FTIR)

FTIR analysis was performed using a Fourier-transform infrared spectrophotometer. Spectra were acquired in the range of 4000 to 500 cm^{-1} with a resolution of 4 cm^{-1}. The results of the FTIR study provided information on the chemical composition and structural features of the film.

2.2.7. Shelf-Life Test

To coat the fruits (here, we selected capsicum and chili), a film solution was prepared, and the fruits were continuously dipped until the solution was even. Dipping was repeated for 3–4 days for an even coating.

2.3. Experimental Design

A two-level three-factor (2^3) design of experiments was implemented to understand the effect of three factors: the concentrations of sodium alginate, castor oil, and D-sorbitol, on various properties. The amount of pectin used was constant at 15 gm in 600 mL of distilled water. Table 1 describes the various runs conducted by varying the three factors mentioned above.

Table 1. High, low, and medium levels of sodium alginate, castor oil, and D-sorbitol.

Run No.	Sodium Alginate (g)	Castor Oil (g)	D-Sorbitol (g)
1	7.5 (High)	2.25 (High)	4.5 (High)
2	3.75 (Low)	2.25	4.5
3	7.5	1.5 (Low)	4.5
4	3.75	1.5	4.5
5	7.5	2.25	2.25 (Low)
6	3.75	2.25	2.25
7	7.5	1.5	2.25
8	3.75	1.5	2.25
9	5.625 (Medium)	1.875 (Medium)	3.375 (Medium)

3. Results and Discussion

3.1. Thickness and Optical Properties (Transparency)

The observed thicknesses of the films were in the range of 0.11 ± 0.004 mm. All prepared films were transparent, with ΔE in the range of 8 to 20.

3.2. Water Vapor Transmission Rate (WVTR)

The WVTR of the "pectin + sodium alginate + castor oil + D-sorbitol" composite films were observed to be in the range of 80 g/m^2/h as shown in Figure 1. Compared to the control pectin films, the WVTR slightly increased. This could be attributed to the presence of hydrophilic sodium alginate and sorbitol, which were used as a plasticizer. This observation is visible from the WVTR comparison of the control pectin and pectin + D-sorbitol. This suggests that D-sorbitol, which is usually a good plasticizer, may not be a suitable additive due to its hydrophilic nature as it compromised the moisture barrier properties of the pectin films.

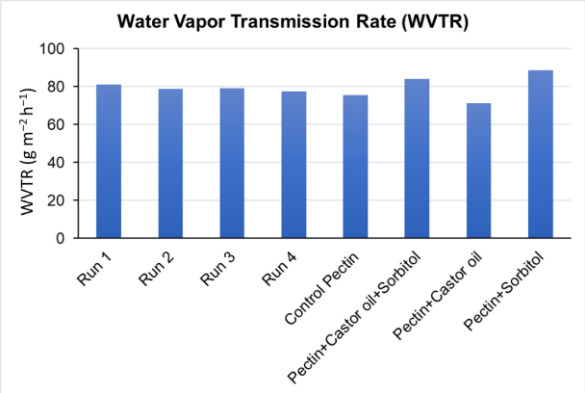

Figure 1. WVTR of the "pectin + sodium alginate + castor oil + sorbitol" films.

3.3. Water Contact Angle (WCA)

The surface hydrophobicity of a thin film can be understood with the help of the WCA. The WCA values after 12 s of sessile drop of two of the composite films and the comparison with the control pectin film are presented in Figure 2. The WCAs of Run 1 and Run 2 were 42.45° and 54.18°, as per Figure 2, which are less than that of the control pectin (67.92°). Therefore, it is evident that in a way, the hydrophilic plasticizer sorbitol reduced the surface hydrophobicity of the pectin films.

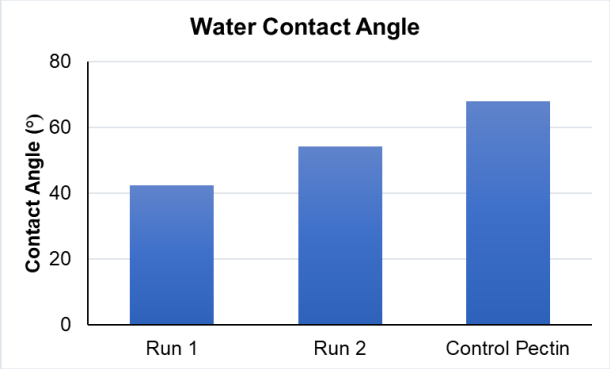

Figure 2. Comparison of the water contact angles.

3.4. Mechanical Properties

The mechanical properties such as tensile strength, load at break, and percent strain at break are compared in Figure 3. The mechanical properties suggest that a low concentration of sodium alginate and castor oil and a high concentration of sorbitol improved the flexibility and strength of the films. This was apparent from the improved elongation at break for Run 2 and Run 3, compared to Run 1 (composition of runs as per Table 1). As per Figure 3, Run 4 had the optimum concentration in terms of acceptable mechanical properties.

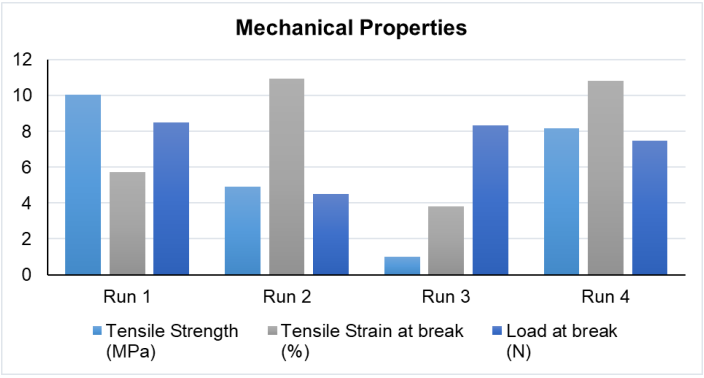

Figure 3. Comparison of mechanical properties.

3.5. Fourier-Transform Infrared Spectroscopy (FTIR)

Figure 4 shows the comparison of the FTIR of the "pectin + sodium alginate + castor oil + D-sorbitol" films with the pure components. The results suggested that no new chemical bonds formed during the film preparation process, and that the film was an outcome of a purely physical process. Peaks at 3300 cm^{-1} were linked to the O–H stretching vibrations of the hydroxyl groups in pectin, whereas peaks at 1650 cm^{-1} were related to the C=O stretching vibration of the carbonyl group in pectin. Peaks at 2920 cm^{-1} and 2850 cm^{-1} were caused by the castor oil's aliphatic chains' C–H stretching vibrations.

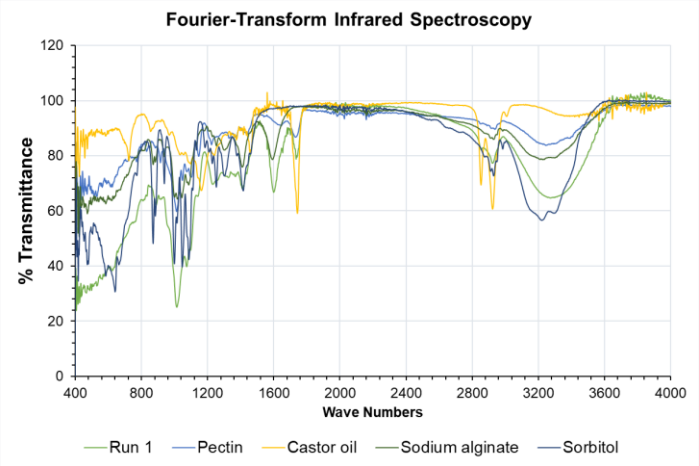

Figure 4. FTIR of film Run 1.

3.6. Shelf-Life Test

As depicted in Figures 5 and 6, the food products coated with the film-forming solution were proven to last longer than those left undisturbed, thus proving that the films extended the shelf-life of the food products.

(**a**) (**b**)

Figure 5. Pictures from Day 3: (**a**) the control chili, kept to compare with the coated one, was wrinkled, and (**b**) the chili coated with the film-forming solution stayed fresh.

(**a**) (**b**)

Figure 6. Pictures from Day 9: (**a**) the control capsicum, kept to compare with the coated one, was wrinkled, and (**b**) the capsicum coated with the film-forming solution stayed fresh.

4. Conclusions

The current study discussed the effect of functional additives such as sodium alginate, castor oil, and sorbitol on improving the properties of pectin-based films. Sodium alginate and D-sorbitol proved to be good plasticizers as they provided flexibility that helped improve the mechanical properties of the films. However, these two hydrophilic components indeed reduced the moisture barrier properties of the films. Castor oil had reduced moisture transmission as it is a good hydrophobic agent. The formed films were thin and transparent, with better moisture barriers and excellent mechanical properties. When tested on food products, they enhanced the shelf-life of the products. The findings in this paper point to the possibility of pectin-based films including additives such as sodium alginate, castor oil, and D-sorbitol in the food-packaging industry to improve the

mechanical and barrier properties. To better understand how they operate in practical packaging applications, more research will be needed. This study might help in developing biodegradable film packaging techniques for food that is to be sustainably packaged. This work provides a strong basis for the future to find out about more such sustainable methods of food packaging that would prove to be better for the environment.

Supplementary Materials: The presentation materials can be downloaded at: https://www.mdpi.com/article/10.3390/ECP2023-14668/s1.

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Conflicts of Interest: The authors declare no conflict of interest.

References

1. Statista. Global Market Value of Food Packaging from 2022–2028. Available online: https://www.statista.com/statistics/876489/food-packaging-market-value-forecast-worldwide/ (accessed on 3 April 2023).
2. Mehraj, S.; Sistla, Y.S. Optimization of process conditions for the development of pectin and glycerol based edible films: Statistical design of experiments. *Electron. J. Biotechnol.* **2022**, *55*, 27–39. [CrossRef]
3. Mohnen, D. Pectin structure and biosynthesis. *Curr. Opin. Plant Biol.* **2008**, *11*, 266–277. [CrossRef] [PubMed]
4. Honeycutt, L. List of Foods High in Pectin. LIVESTRONG.COM. Available online: https://www.livestrong.com/article/289067-list-of-foods-high-in-pectin/ (accessed on 13 September 2019).
5. Min, B.; Bae, I.Y.; Lee, H.G.; Yoo, S.H.; Lee, S. Utilization of pectin-enriched materials from apple pomace as a fat replacer in a model food system. *Bioresour. Technol.* **2010**, *101*, 5414–5418. [CrossRef] [PubMed]
6. Ciriminna, R.; Chavarria-Hernandez, N.; Hernandez, A.I.R.; Pagliaro, M. Pectin: A new perspective from the biorefinery standpoint. *Biofuels Bioprod. Biorefining* **2015**, *9*, 368–377. [CrossRef]
7. Peng, Q.; Xu, Q.; Yin, H.; Huang, L.; Du, Y. Characterization of an immunologically active pectin from the fruits of lycium ruthenicum. *Int. J. Biol. Macromol.* **2014**, *64*, 69–75. [CrossRef]
8. Dranca, F.; Oroian, M. Extraction, purification and characterization of pectin from alternative sources with potential technological applications. *Food Res. Int.* **2018**, *113*, 327–350. [CrossRef]
9. Mellinas, C.; Ramos, M.; Jiménez, A.; Garrigós, M.C. Recent Trends in the Use of Pectin from Agro-Waste Residues as a Natural-Based Biopolymer for Food Packaging Applications. *Materials* **2020**, *13*, 673. [CrossRef] [PubMed]