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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. Present work aims to develop polysaccharide based composite films. Sodium alginate and castor oil blended pectin films were developed as per 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 moisture barrier and antimicrobial properties. D-Sorbitol was used as a plasticizer to improve the flexibility of the films. The amount of sodium alginate (25% & 50% *w*/*w*), castor oil (10% & 15% *w*/*w*) and D-sorbitol (15% & 30% *w*/*w*) with respect to pectin and 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 threefold 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 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 363 billion dollars 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 takes years to degrade and is harmful for human life. Environmental concern with an alternative for the synthetic polymer which has directed for researching on packaging with natural biodegradable and edible products available to us. Biodegradable products don't harm human health and digest soon. These biodegradable products are based on biopolymers such as carbohydrates, lipids, fat and protein which has the tendency to form gels when mixed with water and which upon mixing gives a homogeneous gel [2].

Pectin are a broad group of complex polysaccharides found naturally in the primary cell walls and middle lamella of plants. Fruits and vegetables like apples, citruses, 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 now 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 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 concentration of plasticizer and stabilizer, pH, stirring time, sonication, temperature and humidity for drying, had an effect on the final output. The film formed were tested for transparency, thickness, moisture barrier and mechanical properties and 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 30,000–100,000 g/mole. Degree of esterification (DE) of the pectin was 63–66% whereas methoxyl content was 6–10%. Pure castor oil was procured from a local vendor by cold pressed extraction. Double distilled water was made from our lab.

2.1. Film Preparation

Films were prepared in a lab scale (100 mm diameter) petri dish. Initially control pectin films (2.5% weight/volume) were made by incorporating 2.5 gm 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) 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 was further solubilized using an Ultra-Sonicator (E-Chrom Tech make Ultrasonic Processor UP 100) 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) at a constant relative humidity of 60% and temperature of 40 °C, for 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 castor oil in water. A similar procedure of film forming and drying was implemented to make the films based on the prepared statistical design of experiments.

2.2. Characterization of Films

2.2.1. Thickness

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

2.2.2. Optical Properties (Transparency)

Analysis of transparency was performed using 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, 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 average L, a, b values were reported. Further, these were used to calculate the total colour difference or transparency parameter (ΔE) using equation (1), mentioned below:

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

2.2.3. Water Vapor Transmission Rate (WVTR)

WVTR Analysis (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 in the units of g/h.m^2.

2.2.4. Water Contact Angle

The water contact angle is measured at room temperature (approx. 23° C) using sessile drop method by a video-based contact measuring device (Data Physics OCA15 plus, Germany) and imagining software (SCA20 software, Germany) within 10 s after 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 & Technology, Thiruvananthapuram, India.

2.2.5. Mechanical Properties

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

2.2.6. Fourier-Transform Infrared Spectroscopy (FTIR)

FTIR analysis is studied using Fourier transform infrared spectrophotometer. Spectrum was acquired in the range of 4000–500 cm^{-1} with a resolution of 4 cm^{-1}. The result of the FTIR study gave us 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), 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—concentration 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 taken.

Run No.	Sodium Alginate (gm)	Castor Oil (gm)	D-Sorbitol (gm)
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 thickness of the films were in the range of 0.11 ± 0.004 mm. All films prepared 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 observed to be in the range of 80 g/m²/h. compared to control pectin films the WVTR has slightly increased. This could be attributed to the presence of hydrophilic sodium alginate and sorbitol which was used as plasticizer. This observation is visible from the WVTR comparison of control pectin and pectin + D-sorbitol. This suggests that the D-sorbitol which is usually a good plasticizer, may not be suitable additive due to its hydrophilic nature as it compromises the moisture barrier properties of the pectin films.

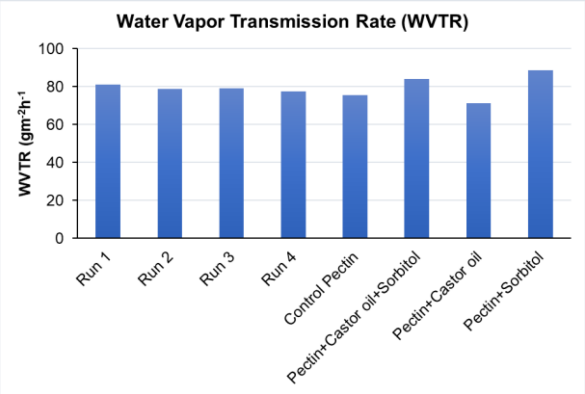

Figure 1. WVTR of the "Pectin + sodium alginate + castor oil + sorbitol" films.

3.3. Water Contact Angle (WCA)

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

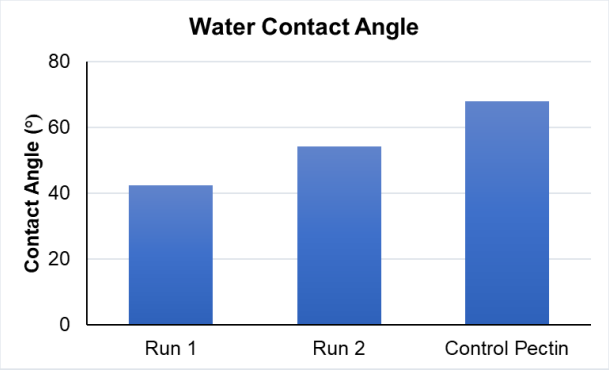

Figure 2. Comparison of Water Contact Angle.

3.4. Mechanical Properties

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

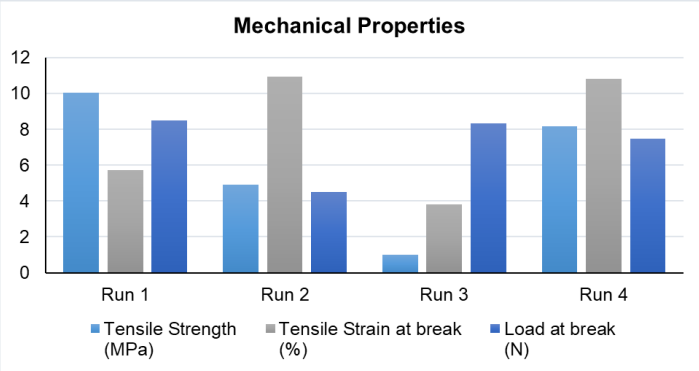

Figure 3. Comparison of Mechanical Properties.

3.5. Fourier-Transform Infrared Spectroscopy (FTIR)

Figure 4 shows the comparison of FTIR of "pectin + sodium alginate + castor oil + D-sorbitol" films with pure components. It suggests that no new chemical bonds formed during the film preparation process and that the film is an outcome of a pure physical process. Peaks at 3300 cm^{-1} are linked to the O–H stretching vibrations of the hydroxyl groups in pectin, whereas peaks at 1650 cm^{-1} are 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.

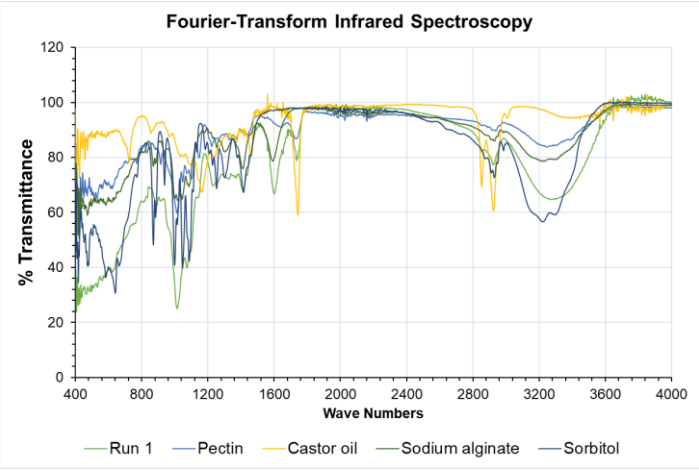

Figure 4. FTIR of film Run 1.

3.6. Shelf Life Test

As depicted in Figures 5 and 6, the food products coated with the film forming solution proved to have lasted longer than the ones left undisturbed. Thus, proving that the films extend the shelf life of food products.

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

Figure 5. Pictures as on Day 3; (**a**) Control Chili kept to compare with the coated one wrinkled; (**b**) Chili coated with film forming solution stayed fresh.

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

Figure 6. Pictures as on Day 9; (**a**) Control Capsicum kept to compare with the coated one wrinkled; (**b**) Capsicum coated with film forming solution stayed fresh.

4. Conclusions

The current study discusses the effect of functional additives such as sodium alginate, castor oil and sorbitol to improve 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 film. However, these two hydrophilic components have indeed reduced the moisture barrier properties of the films. Castor oil has reduced moisture transmission as it is a good hydrophobic agent. The films formed were thin, transparent, with better moisture barrier 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 like sodium alginate, castor oil and D-sorbitol in the food packaging industry to have improved mechanical and barrier properties. To better understand how they operate in practical packaging applications, more research will be needed. This study might help develop biodegradable film packaging techniques for food that is packaged sustainably. This provides a strong base for the future to find out about more such sustainable ways of food packaging that would prove to be better for the environment.

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

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