

## Research Article

# Comparison of the Film Properties of Lemon and Sour Cherry Seed Essential Oil-Added Glycerol and/or Sorbitol-Plasticized Corn, Potato, Rice, Tapioca, and Wheat Starch-Based Edible Films

## Merve Basut Kazak i and Nurcan Tugrul

Faculty of Chemistry and Metallurgy, Department of Chemical Engineering, Davutpaşa Campus, Yıldız Technical University, İstanbul, Türkiye

Correspondence should be addressed to Nurcan Tugrul; ntugrul@hotmail.com

Received 29 February 2024; Revised 9 April 2024; Accepted 22 April 2024; Published 30 April 2024

Academic Editor: Huining Xiao

Copyright © 2024 Merve Basut Kazak and Nurcan Tugrul. This is an open access article distributed under the Creative Commons Attribution License, which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited.

In this study, lemon, and sour cherry seed essential oil-added glycerol and/or sorbitol-plasticized corn, potato, rice, tapioca, and wheat starch-based edible films were produced using the casting method. Starch, essential oil type and glycerol and/or sorbitol effects on the thickness, moisture content, water solubility, swelling index, and water vapor transmission rate of the films have been studied. The interaction of the film components was evaluated by Fourier transform infrared spectroscopy. It was seen that wheat starch-based control films give the lowest thickness value (0.010 mm). Wheat starch-based control films (15.50%), sour cherry seed essential oil-added corn starch (17.80%), and lemon essential oil-added rice starch-based composite films (17.70%) have high moisture content. The lowest solubility values were obtained from wheat starch control (22%) and sour cherry seed essential oil-added corn starch composite (16.40%) films. The highest swelling index values were obtained from wheat starch-based (433.20%) composite films. Rice starch-based control films have the lowest water vapor transmission rate ( $3.30 \times 10^{-8} - 5.70 \times 10^{-8}$ ). FTIR spectra of edible composite films proved that there is no chemical interaction between the film component and that they kept their structure. The main difference of this study from previous studies was the use of sour cherry seed essential oil for the first time in edible film production and the comparison of the film properties of corn, potato, rice, tapioca, and wheat starch-based edible films plasticized with glycerol or sorbitol.

## 1. Introduction

Packaging is especially important in the food industry since it will protect the quality and freshness of the food from the packaging to the consumption and provide to store them [1, 2]. Petroleum-based plastic materials are widely used in food packaging, but they are not degradable and environmentally friendly. So, investigation of alternative biodegradable polymers has been inevitable. Materials used for coating different foods to increase shelf life, which are eaten together with the product, are considered as an edible film. Biodegradable edible films decrease moisture loss, oxygen, and other gas immigration increasing the food shelf life. Several biopolymers, such as starch, protein isolates, pectin, and lipids, are commonly used to prepare edible packaging films. Especially starch has been widely investigated due to its property of forming a continuous matrix, being renewable and available in nature [3–5]. The characteristics of the films formed with starches from various plants are different because of the distinction in particle size, shape, amylopectin/amylose ratio, crystallinity, etc. Global production of starch supply is 75% from corn, 14% from cassava, 7% from wheat, and 4% from potatoes. Corn starch, with high biodegradability, is a mostly used starch polymer for edible film production [6–9].

Native starch edible films have some disadvantages like brittleness and hydrophilic nature. To avoid these drawbacks, plasticizers, emulsifiers, antioxidants, and antimicrobial compound (essential oils) are added to the starch-based edible films, because they increase the barrier and mechanical properties by reducing intermolecular forces, and the mobility of biopolymer chains. Glycerol, sorbitol, propylene glycol, and other polyols are the most used plasticizers [2, 10-13]. Essential oils such as lemon, orange, lime, cinnamon, eucalyptus, and anise are added to the films to ensure antioxidative, antimicrobial, and insecticidal properties and develop barrier properties of the films, due to their hydrophilic nature [14, 15]. Although starch has good filmforming properties, the mechanical properties of the edible films produced are poor. Plasticizers, cross-linking agents, and antibacterial and antioxidant agents can be added to the film-forming solution to improve the film properties of starch-based edible films. Sodium alginate is frequently used in edible film production due to its properties such as thickening, stabilization, suspension formation, film formation, gelatinization, emulsion stabilization, nonflammability, biodegradability, and biocompatibility [16, 17].

The aim of this study was to compare the film properties of corn, potato, rice, tapioca, and wheat starch-based edible films. Effects of starches, essential oil types and glycerol and/or sorbitol on the thickness, moisture content, water solubility, swelling index, and water vapor transmission rate of the edible films were investigated. The most important difference of this study from previous studies was the use of sour cherry seed essential oil for the first time in edible film production. The outcomes of this work will enable to determine the appropriate type of starch, essential oil, and plasticizer to produce long shelf-life edible food packaging films.

#### 2. Materials and Methods

2.1. Materials. The natural corn, potato, rice, tapioca, wheat starches, lemon essential oil (LEO), and sour cherry seed essential oil (SCSEO) used in this study were purchased from Arifoğlu, Istanbul, Turkey. Glycerol was bought from Merck (Darmstadt, Germany), D-sorbitol was bought from Carlo Erba (France), and sodium alginate was bought from AFG Bioscience (USA). All chemicals and solvents used in this study were of analytical grade. The natural starches were dried in an oven at 60°C for 24 h to remove any moisture; then, the dried starches were milled, and the resulting peel powder was stored in low-density polyethylene bags at room temperature.

2.2. Edible Film Preparation. The solution casting method used in past work was used to produce starch-based edible films, with some changes [7, 18]. Starch-based control film solutions were prepared by dissolving starch (3 g), sodium alginate (0.5 g), and different ratios of glycerol and/or sorbitol (indicated in Table 1) in 100 ml distilled water. Starchbased composite film solutions were also prepared following the same procedure, with the addition of LEO (0.5 g) or SCSEO (0.5 g). To obtain a homogeneous solution, they were first mixed (500 rpm, 60 min., 60°C) in Four E's Scientific MI0102003 Hot Plate Magnetic Agitator Stirrer (Four E's Scientific, Guangzhou, China). Then, they were stirred for 10 min. with Bandelin Sonopuls HD 2070 (20 kHz) model

TABLE 1: Film compositions.

Film no.	Starch (g)	Sodium alginate (g)	Water (ml)	Glycerol (g)	Sorbitol (g)	LEO/ SCSEO (g)
1	3	0.5	100	1	0	0.5
2	3	0.5	100	0	1	0.5
3	3	0.5	100	0.5	0.5	0.5
4	3	0.5	100	0.35	0.65	0.5
5	3	0.5	100	0.65	0.35	0.5

ultrasonic homogenizer (Bandelin electronic GmbH & Co. KG, Berlin, Germany). Finally, they were put in an Isolab ultrasonic water bath (Isolab Laborgerate GmbH, Eschau, Germany) for 10 min. at 50°C, till they were fully solubilized. The film solutions were poured in 10 cm diameter Petri dishes and dried at 40°C in an oven for 24 h. Dry films were stored in a constant temperature and humidity desiccator set at 50% RH and room temperature (22 + 0.5°C) until analyzed, after cooling [19]. To achieve different RH levels, water (RH = 100%) and silica gel (RH = 0%) were utilized.

#### 2.3. Characterization of Films

*2.3.1. Film Thickness.* The thickness values of the films (with 0.01 mm. sensitivity) were measured from five different points using a digital micrometer, and the mean value was recorded [20, 21].

2.3.2. Moisture Content. Edible film moisture content was calculated according to the ASTM D4442-20 standard method [22].  $3 \times 3$  cm cutout edible films were dried at 105  $\pm 2^{\circ}$ C for 24 h.  $M_0$  and  $M_1$  values of edible films, before and after drying, respectively, were measured until constant weight was achieved. Experiments were realized three times [23, 24].

Moisture (%) = 
$$\frac{(M_0 - M_1)}{M_0} \times 100.$$
 (1)

 $M_0$  is the initial weight, and  $M_1$  is the dry weight of the films.

2.3.3. Water Solubility. Water solubility was determined by the method of Dash et al. [25] and Go and Song [26] with some modifications. Edible films were cut to  $2 \times 2$  cm, dried at  $105 \pm 2^{\circ}$ C for 24 h, weighed, and stirred at 100 rpm at room temperature ( $22 + 0.5^{\circ}$ C) for 6 h at 10 ml distilled water. The remaining part of the films was filtered and dried in an oven at  $105 \pm 2^{\circ}$ C until a constant weight was obtained. Experiments were realized three times. Calculations were made using

Water solubility (%) = 
$$\frac{W1 - W2}{W1} \times 100.$$
 (2)

 $W_1$  is the dry weight, and  $W_2$  is the dry weight of the insoluble fraction of the film.

2.3.4. Swelling Index. The swelling index of the edible films was defined by the method of Susmitha et al. [9]. Edible films were cut into small pieces (2×2 cm), dried at  $105 \pm 2^{\circ}$ C for 24 h, and weighed ( $W_0$ ). They were immersed in 15 ml distilled water for 1 minute at room temperature (22 + 0.5°C). The swelled samples were wiped with filter paper and weighed ( $W_1$ ). Experiments were realized three times. The adsorbed water amount was calculated using

Swelling index (%) = 
$$\frac{W_1 - W_0}{W_0} \times 100$$
, (3)

where the swelling index (%) was the percentage of the swelling index and  $W_0$  and  $W_1$  were the weights of dried and wet samples.

2.3.5. Water Vapor Transmission Rate (WVTR). WVTR of the film samples was determined by the method of Shafie et al. [27], and ASTM E96/E96-22a [28], with some changes. The glass test tubes were filled with 5 g of silica gel, and their mouth was closed with dried film, surrounded with paraffin, and weighed every 24 h for 5 days. All the experiments were carried out in triplicate. The WVTR was calculated according to

WVTR = 
$$\frac{\Delta W}{\Delta t \times A}$$
, (4)

 $\Delta W/\Delta t$  is the amount of water transferred (g) per unit time (s), and A is the exposed area (m<sup>2</sup>).

2.3.6. Fourier Transform Infrared Spectroscopy (FT-IR). The chemical structure analysis of products was done using an FT-IR spectrometer equipped with a universal attenuation total reflectance sampling accessory with a spectral range between 4000 and 400 cm<sup>-1</sup> with a resolution of  $4 \text{ cm}^{-1}$  and with 16 scans per spectrum. The films were cut to 10 × 10 mm and placed onto the ATR platform.

2.3.7. Statistical Analysis. Statistical analysis by the analysis of variance (ANOVA) was done by SPSS®16. All tests were carried out in three independent runs, and the obtained parameters were averaged and expressed as the mean standard error (±) where each value is considered as significant at p < 0.05.

#### 3. Results and Discussion

#### 3.1. Characterization of Films

3.1.1. Thickness. The thickness is one of the most important factors of the films affecting the mechanical and barrier properties and plays an important role in the film quality. It is preferred that the film thickness is less than 0.3 mm so that the packaged food can be eaten together with the edible film and effective food protection can be provided. Lower edible film thickness also increases the solubility in the mouth and the digestibility [29, 30]. The difference in the thickness of edible films is due to the amylose content of the starch type. The amylose content

of wheat starch is 20.90%, corn starch is 25%, tapioca starch is 27%, potato starch is 26.90% and rice starch is 28.58%. It was observed that the thickness of the edible films increased with increasing amylose content. The high amylose content will increase the interaction between amylose molecules to compose stronger hydrogen bonds. So, the thickness of the film matrix increases [2, 31]. The changes in the thickness of the films may be due to the interaction and physical bonding between the film and the essential oil droplets which acted as a film filler [32, 33]. Moreover, the different chemical constituents of LEO and SCSEO may increase the distances between the particles in the matrix, thereby resulting in relatively thicker films [34, 35]. The corn, tapioca, and wheat starch edible film thicknesses were increased, and the potato and rice starch thicknesses were decreased after SCSEO and LEO addition to the film-forming solution. The decrease in film thickness of potato and rice starch-based edible films may be due to the inability of LEO and SCSEO to disperse in the film matrix of potato and rice starch. In this study, all the produced film thicknesses were less than 0.3 mm. The thickness values of the edible control and composite films are given in Tables 2-4. The lowest thickness (0.010 mm) value was observed in WS-1. Numerous studies have shown that essential oil incorporation can increase the film thickness [36-38].

3.1.2. Moisture Content. Moisture content value represents the total void volume occupied by water molecules in the microstructure network of the edible film and displays the possible effect of hydrocolloid interaction on the affinity of films to water [25, 30]. The moisture content of the WS-3 (wheat control film plasticized with glycerol and sorbitol) was the highest (15.50%). The addition of essential oils (SCSEO and LEO) to the films decreases moisture content, due to their hydrophobic nature. WS-SCSEO-3 moisture content decreases to 8.50%, and WS-LEO-3 moisture content decreases to 10.0% [33]. Edible films plasticized with glycerol have higher hydrophilicity than films plasticized with sorbitol due to the higher moisture content of glycerol.

Glycerol is a hydrophilic plasticizer, and when added to the starch, it shows high water-retaining capability. Also, the addition of glycerol decreases the interactions among starch macromolecules, simplifying the adsorption of water from the surroundings [38].

The moisture values of the edible control and composite films are given in Tables 2–4. Films with high moisture content are more flexible and can be used in different areas of food [9]. CS-SCSEO-1 (17.80%) and RS-LEO-5 (17.70%) were high moisture content edible composite films [39]. Similar results were obtained by Yang et al. [40] who studied the properties of corn starch films incorporated with *Zanthoxylum bungeanum* essential oil.

*3.1.3. Water Solubility.* The water solubility is a critical edible film property for the food preservation especially in humid environments, and antioxidants and plasticizer molecules are highly effective in water solubility [29, 32].

Starch film	Thickness (mm)	Moisture (%)	Solubility (%)	Swelling (%)	WVTR (g $*$ cm <sup>2</sup> /s)
CS-1	$0.016 \pm 0.051^{a}$	$9.60 \pm 3.16^{a}$	$92.20 \pm 3.05^{b}$	$54.20 \pm 3.19^{b}$	$10.40 \times 10^{-8} \pm 1.53^{a}$
CS-2	$0.070\pm0.025^b$	$7.60\pm1.20^{\rm b}$	$92.60 \pm 3.18^{a}$	$57.80 \pm 1.23^{a}$	$10.60 \times 10^{-8} \pm 1.76^{\rm b}$
CS-3	$0.080\pm0.033^{\mathrm{b}}$	$10.10\pm1.38^{\rm b}$	$88.60 \pm 2.56^{a}$	$230.70 \pm 3.75^{a}$	$7.00 \times 10^{-8} \pm 0.69^{a}$
CS-4	$0.060 \pm 0.046^{a}$	$8.90 \pm 2.16^{a}$	$99.70 \pm 3.27^{b}$	$76.10\pm1.88^{\mathrm{b}}$	$10.40 \times 10^{-8} \pm 1.15^{\rm b}$
CS-5	$0.100 \pm 0.002^{b}$	$9.50\pm2.15^a$	$82.10 \pm 2.68^{a}$	$12.20\pm0.11^{\rm b}$	$11.10 \times 10^{-8} \pm 1.55^{b}$
PS-1	$0.240\pm0.058^b$	$10.70 \pm 2.92^{b}$	$61.30 \pm 2.16^{a}$	$158.70 \pm 2.64^{a}$	$10.60 \times 10^{-8} \pm 0.75^{a}$
PS-2	$0.200\pm0.017^{b}$	$7.50\pm1.65^{a}$	$78.60\pm2.35^{\mathrm{b}}$	$132.70\pm2.38^a$	$8.90 \times 10^{-8} \pm 0.11^{a}$
PS-3	$0.075 \pm 0.015^{\rm b}$	$9.30\pm2.93^{\mathrm{b}}$	$97.20 \pm 3.76^{a}$	$276.30\pm3.24^a$	$8.60 \times 10^{-8} \pm 0.89^{b}$
PS-4	$0.240\pm0.068^a$	$6.90 \pm 2.31^{b}$	$63.20 \pm 2.13^{b}$	$164.50 \pm 2.76^{b}$	$8.70 \times 10^{-8} \pm 1.98^{b}$
PS-5	$0.160\pm0.019^{\rm b}$	$7.40\pm1.24^{\rm a}$	$89.90 \pm 2.78^{a}$	$172.80 \pm 2.59^{b}$	$7.00 \times 10^{-8} \pm 1.04^{a}$
RS-1	$0.120\pm0.034^a$	$12.70 \pm 2.58^{a}$	$81.60\pm2.08^{\rm b}$	$145.20 \pm 2.28^{a}$	$5.70 \times 10^{-8} \pm 0.97^{a}$
RS-2	$0.140\pm0.041^a$	$2.30\pm1.04^b$	$82.40 \pm 2.15^{a}$	$132.90 \pm 2.13^{b}$	$5.20 \times 10^{-8} \pm 1.31^{b}$
RS-3	$0.110\pm0.028^b$	$12.60 \pm 2.36^{b}$	$73.10 \pm 1.28^{a}$	$121.90 \pm 1.65^{a}$	$3.60 \times 10^{-8} \pm 0.47^{a}$
RS-4	$0.120\pm0.032^a$	$7.10\pm1.38^{\rm b}$	$77.90 \pm 1.46^{b}$	$85.50 \pm 3.26^{b}$	$3.30 \times 10^{-8} \pm 0.04^{b}$
RS-5	$0.150\pm0.065^a$	$7.30 \pm 1.46^{\rm a}$	$88.00 \pm 1.78^{\mathrm{b}}$	$169.60 \pm 1.96^{b}$	$4.10 \times 10^{-8} \pm 0.08^{b}$
TS-1	$0.060 \pm 0.056^{\mathrm{b}}$	$3.40\pm2.55^{\mathrm{b}}$	$56.30 \pm 2.73^{a}$	$218.70 \pm 2.37^{a}$	$5.70 \times 10^{-8} \pm 0.09^{a}$
TS-2	$0.160 \pm 0.009^{a}$	$7.40\pm1.64^{\rm b}$	$45.10 \pm 1.23^{b}$	$176.70 \pm 3.04^{a}$	$7.50 \times 10^{-8} \pm 0.54^{a}$
TS-3	$0.020\pm0.045^a$	$11.60 \pm 3.89^{a}$	$78.80 \pm 1.36^{\mathrm{b}}$	$149.00 \pm 2.97^{b}$	$9.40 \times 10^{-8} \pm 1.23^{a}$
TS-4	$0.060 \pm 0.071^{b}$	$7.60 \pm 1.37^{a}$	$65.60 \pm 2.68^{a}$	$245.40 \pm 3.33^{b}$	$10.30 \times 10^{-8} \pm 0.49^{b}$
TS-5	$0.035 \pm 0.082^{b}$	$6.90 \pm 3.86^{b}$	$88.80\pm2.09^{b}$	$211.50 \pm 2.85^{a}$	$8.20 \times 10^{-8} \pm 1.05^{\rm b}$
WS-1	$0.010\pm0.003^a$	$11.10 \pm 2.53^{a}$	$26.20 \pm 1.55^{a}$	$210.90\pm2.14^{\rm a}$	$6.10 \times 10^{-8} \pm 0.35^{a}$
WS-2	$0.100\pm0.066^a$	$3.30\pm2.75^{b}$	$22.30\pm1.19^{\rm b}$	$269.20\pm2.96^b$	$6.60 \times 10^{-8} \pm 0.21^{a}$
WS-3	$0.040 \pm 0.059^{\rm b}$	$15.50\pm0.38^{b}$	$40.30\pm3.57^a$	$247.10\pm1.78^b$	$7.50 \times 10^{-8} \pm 0.64^{b}$
WS-4	$0.080 \pm 0.012^{\mathrm{b}}$	$7.80 \pm 0.29^{a}$	$53.40 \pm 3.45^{a}$	$289.00\pm3.07^a$	$5.90 \times 10^{-8} \pm 0.15^{b}$
WS-5	$0.010\pm0.095^a$	$12.70 \pm 2.65^{a}$	$33.70\pm3.18^b$	$271.10 \pm 2.52^{b}$	$3.80 \times 10^{-8} \pm 0.07^{a}$

TABLE 2: Physical properties of starch-based control films.

Values are the mean of three replicates  $\pm$  SD. Different letters in the same column indicate significant differences among film samples (p < 0.05).

TABLE 3: Physical properties of starch-based composite films with SCSEO.

Starch film	Thickness (mm)	Moisture (%)	Solubility (%)	Swelling (%)	WVTR (g $*$ cm <sup>2</sup> /s)
CS-SCSEO-1	$0.023 \pm 0.048^{a}$	$17.80 \pm 2.76^{a}$	$83.80 \pm 2.35^{b}$	$109.70 \pm 1.14^{b}$	$0.06 \times 10^{-8} \pm 0.56^{b}$
CS-SCSEO-4	$0.070 \pm 0.025^{\rm b}$	$14.80\pm1.92^{\rm b}$	$52.30 \pm 1.16^{\mathrm{b}}$	$117.50 \pm 1.65^{a}$	$0.07 \times 10^{-8} \pm 0.25^{a}$
PS-SCSEO-1	$0.026 \pm 0.069^{a}$	$5.30\pm2.35^{\rm a}$	$30.30\pm1.04^a$	$159.00 \pm 2.63^{a}$	$12.20 \times 10^{-8} \pm 1.76^{\rm b}$
PS-SCSEO-5	$0.060 \pm 0.056^{\mathrm{b}}$	$14.30\pm1.76^{\rm b}$	$50.00 \pm 1.48^{\rm a}$	$213.50\pm2.07^b$	$13.30 \times 10^{-8} \pm 1.55^{b}$
RS-SCSEO-1	$0.120\pm0.032^{b}$	$5.90 \pm 1.33^{a}$	$52.90\pm2.89^{b}$	$92.50 \pm 1.26^b$	$11.60 \times 10^{-8} \pm 1.53^{\rm a}$
RS-SCSEO-5	$0.100 \pm 0.027^{a}$	$2.80\pm1.06^{\rm a}$	$64.60 \pm 2.68^{a}$	$86.90 \pm 3.25^{a}$	$9.80 \times 10^{-8} \pm 0.11^{b}$
TS-SCSEO-1	$0.080 \pm 0.012^{a}$	$13.90\pm2.38^{\mathrm{b}}$	$50.30 \pm 3.13^{a}$	$388.80\pm3.07^b$	$10.50 \times 10^{-8} \pm 0.75^{a}$
TS-SCSEO-5	$0.095 \pm 0.083^{\rm b}$	$3.70 \pm 1.21^{a}$	$41.20\pm1.79^{b}$	$215.50 \pm 2.34^{a}$	$7.10 \times 10^{-8} \pm 1.04^{\rm b}$
WS-SCSEO-1	$0.014 \pm 0.041^{\rm b}$	$6.40 \pm 1.64^{a}$	$85.90\pm2.12^{\rm b}$	$329.00\pm1.88^b$	$9.90 \times 10^{-8} \pm 1.23^{a}$
WS-SCSEO-3	$0.090 \pm 0.074^{a}$	$8.50\pm1.15^{\rm b}$	$51.60 \pm 3.27^{a}$	$248.90 \pm 1.78^{b}$	$16.80 \times 10^{-8} \pm 0.51^{a}$

Values are the mean of three replicates  $\pm$  SD. Different letters in the same column indicate significant differences among film samples (p < 0.05).

The solubility of SCSEO- and LEO-added corn, potatoes, rice, and tapioca starch edible films decreased compared to the starch control films. Only wheat starch composite film solubility increased. Essential oils combined with the starch hydroxyl group decreased the solubility of the com-

posite films. The interaction between the essential oil components and the hydroxyl groups of the film decreased the water solubility of the film; therefore, a more waterresistant film was obtained. Marzlan et al. [41] studied starch-based edible film for chicken meat packaging and

Starch film	Thickness (mm)	Moisture (%)	Solubility (%)	Swelling (%)	WVTR ( $g * cm^2/s$ )
CS-LEO-1	$0.120\pm0.033^a$	$16.40 \pm 2.56^{b}$	$16.40\pm014^b$	$371.20 \pm 3.84^{a}$	$20.10 \times 10^{-8} \pm 2.36^{b}$
CS-LEO-4	$0.080\pm0.014^a$	$7.90 \pm 0.39^{a}$	$19.50\pm1.19^{\rm a}$	$424.40\pm2.85^b$	$11.30 \times 10^{-8} \pm 1.47^{a}$
PS-LEO-1	$0.075 \pm 0.025^{b}$	$9.30 \pm 2.95^{a}$	$56.70 \pm 2.73^{\rm b}$	$307.90 \pm 1.76^{b}$	$14.90 \times 10^{-8} \pm 2.56^{\rm b}$
PS-LEO-5	$0.070 \pm 0.036^{a}$	$11.60\pm3.68^{\rm b}$	$66.20 \pm 2.13^{a}$	$433.20 \pm 3.47^{a}$	$26.00 \times 10^{-8} \pm 2.68^{b}$
RS-LEO-1	$0.065 \pm 0.058^{\mathrm{b}}$	$11.20 \pm 2.53^{b}$	$39.20 \pm 3.09^{a}$	$250.50 \pm 3.23^{a}$	$6.70 \times 10^{-8} \pm 0.21^{a}$
RS-LEO-5	$0.064 \pm 0.067^{b}$	$17.70 \pm 2.76^{a}$	$35.20 \pm 3.18^{a}$	$218.80\pm2.38^b$	$7.80 \times 10^{-8} \pm 0.69^{b}$
TS-LEO-1	$0.170 \pm 0.009^{a}$	$13.50 \pm 2.18^{a}$	$48.40 \pm 3.57^{a}$	$265.70 \pm 2.95^{a}$	$15.40 \times 10^{-8} \pm 1.65^{\rm b}$
TS-LEO-5	$0.100\pm0.027^{b}$	$8.70\pm2.16^{\rm b}$	$31.00\pm1.14^{\rm b}$	$236.30\pm3.75^a$	$9.20 \times 10^{-8} \pm 1.58^{a}$
WS-LEO-1	$0.070\pm0.038^a$	$10.70\pm2.92^{\rm b}$	$77.40 \pm 2.35^{b}$	$336.40\pm1.98^{b}$	$13.40 \times 10^{-8} \pm 1.77^{\rm b}$
WS-LEO-3	$0.130 \pm 0.041^{b}$	$10.00\pm1.38^{b}$	$51.60 \pm 3.29^{a}$	$266.20\pm2.92^b$	$17.90 \times 10^{-8} \pm 2.18^{b}$

TABLE 4: Physical properties of starch-based composite films with LEO.

Values are the mean of three replicates  $\pm$  SD. Different letters in the same column indicate significant differences among film samples (p < 0.05).

obtained similar results. The lowest solubility values were obtained from wheat starch control (22%) and SCSEO-added corn starch composite (16.40%) films. The water solubility values of the edible control and composite films are given in Tables 2–4.

3.1.4. Swelling Index. The physical and barrier properties and water resistance of the edible film are influenced by the swelling index. A very important feature of the films is that the food product coated with these materials meets water. The water resistance of edible starch films, measured in terms of swelling capacity, depends on the character and chemical composition of the starch used in their production [42]. The swelling index characterizes the water resistance of packaging materials from hydrophilic polymers and demonstrates the conservation of quality during packaging and storage of food products. The swelling index of starch films is a very important property of films, when food products coated with these materials are in contact with water. The swelling index values of corn, potatoes, rice, tapioca, and wheat starch-based control and composite films are given in Tables 2–4. The lowest swelling index values were found for corn starch-based edible films (12.20-230.70%), and higher values were found for wheat starch-based edible films (210.90-289.0%). The swelling index of SCSEO- and LEOadded corn, potatoes, rice, and tapioca starch edible films was increased compared to the starch control films. Only SCSEO-added rice starch composite film solubility was decreased. These results were consistent with the studies of Girgin et al. [39]. The highest swelling index values for SCSEO-added films were obtained from TS-SCSEO-1 (388.80%) and for LEO-added films were obtained from PS-LEO-5 (433.20%). The lowest swelling index values for SCSEO-added films were obtained from TS-SCSEO-1 (388.80%) and for LEO-added films were obtained from PS-LEO-5 (433.20%).

3.1.5. Water Vapor Transmission Rate (WVTR). WVTR plays an important role in food coating. Starch films help diminish moisture transfer among food and the surrounding atmosphere or between two components of heterogeneous

food products. The lower the WVTR, the better the packaging film [43, 44]. The WVTR of corn, potatoes, rice, tapioca, and wheat starch-based control and composite films are given in Tables 2-4. Among wheat, corn, tapioca, potato, and rice starches, rice starch has the highest amylose content (28.58%) and the lowest WVTR. High amylose content could influence the WVTR of the film. Rice starch-based edible films showed lower WVP indicating that the films exhibited high water resistance [1]. The WVTR of SCSEO-added potatoes, rice, tapioca, and wheat starchbased composite films  $(6 \times 10^{-8} - 16.80 \times 10^{-8} \text{ g} * \text{ cm}^2/\text{s})$ and LEO-added corn, potatoes, rice, tapioca, and wheat starch-based composite films  $(6.70 \times 10^{-8} - 26.00 \times 10^{-8} \text{ g} *$ cm<sup>2</sup>/s) were higher than the control films. Only WVTR of SCSEO-added corn starch-based composite films was lower than the control films  $(6 \times 10^{-8} - 7 \times 10^{-8} \text{ g} * \text{ cm}^2/\text{s})$ . The increase in WVTR of the films may be due to the negative impact of SCSEO and LEO on the microstructure of films. These results are consistent with the studies of Wigati et al. [45]

3.1.6. Fourier Transform Infrared Spectroscopy (FT-IR). Figure 1 shows FTIR spectra of corn, potatoes, rice, tapioca, and wheat starch-based control and composite films plasticized with 1% glycerol. When the FTIR spectrums were examined, it was seen that all spectrums had remarkable peaks at around 3265 cm<sup>-1</sup> (O-H groups of starches, SCSEO, and LEO), 2927 cm<sup>-1</sup> (stretching vibration of C-H of alkyl groups, SCSEO, and LEO), 1740 cm<sup>-1</sup> (C=O absorption for the LEO major compound citral), 1605 cm<sup>-1</sup> (H-O-H bending vibration from water), 1409 cm<sup>-1</sup> (C-O-H bending vibration), and 997 cm<sup>-1</sup> (C-OH and C-O-C bonds and C-C skeleton stretching vibrations). Since glycerol contains more hydroxyl groups than starch, the hydroxyl groups are more in starch-based films plasticized with glycerol. Moreover, this band of starch films, the broadband at around  $3265 \text{ cm}^{-1}$ , corresponded to the stretching of hydroxyl groups [7] and moved to a lower wavenumber than that of the corresponding starches. This showed that hydrogen bonds were formed between starch and glycerol [46, 47].



FIGURE 1: FTIR spectra of corn, potatoes, rice, tapioca, wheat starch control films, and glycerol-added composite films.

The FTIR spectra of edible composite films showed that no new peaks were formed, proving that there is no chemical interaction between the edible film component and that they kept their structure.

## 4. Conclusion

This study investigated and compared the film properties of corn, potato, rice, tapioca, wheat starch-based edible film-

added SCSEO or LEO, and plasticized with glycerol, and/or sorbitol produced using the casting method. When the FTIR spectrum was examined, it was seen that no new peaks were formed, indicating that there was no chemical interaction between the edible film components and that the starches, essential oils, and plasticizers were successfully combined. It was seen that wheat starch-based (having the lowest amylose content (20.90%)) control and composite films have the lowest thickness (0.010 mm) value. Wheat starch-based control film (15.50%), SCSEO-added corn starch (17.80%), and LEO-added rice starch-based (17.70%) composite films have high moisture content. Edible films plasticized with glycerol have higher hydrophilicity than films plasticized with sorbitol due to the higher moisture content of glycerol. The interaction between the essential oil components and the hydroxyl groups of the film decreased the water solubility of the film; therefore, a more water-resistant film was obtained. The lowest solubility values were obtained from wheat starch-based control (22%) and SCSEO-added corn starch-based composite (16.40%) films. The highest swelling index values were obtained from wheat starch-based control film (289.0%), SCSEO-added tapioca starch-based composite films (388.80%), and LEO-added potato starch-based composite films. Rice starch has the highest amylose content (28.58%), and rice starch-based edible control films have the lowest water vapor transmission rate value among the five types of edible film. As a result of this study, it was concluded that

- (i) Starch, essential oil, and plasticizer type influence edible film properties
- (ii) The film-forming properties of corn, rice, and wheat starch are better than potato and tapioca starch
- (iii) SCSEO can be successfully used in edible film production

## Data Availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.

## **Conflicts of Interest**

The authors declared that there is no conflict of interest.

## **Authors' Contributions**

Merve Basut Kazak conducted the data curation, writing of the original draft preparation, visualization, investigation, software, and validation. Nurcan Tugrul conducted the conceptualization, methodology, software supervision, writing, reviewing, and editing.

#### Acknowledgments

This research was performed at Yildiz Technical University Chemical Engineering Department Laboratories.

#### References

- P. B. Wilfer, G. Giridaran, J. J. Jeevahan, G. B. Joseph, G. S. Kumar, and N. J. Thykattuserry, "Effect of starch type on the film properties of native starch based edible films," *Materials Today: Proceedings*, vol. 44, no. 5, pp. 3903–3907, 2021.
- [2] T. R. Putri, A. Adhitasari, V. Paramita, M. E. Yulianto, and H. D. Ariyanto, "Effect of different starch on the characteristics of edible film as functional packaging in fresh meat or meat products: a review," *Materials Today: Proceedings*, vol. 87, no. 2, pp. 192–199, 2023.
- [3] S. Mao, F. Li, X. Zhou, C. Lu, and T. Zhang, "Characterization and sustained release study of starch-based films loaded with carvacrol: a promising UV-shielding and bioactive nanocomposite film," *LWT*, vol. 180, article 114719, 2023.
- [4] M. A. R. Sanches, C. Camelo-Silva, L. Tussolini, M. Tussolini, R. C. Zambiazi, and P. B. Pertuzatti, "Development, characterization and optimization of biopolymers films based on starch and flour from jabuticaba (*Myrciaria cauliflora*) peel," *Food Chemistry*, vol. 343, article 128430, 2021.
- [5] W. G. Sganzerla, G. B. Rosa, A. L. A. Ferreira et al., "Bioactive food packaging based on starch, citric pectin and functionalized with Acca sellowiana waste by-product: characterization and application in the postharvest conservation of apple," *International Journal of Biological Macromolecules*, vol. 147, pp. 295–303, 2020.
- [6] X. Song, G. Zuo, and F. Chen, "Effect of essential oil and surfactant on the physical and antimicrobial properties of corn and wheat starch films," *International Journal of Biological Macromolecules*, vol. 107: A, pp. 1302–1309, 2018.
- [7] L. Dai, J. Zhang, and F. Cheng, "Effects of starches from different botanical sources and modification methods on physicochemical properties of starch-based edible films," *International Journal of Biological Macromolecules*, vol. 132, pp. 897–905, 2019.
- [8] O. F. Vilpoux and J. F. S. S. Junior, "Chapter 3- Global production and use of starch," in *Starchy Crops Morphology, Extraction, Properties and Applications*, pp. 43–66, Academic Press, 2023.
- [9] A. Susmitha, K. Sasikumar, D. Rajan, M. A. Padmakumar, and K. M. Nampoothiri, "Development and characterization of corn starch-gelatin based edible films incorporated with mango and pineapple for active packaging," *Food Bioscience*, vol. 41, article 100977, 2021.
- [10] L. Cao, W. Liu, and L. Wang, "Developing a green and edible film from Cassia gum: the effects of glycerol and sorbitol," *Journal of Cleaner Production*, vol. 175, pp. 276–282, 2018.
- [11] N. Nordin, S. H. Othman, S. A. Rashid, and R. K. Basha, "Effects of glycerol and thymol on physical, mechanical, and thermal properties of corn starch films," *Food Hydrocolloids*, vol. 106, article 105884, 2020.
- [12] M. Jouki, N. Khazaei, and A. Jouki, "Fabrication and characterization of an active biodegradable edible packaging film based on sesame seed gum (*Sesamum indicum* L.)," *Journal* of Food Measurement and Characterization, vol. 15, no. 5, pp. 4748–4757, 2021.
- [13] M. G. De Paola, D. Mammolenti, F. R. Lupi, M. P. De Santo, D. Gabriele, and V. Calabrò, "Formulation and process investigation of glycerol/starch suspensions for edible films production by tape casting," *Chemical Papers*, vol. 76, no. 3, pp. 1525–1538, 2022.

- [14] F. Xue, Y. Gu, Y. Wang, C. Li, and B. Adhikari, "Encapsulation of essential oil in emulsion based edible films prepared by soy protein isolate-gum acacia conjugates," *Food Hydrocolloids*, vol. 96, pp. 178–189, 2019.
- [15] H. Çakmak, Y. Ozselek, O. Y. Turan, E. Fıratlığil, and F. Karbancioglu-Güler, "Whey protein isolate edible films incorporated with essential oils: antimicrobial activity and barrier properties," *Polymer Degradation and Stability*, vol. 179, article 109285, 2020.
- [16] R. Jafari, M. Zandi, and A. Ganjloo, "Characterization of alginate-gelatin edible film containing anise (Pimpinella anisum L.) essential oil," *Journal of Polymers and the Environment*, vol. 31, no. 4, pp. 1568–1583, 2023.
- [17] M. Zhang and H. Chen, "Development and characterization of starch-sodium alginate-montmorillonite biodegradable antibacterial films," *International Journal of Biological Macromolecules*, vol. 233, article 123462, 2023.
- [18] I. Choi, D. Shin, J. S. Lyu et al., "Physicochemical properties and solubility of sweet potato starch-based edible films," *Food Packaging and Shelf Life*, vol. 33, article 100867, 2022.
- [19] E. Basiak, A. Lenart, and F. Debeaufort, "Effect of starch type on the physico-chemical properties of edible films," *International Journal of Biological Macromolecules*, vol. 98, pp. 348– 356, 2017.
- [20] Y. Rahmasari and G. P. Yemis, "Characterization of ginger starch-based edible films incorporated with coconut shell liquid smoke by ultrasound treatment and application for ground beef," *Meat Science*, vol. 188, article 108799, 2022.
- [21] R. Thakur, P. Pristijono, J. B. Golding et al., "Development and application of rice starch based edible coating to improve the postharvest storage potential and quality of plum fruit (*Prunus salicina*)," *Scientia Horticulturae*, vol. 237, pp. 59–66, 2018.
- [22] ASTM, ASTM D 4442-2020: Standard Test Methods for Direct Moisture Content Measurement of Wood and Wood-Base Materials, ASTM, West Conshohocken, 2020.
- [23] S. Paudel, S. Regmi, and S. Janaswamy, "Effect of glycerol and sorbitol on cellulose-based biodegradable films," *Food Packaging and Shelf Life*, vol. 37, article 101090, 2023.
- [24] E. Asiamah, W. Arthur, V. Kyei-Barfour, F. Sarpong, and H. K. Ketemepi, "Enhancing the functional and physicochemical properties of tomato (*Solanum lycopersicum* L.) fruit through polysaccharides edible dipping technique coating under various storage conditions," *Bioactive Carbohydrates and Dietary Fibre*, vol. 30, article 100373, 2023.
- [25] K. K. Dash, N. A. Ali, D. Das, and D. Mohanta, "Thorough evaluation of sweet potato starch and lemon-waste pectin based-edible films with nano-titania inclusions for food packaging applications," *International Journal of Biological Macromolecules*, vol. 139, pp. 449–458, 2019.
- [26] E.-J. Go and K. B. Song, "Effect of java citronella essential oil addition on the physicochemical properties of Gelidium corneum-chitosan composite films," *Food Science and Biotechnology*, vol. 29, no. 7, pp. 909–915, 2020.
- [27] M. H. Shafie, R. Yusof, D. Samsudin, and C.-Y. Gan, "Averrhoa bilimbi pectin-based edible films: effects of the linearity and branching of the pectin on the physicochemical, mechanical, and barrier properties of the films," *International Journal of Biological Macromolecules*, vol. 63, no. 15, pp. 1276–1282, 2020.
- [28] ASTM, E96/E96-22a Standard Test Methods for Gravimetric Determination of Water Vapor Transmission Rate of Materials, ASTM International, West Conshohocken, PA, USA, 2022.

- [29] B. Karkar, İ. Patır, S. Eyüboğlu, and S. Şahin, "Development of an edible active chitosan film loaded with *Nigella sativa* L. extract to extend the shelf life of grapes," *Biocatalysis and Agricultural Biotechnology*, vol. 50, article 102708, 2023.
- [30] D. Khodaei, K. Oltrogge, and Z. Hamidi-Esfahani, "Preparation and characterization of blended edible films manufactured using gelatin, tragacanth gum and Persian gum," *LWT*, vol. 117, article 108617, 2020.
- [31] L. Boetje, X. Lan, J. V. Dijken et al., "Starch ester film properties: the role of the casting temperature and starch its molecular weight and amylose content," *Carbohydrate Polymers*, vol. 316, article 121043, 2023.
- [32] Y. Zhou, X. Wu, J. Chen, and J. He, "Effects of cinnamon essential oil on the physical, mechanical, structural and thermal properties of cassava starch-based edible films," *International Journal of Biological Macromolecules*, vol. 184, pp. 574–583, 2021.
- [33] Y. Wang, X. Wang, G. Hu et al., "Effect of green coffee oil as a natural active emulsifying agent on the properties of corn starch-based films," *LWT*, vol. 170, article 114087, 2022.
- [34] Y. Jiang, W. Lan, D. E. Sameen et al., "Preparation and characterization of grass carp collagen-chitosan-lemon essential oil composite films for application as food packaging," *International Journal of Biological Macromolecules*, vol. 160, pp. 340–351, 2020.
- [35] S. Bhatia, A. Al-Harrasi, Y. A. Shah et al., "Structural, mechanical, barrier and antioxidant properties of pectin and xanthan gum edible films loaded with grapefruit essential oil," *Heliyon*, vol. 10, no. 3, article e25501, 2024.
- [36] S. Salimiraad, S. Safaeian, A. A. Basti, A. Khanjari, and R. M. Nadoushan, "Characterization of novel probiotic nanocomposite films based on nano chitosan/nano cellulose/gelatin for the preservation of fresh chicken fillets," *LWT*, vol. 162, article 113429, 2022.
- [37] C. Qin, Z. Li, J. Zhang, H. Meng, and C. Zhu, "Preparation, physicochemical properties, antioxidant, and antibacterial activities of quaternized hawthorn pectin films incorporated with thyme essential oil," *Food Packaging and Shelf Life*, vol. 41, article 101235, 2024.
- [38] L. D. Pérez-Vergaraa, M. T. Cifuentesa, A. P. Francoa, C. E. Pérez-Cerveraa, and R. D. Andrade-Pizarrob, "Development and characterization of edible films based on native cassava starch, beeswax, and propolis," *NFS Journal*, vol. 21, pp. 39– 49, 2020.
- [39] B. Girgin and N. Tugrul, "Development of dry walnut shell edible film plasticized with glycerol and/or sorbitol and incorporated with aloe vera gel and lemon essential oil," *Periodica Polytechnica Chemical Engineering*, vol. 67, no. 2, pp. 193– 204, 2023.
- [40] Q. Yang, F. Zheng, Q. Chai et al., "Effect of emulsifiers on the properties of corn starch films incorporated with *Zanthoxylum bungeanum* essential oil," *International Journal of Biological Macromolecules*, vol. 256, no. 1, article 128382, 2024.
- [41] A. A. Marzlan, B. J. Muhialdin, N. H. Z. Abedin et al., "Incorporating torch ginger (*Etlingera elatior* Jack) inflorescence essential oil onto starch-based edible film towards sustainable active packaging for chicken meat," *Industrial Crops and Products*, vol. 184, article 115058, 2022.
- [42] P. Pająk, I. Przetaczek-Rożnowska, and L. Juszczak, "Development and physicochemical, thermal and mechanical properties of edible films based on pumpkin, lentil and quinoa

starches," International Journal of Biological Macromolecules, vol. 138, pp. 441–449, 2019.

- [43] C. Dularia, A. Sinhmar, R. Thory, A. K. Pathera, and V. Nain, "Development of starch nanoparticles based composite films from nonconventional source-water chestnut (*Trapa bispinosa*)," *International Journal of Biological Macromolecules*, vol. 136, pp. 1161–1168, 2019.
- [44] K. Roy, R. Thory, A. Sinhmar, A. K. Pathera, and V. Nain, "Development and characterization of nano starch-based composite films from mung bean (*Vigna radiata*)," *International Journal of Biological Macromolecules*, vol. 144, pp. 242– 251, 2020.
- [45] L. P. Wigati, A. A. Wardana, F. Tanaka, and F. Tanaka, "Edible film of native jicama starch, agarwood aetoxylon bouya essential oil and calcium propionate: processing, mechanical, thermal properties and structure," *International Journal of Biological Macromolecules*, vol. 209: A, pp. 597–607, 2022.
- [46] M. S. A. Jorge, C. M. Gaspar, H. F. M. Henriques, and E. M. M. Braga, "Edible films produced from agrifood by-products and wastes," *Innovative Food Science & Emerging Technologies*, vol. 88, article 103442, 2023.
- [47] E. P. Da Cruz, J. B. Pires, F. N. dos Santos et al., "Encapsulation of lemongrass essential oil into cassava starch fibers for application as antifungal agents in bread," *Food Hydrocolloids*, vol. 145, article 109105, 2023.