Biodegradable edible film based on basil seed gum: the effect of gum and plasticizer concentrations

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Research Article

Keywords: Basil seed gum, plasticizer, biodegradable, edible film, packaging

Posted Date: February 28th, 2023

DOI: https://doi.org/10.21203/rs.3.rs-2626369/v1

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Abstract

In this research, edible films produced from basil seed gum (BSG) with 3 different gum (0.5%, 1%, 1.5%) and plasticizer concentrations (1%, 3%, 5%) were developed, and the physical, thermal, barrier and microstructural properties of these films were measured. As a result of XRD, AFM, DSC, and FT-IR spectroscopy analyses, it was concluded that the mechanical and barrier properties and thermal stability of BSG-based films are quite good. The increase in gum and glycerol concentrations increased the crystallinity also strengthened the mechanical and barrier properties of the film. Also, films with low gum and high glycerol ratio have almost smooth surfaces and appropriate transparency for packaging applications. As the glycerol and BSG concentration increased, WVP values of the films increased. The complete dissolution of this film in the soil within 60 days, even at the highest gum concentration, showed that this material could be considered eco-friendly packaging. For this reason, it is thought that BSG-based films and coatings with suitable gum and plasticizer concentrations can be a potential packaging material for foods since they can be obtained at low cost, have a very good barrier, thermal and structural properties, and are edible and biodegradable.

1. Introduction

The most widely used polymeric-based materials for food packaging are derived from synthetic and non-biodegradable sources, which can cause global challenges and serious environmental concerns [1]. To solve this problem, many studies have been carried out for the production of environmentally friendly materials, and recently, the interest in natural polymers has increased [2]. Edible coatings and films made from renewable and natural resources have the qualities to replace synthetic plastic packaging. Edible films are thin layers of biopolymers that have potential as food packaging without altering the original ingredients or processing methods of the final food product. The advantages of biopolymer-based films over synthetic polymers include availability, edibility, biodegradability, affordability, good barrier properties to gases, and is a good carrier matrix, especially for natural antimicrobial, antioxidant, flavor, and other bioactive ingredients [3–5].

Protein, polysaccharides, and lipid-based hydrocolloids of animal or plant origin can be used to develop edible films and coatings. Polysaccharides and proteins are excellent materials for film formation as they show excellent mechanical and structural properties, but have a poor barrier capacity to moisture transfer. This problem is not found in lipids, especially those with high melting points, due to their hydrophobic properties [6]. Plasticizers are also often added to film-forming solutions to improve the properties of the final film. Common food-grade plasticizers such as sorbitol, glycerol, mannitol, sucrose, and polyethylene glycol reduce brittleness and increase film flexibility, which is important in packaging applications [7].

In recent years, new hydrocolloid sources to be used in the production of edible and biodegradable films have been researched to meet the demands of the food industry. In this context, research on the extraction of gum, its properties, and film-coating production has recently started on Basil seed gum (BSG). Due to its high mucilage content (20%), basil seeds can be a natural source of hydrocolloids that can be an alternative to commercial gums with invaluable functional properties [8, 9]. Although BSG has a high coefficient of consistency and shear stress dependent on temperature and concentration. In addition to being a stabilizer and thickening agent, it has also been reported to be a surfactant and emulsifying hydrocolloid [10]. The surface activity of crude, purified and protein-free forms of BSG was investigated and it was concluded that BSG solutions at different concentrations reduced the surface tension. It has also been found that BSG has emulsifying properties and low protein impurities can stabilize oil/water emulsions [11, 12]. BSG is a material that attracts attention as a film agent and hydrocolloid in foods due
to many important properties such as exhibiting good functional properties compared to other polysaccharides, low cost, heat resistance, hydrophilic structure, biocompatibility, biodegradability, and excellent film-forming agent [13, 14].

In this study, the edible film was produced with gum obtained from basil seeds. For the optimization of the coating solutions, 3 gum ratios (0.5%, 1% and 1.5%) and 3 plasticizer (glycerol) ratios (1%, 3% and 5%) were applied. Although some studies have been done on BSG-based films, there is no research on the optimization of the gum and plasticizer ratios of the film. It is thought that optimizing the film formulation in terms of physical, thermal, barrier, and microstructural properties will make a great contribution to making the film a good alternative to other packaging as a packaging material. In addition to investigating the applicability of the film to the food industry, it is thought that this research will be a reference for adjusting the concentrations of gum and plasticizer according to the characteristics of the food to be applied.

2. Material Methods

2.1 Material

The basil seeds used in the research were obtained from Arzuman Agro and Seed Company (Konya/Turkey). 95% pure glycerol used as a plasticizer in film production was obtained from Merck (Darmstadt, Germany).

2.2 Methods

2.2.1 Gum extraction from basil seeds

Basil seeds are washed 3–4 times with ethanol to purify them from impurities, then excess ethanol is removed with the help of an oven. The pH of the distilled water was adjusted to 8 with HCl and H\textsubscript{2}SO\textsubscript{4} and heated to 50–55 °C. Then basil seeds were added to water at a ratio of 30:1. The prepared mixture was then kept at 50 °C for 2 h in a magnetic stirrer with gentle stirring. To separate the mucilage from the seed surface, the mixture was stirred at 1500 rpm for 10 min. Then, it was centrifuged at 9000 rpm for 15 min (Nüve-NF 800R, Turkey) to separate the mixture in the pericarp layer of the seed. Basil seed mucilage separated from the upper phase was left to dry in an oven set at 50 °C for 24 h.

The impurities in the gum directly affect the extraction efficiency. For this reason, after obtaining the pure extract, 1 volume of the extract was mixed with 3 volumes of 95% ethanol and kept at 4 °C overnight. The mixture was then filtered using a sieve. Pure water was added to the basil seed gum (BSG) extract remaining on the sieve surface and mixed for 30 minutes. Then the obtained BSG was dried at 50 °C. After drying BSG was ground and transferred to zip lock bags. It was stored at +4 °C until used in the analysis. The extraction yield of gum from basil seed was calculated with the following formula [9]:

\[
\text{Extraction yield} = \left( \frac{\text{amount of extracted gum (g)}}{\text{Amount of basil seed (g)}} \right) \times 100
\]

The extraction yield of BSG used in the study was determined as 20.2%.

2.2.2 Preparation of film samples

In the study, a total of 9 different coating solutions were prepared by using 3 different gum ratios (1%, 1.5%, 2%) and 3 different plasticizer ratios (1%, 3%, 5%). In this context, the obtained BSG was dissolved in distilled water at the rates stated above, and glycerol was added. The pH of the mixture was then adjusted to 7. The prepared coating
solutions were heated with stirring at 50 °C for 2 h and then pasteurized for 1 h in a shaking water bath adjusted to 80–85 °C. It was centrifuged at 6000 rpm for 5 minutes (Nüve-NF 800R, Turkey) to remove air bubbles and impurities in the solutions. The prepared coating solutions were kept at +4 °C for 24 h and then transferred to 90 mm diameter Petri dishes. Petri dishes were kept in an oven at 50 °C for 24 h. At the end of the period, the lids of the Petri dishes were closed, placed in a desiccator containing saturated Mg(NO₃)₂ (53% relative humidity), and stored until the analysis.

### 2.2.3 Film moisture

The drying cups were kept at 105 °C for 1 hour and brought to constant weight. The film samples (2cm x 2cm square pieces) were weighed into the drying cups and dried in an oven at 105 °C until they reach a constant weight [15].

### 2.2.4 Film thickness

Film thickness was measured with a 0.001 mm precision digital micrometer (Loyka 5202-25, Germany). Measurements were repeated in 10 different regions of the film.

### 2.2.5 Water vapor permeability (WVP)

Glass containers are filled with anhydrous calcium chloride to provide 0% relative humidity (RH). Glass containers were sealed with paraffin with sample films to isolate their inner atmosphere from the outside. It was then placed in a desiccator containing saturated sodium chloride (75% RH). The inner atmosphere of the container with low RH absorbed moisture from the desiccator, resulting in an increase in the weight of anhydrous calcium chloride in the container. These weight gains were determined after 24 h of cultivation and the water vapor permeability values of the films were calculated with the formula given below [16].

\[
\text{Water vapor permeability} = \frac{G \times L}{A \times t \times \Delta P}
\]

G: increase in weight (g), L: film thickness (mm), cross-sectional area (mm²), t: permeation time, ΔP: pressure difference between container atmosphere and desiccator atmosphere

### 2.2.6 Water solubility

Film pieces (3x3 cm) whose initial weights were determined were dried in an oven (105 °C) until they reached constant weight. The dried films were dipped into tubes containing 20 ml of distilled water. Then, the tubes were kept in a shaking oven at 95 rpm, 25 °C for 24 h. The films insoluble in water were taken and dried in an oven at 105 °C for 24 h and weighed [17].

### 2.2.7 Color

Minolta (CR 400; Minolta, Japan) colorimeter was used for color analysis, and L, a, b values (L: represents the range of black and white, a: represents the color range of red-green, b: the range of yellow-blue) were recorded [18]. The values were measured from 5 different areas of each film and the average value was recorded.

### 2.2.8 Opacity / transparency

The opacity of the films was determined by measuring at 600 nm using a UV spectrophotometer (Optizen 3220/Korea). The films were cut in 4.5 cm x 1 cm dimensions and placed in spectrophotometer cuvettes, and then the absorbances were measured. Opacity was calculated with the following formula. According to this formula, the film with a high opacity value has low transparency [19].
2.2.9 X-ray diffraction (XRD)

XRD analysis was performed using X-ray diffractometry (Bruker D8 Advance/Germany), modifying the method developed by Motalebi Monghanjougi et al. [20]. The diffraction angle is set to 4°-80°. Current and voltage are set to 40 kV and 40 mA, respectively.

2.2.10 Atomic force microscopy (AFM)

This technique was used to examine the surface morphology of the films. First, the microscope (Ntegra/Solaris) is set to non-contact mode. The film samples were then fixed on the sample tray with double-sided tape. The force constant was set to 25–60 N/m, images of 5–50 µM sizes were taken from different regions of the films and surface roughness data were recorded [21].

2.2.11 Fourier transform infrared (FT-IR) spectroscopy analysis

Spectrophotometric measurements were made with a Bruker/Vertex70 (Germany) brand spectrophotometer. Measurements were made at a scanning range of 400–4000 cm⁻¹ and a resolution of 4 cm⁻¹ [22].

2.2.12 Thermal analysis

The thermal properties of the film samples were measured with the help of differential scanning calorimetry (Mettler-Toledo, Switzerland). The samples were scanned at a heating rate of 10 °C/min in the range of -80 to + 400 °C. Glass transition temperature ($T_G$) midpoint temperature of change in baseline due to change in heat capacity during glass transition) and melting point ($T_m$) were determined [23].

2.2.13 Biodegradability analysis

The analysis medium was prepared by pouring soil approximately 4 cm high into plastic containers. Then, the films (2x2 cm) were buried in the soil at a depth of 1 cm. The containers were kept at room conditions and humidity. Water was sprayed on it twice a day to preserve the moisture of the soil. On the 15th, 21st, 30th, 45th and 60th days, the soil on the films was removed and the changes in the films were recorded with the camera. [24].

2.2.14 Stability of films in acidic/alkaline conditions

The films can also be used to detect the deterioration of the product (smart packaging). For this purpose, the film with a diameter of 16 mm was placed in Petri dishes containing 10 ml of acidic (pH:3) and alkaline solution (pH:12). The color change in the films was observed, photographs were taken and the results were recorded [24].

2.2.15 Statistical Analysis

SPSS PASW 21.0 statistical analysis program was used for data analysis. Differences were determined at the p < 0.05 level. Means with significant differences were subjected to the Duncan multiple comparison test (MSTAT-C).

3. Result And Discussion

3.1 Moisture content
High moisture content in films ensures better adhesion of the film to aqueous food products, films with low moisture content are more suitable for fatty foods [25]. For this reason, it is very important to determine the moisture content of the films. The moisture content of the film samples produced according to different BSG and glycerol concentrations are given in Table 1. The differences between the samples were found to be statistically insignificant while the moisture content of the film samples decreased due to the increase in the glycerol ratio. Contrary to the results of this study, many previous studies reported that the moisture content of the films increased due to the increase in the glycerol ratio [18, 26, 27]. Due to the hygroscopic nature of glycerol, it has been suggested that increasing the glycerol ratio causes an increase in the amount of moisture retained in the films at the end of drying [28].
Table 1
The effect of different BSG and glycerol concentrations on the physical properties of the film samples

<table>
<thead>
<tr>
<th>BSG ratio (%)</th>
<th>Glycerol Ratio (%)</th>
<th>Moisture (%)</th>
<th>Film thickness (mm)</th>
<th>Water solubility (%)</th>
<th>Water Vapor Permeability $x10^{-10}$ (g/ Pa mm h)</th>
<th>L</th>
<th>a</th>
<th>b</th>
<th>Opacity ($A_{600}/\text{mm}$) %</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5</td>
<td>1</td>
<td>44.78 ± 0.572^a</td>
<td>0.064 ± 0.003^e</td>
<td>0.131 ± 0.000^de</td>
<td>1.72 ± 0.08^d</td>
<td>79.56 ± 0.71^a</td>
<td>1.01 ± 0.15^e</td>
<td>14.37 ± 0.46^e</td>
<td>9.31 ± 0.92^b</td>
</tr>
<tr>
<td>3</td>
<td>0.28^ab</td>
<td>38.81 ± 0.136</td>
<td>0.0136 ± 0.009^d</td>
<td>0.540 ± 0.12^bcd</td>
<td>3.58 ± 0.26^bcd</td>
<td>75.72 ± 0.47^bc</td>
<td>1.94 ± 0.14^cd</td>
<td>16.54 ± 0.32^d</td>
<td>4.18 ± 0.15^de</td>
</tr>
<tr>
<td>5</td>
<td>3.11^ab</td>
<td>28.68 ± 0.170</td>
<td>0.0109 ± 0.001^e</td>
<td>0.959 ± 0.07^abc</td>
<td>4.23 ± 0.19^bcd</td>
<td>76.72 ± 1.29^ab</td>
<td>1.58 ± 0.18^de</td>
<td>15.67 ± 0.90^de</td>
<td>2.83 ± 0.08^f</td>
</tr>
<tr>
<td>1</td>
<td>1</td>
<td>40.56 ± 0.12^ab</td>
<td>0.085 ± 0.005^e</td>
<td>0.109 ± 0.01^e</td>
<td>2.14 ± 0.07^d</td>
<td>73.15 ± 3.14^c</td>
<td>2.34 ± 0.68^c</td>
<td>19.80 ± 1.82^c</td>
<td>11.91 ± 0.46^a</td>
</tr>
<tr>
<td>3</td>
<td>7.35^ab</td>
<td>36.40 ± 0.170</td>
<td>0.0109 ± 0.001^e</td>
<td>0.959 ± 0.07^abc</td>
<td>5.99 ± 0.34^ab</td>
<td>67.80 ± 1.45^d</td>
<td>3.41 ± 0.37^b</td>
<td>21.18 ± 0.68^c</td>
<td>4.90 ± 0.18^cd</td>
</tr>
<tr>
<td>5</td>
<td>2.04^b</td>
<td>27.49 ± 0.230</td>
<td>0.0160 ± 0.009^ab</td>
<td>0.931 ± 0.09^abc</td>
<td>6.16 ± 1.89^ab</td>
<td>66.44 ± 1.40^de</td>
<td>3.63 ± 0.40^b</td>
<td>21.10 ± 0.86^c</td>
<td>3.48 ± 0.18^ef</td>
</tr>
<tr>
<td>1.5</td>
<td>0.57^ab</td>
<td>35.39 ± 0.121</td>
<td>0.0117 ± 0.000^de</td>
<td>0.117 ± 0.000^de</td>
<td>2.96 ± 1.00^cd</td>
<td>66.84 ± 1.45^de</td>
<td>3.90 ± 0.39^b</td>
<td>23.52 ± 0.66^b</td>
<td>12.34 ± 0.12^a</td>
</tr>
<tr>
<td>3</td>
<td>5.44^ab</td>
<td>31.70 ± 0.201</td>
<td>0.0160 ± 0.009^ab</td>
<td>0.466 ± 0.18^de</td>
<td>5.77 ± 0.08^abc</td>
<td>63.87 ± 2.44^ef</td>
<td>4.68 ± 0.41^a</td>
<td>25.09 ± 0.68^a</td>
<td>5.25 ± 0.17^c</td>
</tr>
<tr>
<td>5</td>
<td>6.03^ab</td>
<td>29.71 ± 0.329</td>
<td>0.042 ± 0.02^a</td>
<td>1.141 ± 0.02^a</td>
<td>7.29 ± 0.05^a</td>
<td>62.35 ± 2.17^f</td>
<td>4.60 ± 0.39^a</td>
<td>23.61 ± 0.39^b</td>
<td>4.19 ± 0.19^de</td>
</tr>
</tbody>
</table>

*The different lower-case letters show statistically the difference between the film samples ($p \leq 0.05$).

In addition, although the increase in the amount of BSG in the film samples slightly decreased the moisture content of the films. This decrease may be due to the strong electrostatic interaction between BSG molecules. This interaction may have caused a decrease in moisture content by weakening the hydrogen bond between BSG molecules and water molecules [19]. Similar to the results of this study, Zhang et al. [29] reported that the moisture content of edible films produced based on ghatti gum decreased due to the increase in gum content.

3.2 Film thickness
It has been determined that the thickness of the film samples varies in the range of 0.064–0.324 mm (Table 1). The increase in the glycerol ratio caused a significant increase in the film thickness ($p < 0.05$). The increase in film thickness can be attributed to the penetration of glycerol molecules into mucilage polymer chains and breaking molecular interactions, especially hydrogen bonds. Thus, the molecular volume of the film network increases and it absorbs more water vapor into its structure, causing an increase in film thickness [27]. Similarly, Chen et al. [30] reported that the film thickness increased as the glycerol content increased in the edible films produced from tapioca starch/decolored Hsian-Tsao leaf gum. Furthermore, at constant glycerol ratios in the film samples, the increase in BSG content significantly increased the film thickness ($p < 0.05$). In line with our results, Sandhu et al. [31] reported that films containing higher concentrations of starch and carrageenan have a higher thickness in composite edible films prepared from millet starch and carrageenan gum.

### 3.3 Solubility of films

Edible films are generally water-soluble. Solubility tests evaluate the water-resistance properties of the films and demonstrate hydrophobicity in practical terms. The water solubility ratios of the films are presented in Table 1. It was determined that the solubility of the film samples increased significantly due to the increase in glycerol concentration in all applied BSG ratios ($p < 0.05$). Similar results were reported by Jouki et al. [32] on film samples produced based on cress seed gum. The water solubility of polymers is affected by cross-linked network structures and crystallization between polymer chains. Polymer-glycerol bonds in glycerol-added films cause an increase in the hydrophilic property of the edible film, resulting in increased water solubility [27]. On the other hand, the water solubility of the films did not show a constant change depending on the increase in the amount of BSG. Similar behavior was observed in edible films produced from the Ghatti gum [29].

### 3.4 Water Vapor Permeability

The water vapor permeability (WVP) of the composite films represents the moisture exchange with the surrounding environment. A lower WVP means that the films form a better barrier against water vapor [33]. As seen in Table 1, the WVP values of the film samples vary in the range of $1.72–7.29 \times 10^{-10}$ g Pa$^{-1}$ mm$^{-1}$ h$^{-1}$. The increase in glycerol and BSG concentration significantly increased the WVP values of the films ($p < 0.05$). The increase in plasticizer concentration causes a reorganization of the polysaccharide network, resulting in an increase in free volume and segmental movements. This allows water molecules to disperse more easily and increases the WVP of hygroscopic or hydrophilic films [34]. In addition, at higher glycerol concentrations, glycerol can aggregate within itself to open the polymer structure and increase the moisture permeability of the film. Similar to the results of the current study, it has been reported in many previous studies that the WVP of hydrocolloid films increases with the concentration of added plasticizers [35, 36]. The WVP of edible films depends on their water solubility. Considering the water solubility values of the films (Table 1), it was determined that the increase in glycerol concentration significantly increased the water solubility of the films. This causes an increase in the porosity of the edible film and increases its water vapor permeability. Also, in our study, the increase in gum ratio caused an increase in the water vapor permeability of the films. This can be explained by the fact that BSG added at high concentrations causes gaps between the polymeric chains.

### 3.5 Color and Opacity

The color and transparency of edible films are two important criteria in terms of overall appearance and consumer acceptance. In general, the clearer the biopolymer film and the more similar it is to the plasticity of synthetic polymers, the higher its acceptability and application rate [37]. The color properties of BSG-based films were evaluated in terms of L, a, and b values, and the results are presented in Table 1. Depending on the increase in the
glycerol ratio, the brightness (L*) of the films decreased, while the redness (a*) value increased significantly (p < 0.05). When the glycerol ratio increased from 1–3%, it was observed that the yellowness (b*) values of the film samples increased but then decreased in all gum ratios. In line with our findings, Zhang et al. [29] reported that the brightness of edible films produced based on ghatti gum decreases with glycerol concentration. The increase in the amount of gum significantly decreased the L value, which expresses the brightness (p < 0.05). Therefore, the darkness of the BSG-based films varied directly with the gum content. On the other hand, with the increase in gum ratio, the redness (a*) and yellowness (b*) values of the films increased significantly (p < 0.05).

Examining the opacity and transparency of films is very important in food packaging. With high opacity or low transparency, possible photooxidation effects that lead to light-catalyzed food spoilage can be avoided [38]. Opacities of the film samples are reported in Table 1. Due to the increase in glycerol ratio, the opacity values of BSG-based films showed a significant decrease (p < 0.05). Although the opacity of the film samples increased depending on the increasing amount of BSG (Table 1), it was observed that all films had appropriate transparency for packaging applications. This can be explained by the increased BSG content, which leads to an increase in opacity for the edible film, and the increased intermolecular interactions between polymer chains and decreased mobility [27]. Saberi et al. [39] stated that starch had a significant linear positive effect on transparency in biodegradable edible films based on pea starch and guar gum, but glycerol and guar gum had a negative effect on transparency.

3.6 XRD

X-ray diffraction analysis was performed to characterize the crystal pattern of the films. As shown by the X-ray diffraction patterns in Fig. 1, the amorphous structure is dominant in BSG films. All films showed a specific peak at ~ 20°. If the film components have low compatibility, each component exhibits its crystal region [40]. However, in our study, the same peak appeared in the XRD spectra of all films containing different concentrations of gum and glycerol. Therefore, the components were compatible with each other. As the glycerol ratio and gum concentration in the film composition increased, the intensity of the diffraction peaks increased at 2θ. In X-ray diffraction patterns, the material with high crystallinity shows a narrow and high peak shape, while the material with low crystallinity shows a wider peak shape [41]. The increase in peak density is interpreted as the formation of new hydrogen bonds and the increase of regular areas, thus increasing the crystallinity of the films [19]. Therefore, the increase in gum and glycerol concentrations increased the crystallinity of the film. The increase in crystallinity also strengthens the mechanical and barrier properties of the film, increasing its stability [42].

3.7 AFM

The roughness of the film surfaces was studied to understand the surface morphology, integrity, and compatibility of the films. For this purpose, surface topographies and roughnesses were determined by observations made with atomic force microscopy (AFM). As seen in Fig. 2, as the glycerol ratio increases, peak-to-peak (Sy), ten-point height (Sz), and average roughness (Sa) values decrease in all films. Hasan et al. [43] stated that plasticizers can reduce intermolecular forces and increase film flexibility by expanding the molecular free space and weakening the hydrogen bonds of polymer chains. The decrease in surface roughness depending on the glycerol ratio can be explained by this expression. Based on the gum concentration, the above values increased as the gum concentration in the film samples increased.

The films with low glycerol and high gum ratio exhibited a surface topography with dense sharp spikes. The symmetry of the surface profiles is characterized by surface skewness (Ssk), and the distribution of spikes above and below the mean line is characterized by the coefficient of kurtosis (Ska) values. The skew profile is used to distinguish the porosity, and load capacity of the surfaces, and the negative sign of skewness is an optimum
criterion for good bearing surfaces [44]. As the roughness increased due to the increase in the gum concentration and the decrease in the glycerol ratio in the examined film samples, the skewness took a positive value. However, films with low gum and high glycerol ratio generally have slight positive distortion and coefficient of kurtosis values are quite good.

3.8 FT-IR Spectroscopy

FT-IR spectroscopic analysis was performed to understand the intermolecular interactions and structural changes in the films at the molecular level. The FT-IR spectra of BSG films from 3500 to 500 cm$^{-1}$ are shown in Fig. 3. The peak occurring at approximately 3290 cm$^{-1}$ reflects the stretching vibration of OH groups affected by intermolecular and intramolecular hydrogen bonds. This stretching vibration is due to the interaction of hydroxyls with bound water [45]. The peak occurring around 2930 cm$^{-1}$ represents the C-H aliphatic absorption peak and is related to the vibration of the methyl group in the polymer chain. The peak occurring between 1600 and 1640 cm$^{-1}$ is a typical indicator of the presence of water [46]. A peak around 1400 cm$^{-1}$ indicates the presence of uronic acid in the basil seed mucilage polysaccharide [47]. The peak observed at approximately 1034 cm$^{-1}$ for all film samples generally corresponds to the C-O-C asymmetric and symmetric stretching modes from the ether group in the pyranose rings and indicates the presence of monosaccharide. The adsorption peaks observed at 920 and 850 cm$^{-1}$ are the stretching vibration of C-C bonds [48]. The peak observed around 920 cm$^{-1}$ generally reflects the vibration of α-1, 4-glycosidic bonds [49].

The obtained FTIR spectra were compatible with many studies on BSG-based edible films [50, 51]. When the spectra were examined, it was observed that the O-H stretch bands became sharper and shifted to lower wave numbers with increasing glycerol concentration. The increase in glycerol concentration increases the number of hydroxyl bonds, resulting in a sharper O-H peak. On the other hand, glycerol associated with BSG molecules forms hydrogen bonds and causes the O-H stretching vibration to shift to lower wave numbers [52]. At low glycerol concentration, all the hydroxyl groups in the polymer interact with the hydroxyl groups in the glycerol molecules to replace the interaction between the polymer molecules due to the solvent effect of the plasticizer. With increasing glycerol concentration, free hydroxyl groups in glycerol become dominant, reducing the intermolecular force between BSG chains [22]. It was observed that the wavelength of the O-H stretching vibration peaks increased, while the wavelength of the C-O-C peaks decreased as the gum concentration increased. In addition, when the spectra were examined, it was seen that all BSG films had similar models and no new peaks were found between the films. This showed that BSG has good compatibility with glycerol.

3.9 Thermal Analysis

The thermal properties of films are physical property that plays an important role in the processing condition as well as in the application of the composite in food packaging. Determination of the glass transition temperature ($T_g$) value is one of the basic approaches used to determine the shelf life of food products. Molecules can feel more comfortable at low $T_g$ temperatures as the free space increases. However, the water vapor and gas permeability of polymer films increase as the $T_g$ temperature rises. At low and high values of $T_g$, the film samples are hard or rough and soft or flexible, respectively [32]. The films exhibit only one glass transition temperature, indicating a homogeneous mixture of compounds, particularly glycerol, in the film matrix [53]. A single $T_g$ value was obtained in all the films examined, and the glass transition temperature was below 0 (in the range of -41.33 °C to -45.53 °C) in all samples (Online Resource 1). The $T_g$ value of the film samples was found to be higher than the $T_g$ value of materials such as high-density polyethylene (-125 °C), gellan (-84 °C), and pure liquid glycerol (-93 °C) [27]. This effect is probably due to the good miscibility-compatibility of the glycerol and BSG phases, resulting in a higher
glass transition temperature due to the increase in average molecular weight. This shows that BSG has good potential in packaging applications.

Plasticizers change the interaction between polymers, leading to an increase in the free volume of the system and thus an increase in chain mobility and a decrease in the glass transition temperature of the system [54]. In the film samples in our study, the $T_g$ value increased when the plasticizer concentration increased from 1–3%, but this increase was not statistically significant. A lower $T_g$ value was recorded in the samples with 5% compared to the samples with 1%. When examined based on the gum concentration, the $T_g$ value increased when the concentration was increased from 0.5–1%, then decreased. Similar to our results Wang et al. [55] stated that the $T_g$ value increased between 0%-10% and decreased between 10% and 30% in films to which zein was added.

During the thermal analysis, the continuous heating process initiates the thermal decomposition of the films. This temperature expresses the melting temperature ($T_m$) of the film. An endothermic activity (initial decomposition temperature) was observed in the film samples in the range of 85°C to 131°C, possibly due to the evaporation of water. The exothermic peaks formed between 248 °C and 292 °C represent the maximum decomposition temperature at which the films decompose (Online Resource 1). This temperature is also an indicator of thermal stability. Thermal stability increased with increasing glycerol concentration in the film. Based on the gum concentration, the stability increased up to 1%, while the thermal stability of the samples with 1.5% gum added decreased. The best thermal stability value was recorded in the film sample with the lowest ratio of gum (0.5%) and the highest ratio of glycerol (5%).

### 3.10 Biodegradability

Biodegradability results are important in terms of qualifying the film as environmentally friendly packaging. Degradation of polymers in a bioactive environment; occurs through the breakdown of material followed by mineralization. The effect of heat and moisture, as well as the enzymatic activity of microorganisms, shortens and weakens polymer chains [56]. For this reason, the degradation times of the films in the soil vary depending on the material used in film production and environmental conditions. For example, Cerruri et al. [57] reported the period of complete dissolution and destruction in the soil as 30 days in their studies where they developed starch-based polymers. Similarly, Xiong et al. [58] who developed starch-based films, determined the degradation time of the films as 100 days.

The time-dependent degradation changes of BSG-based films buried in the soil within the scope of the research are given in Fig. 4. During the first 15 days of burial, no change was observed in the film samples. As of the 21st day, the film samples started to degrade by showing a mass loss. Since the increase in the gum concentration increases the film thickness, the films with high gum ratio were completely dissolved in a long time compared to the others (60th day). The low water solubility of BSG-based films is also thought to be effective in prolonging the degradation time. The increase in the glycerol ratio had a positive effect on the degradation. As the glycerol ratio in the film increased, the solubility of the film in the soil increased.

### 3.11 Color changes of films in acid/alkaline conditions

The onset of spoilage in some foods results in changes in the acidity of the food. In this case, if the food is covered with a film, the color tone of the film will change depending on the increase or decrease in pH. For this reason, the color change of the film in an acid and alkali environment is important in terms of helping the consumer to control the food quality based on the color change. Color changes were observed after BSG films were placed in both media
When the samples were immersed at pH = 3, the basic color greenish yellow changed to very light yellow. When immersed in pH = 12, the greenery in the films increased and became darker than the original state (Online Resource 2). This difference was observed more clearly as the gum concentration in the films increased. Increasing the glycerol ratio negatively affected the monitoring of the acid/alkali effect. For this reason, the most ideal film can be obtained with the choice of high gum concentration and low glycerol ratio. Similar to our research result, Medina-Jaramillo et al. [24] also reported that when the film they prepared with the addition of basil extract was immersed in an acid medium, they obtained a yellow color close to white.

4. Conclusion

In this research, the gum obtained from basil seed was used as a film component, and its potential for use in the food industry was discussed by investigating the various properties of this film. When the BSG-based film results were examined, it was determined that the water solubility and water vapor permeability of the films especially with low gum and high plasticizer concentrations was high. This result showed that BSG film is not a suitable alternative, especially for some food products that should not absorb moisture. On the other hand, optimum results were obtained from the film samples in terms of color and opacity criteria. The compatibility of BSG and glycerol in the film matrix was supported by AFM images, XRD patterns, and FTIR spectra. It has been found that films with low gum and high glycerol ratio produce almost smooth surfaces. As a result of all these analyzes, it was concluded that the mechanical and barrier properties and thermal stability of BSG-based films are quite good. In addition, the complete dissolution of this material in the soil within 60 days showed that this material could be considered an environmentally friendly packaging. The results obtained within the scope of the research have shown that BSG-based films with appropriate gum and plasticizer ratios exhibit properties that can compete with other edible films and synthetic packaging.

Declarations

Declaration of competing interest

The authors have declared that there is no conflict of interest.

Acknowledgements

The authors thank the Selcuk University Scientific Research Projects Coordinatorship for the financial support to this research with grant number 20401023.

References


Figures

Figure 1
X-ray diffraction patterns of film samples (a:b:c/0.5%BSG:1,3,5% GLY; d:e:f/1%BSG: 1,3,5 GLY; g:h:i/ 1.5BSG: 1, 3, 5 GLY; BSG: basil seed gum, GLY: Glycerol)

Figure 2

AFM images of film samples (a:b:c/0.5%BSG:1,3,5% GLY; d:e:f/1%BSG: 1,3,5 GLY; g:h:i/ 1.5BSG: 1, 3, 5 GLY; BSG: basil seed gum, GLY: Glycerol)
Figure 3

FT-IR Spectrum of film samples (a:b:c/0.5%BSG:1,3,5% GLY; d:e:f/1%BSG: 1,3,5 GLY; g:h:i/ 1.5BSG: 1,3,5 GLY; BSG: basil seed gum, GLY: Glycerol)
Figure 4

Biodegradability of film samples at 15th, 21st, 30th, and 45th days (1,2,3: 0.5% BSG/1,3,5 % GLY; 4,5,6: 1% BSG/1,3,5 % GLY; 7,8,9: 1.5% BSG/1,3,5 % GLY; BSG: basil seed gum, GLY: Glycerol)

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