DEVELOPMENT AND CHARACTERIZATION OF BIODEGRADABLE FOOD PACKAGING FILMS FROM FOOD INDUSTRIAL WASTES

Muhammad Adnan Hafeez 1,*, Abid Aslam Maan1, Masood Sadiq Butt1 and Muhammad Anjum Zia2

1National Institute of Food Science and Technology, Faculty of Food, Nutrition and Home Sciences, University of Agriculture, Faisalabad, Pakistan
2Department of Biochemistry, Faculty of Sciences, University of Agriculture, Faisalabad, Pakistan
*Corresponding author’s e-mail: Hafeez_ma@outlook.com

The demand of food packaging has increased along with the food consumption. Among all types of packaging materials plastics are most commonly used owing to their good mechanical, barrier and permeability characteristics. However, plastics are totally non-biodegradable and their raw materials come from non-renewable resources. This has urged the researchers to explore biodegradable food packaging films from renewable resources. Natural polymers such as carbohydrate, starch, cellulose, hemicellulose or pectin can be effectively used for the synthesis of biodegradable food packaging films. Fruits and vegetables processing industries are producing huge amount of waste in the form of peels, seeds and pomace. Being rich source of biopolymers, these wastes can serve as a good source of raw material for synthesis of biodegradable packaging films. In the present research, three types of food industrial wastes (potato, citrus and apple residues) were used with varying glycerol concentrations to develop biodegradable food packaging films. The prepared films were analyzed on the basis of their thickness, water vapor permeability (WVP), opacity, mechanical properties and photo oxidation. Results showed that potato residue film was thicker (0.17 mm) and more opaque (97.45 AU nm) in comparison with citrus and apple residue films. Results for mechanical properties showed that potato residue film possessed greater tensile (8.99 MPa) and puncture strength (13.78 N). However, it exhibited lower elongation at break (9.54 %) compared to citrus and apple residue films. While citrus residue film showed higher elastic modulus compared to potato and apple residue films. Water vapor permeability of all the prepared films was comparable. Increased glycerol concentration in film forming solutions resulted in the increase in thickness, opacity, water vapor permeability and elongation at break of all the films. However, increased glycerol concentration caused a decrease in tensile strength, elastic modulus and puncture strength of all the films. The structure of organic compounds present in prepared biodegradable films was evaluated using Fourier Transform Infrared Spectroscopy (FTIR) technique.

Keywords: Food packaging, plastic packaging, biodegradable films, food industrial wastes.

INTRODUCTION

The global packaging industry was reported to be of worth $931.1 billion in 2018 and is expected to reach $1177.7 billion in 2025 (Expresswire, 2019). The food packaging comprises the largest portion of this packaging cost amounting $293 billion in 2018 and expected to reach $423.27 billion in 2025 (Shahbandeh, 2019). Currently, petrochemical-based (polythelene, polyvenyl chloride, and polypropylene etc.) plastic films are used abundantly as packaging materials owing to their large availability and good mechanical strength (tensile properties), permeability to water vapors, carbon dioxide, oxygen, aroma compounds etc. and heat seal ability (Sorrentino et al., 2007). These plastics originate from non-renewable resources and are totally non-biodegradable leading to serious disposal problems. The manufacturing process of plastic packaging also involves higher energy cost (Ferreira et al., 2016). Toxic chemical ingredients present in the packaging material often contaminate the food stuff. Keeping in view the issues related to plastic packaging materials, development of biodegradable packaging is not only a useful necessity but also an important environmental requirement (Siracusa et al., 2008). Biodegradable packaging materials are based on renewable resources, cost effective and are environment friendly (Shen et al., 2015). Researchers are continuously exploring the biological sources suitable for the preparation of biodegradable films and coatings. Biodegradable films are prepared using natural biopolymers and biological materials such as polysaccharides, proteins, lipids, polyeseters and derivatives (Franco et al., 2017). Use of these natural biopolymers in food packaging films is highly safe because of the absence of toxins or harmful chemicals. These biopolymers break down into harmless products and get absorbed into the soil after being used (Khalil et al., 2018). Biodegradable films and coatings possess ability to lower the metabolic rate of fruits and vegetables (Cardoso et al., 2017). Properly formulated biodegradable films have been reported
to maintain the quality characteristics of a variety of fresh fruits and vegetables including strawberries (Mali and Grossmann, 2003; Gol et al., 2013; Wang and Gao, 2013; Franco et al., 2017), blueberries (Almenar et al., 2010), carambola (Gol et al., 2015), avocado (Aguilar-Mendez et al., 2008), carrots (Mensiti et al., 2011) fresh cut carrots (Fai et al., 2016), carrot slices (Moreira et al., 2011) baby corn and Chinese cabbage (Pitak and Rakshit, 2011), minimally processed lettuce (Del Nobile et al., 2008) fresh cut apples (McHugh and Senesi, 2000; Sangsawan et al., 2008; Chiumarelli and Hubinger, 2012), fresh cut papaya (Tapia et al., 2008), acerolas (Ferreira et al., 2016), tomatoes (Athinasevî et al., 2013), mango (Chonhenchob et al., 2007), mango puree (Azeredo et al., 2009), Hayward kiwifruit (Benitez et al., 2013), grapes (Chauhan et al., 2014), Red Crimson grapes (Fakhouri et al., 2015), pineapple (Chonhenchob et al., 2007) etc. by reducing respiration, gaseous exchange, moisture and solute migration and oxidative reaction rates (Cortez-Vega et al., 2014). Fruits and vegetables have been used as an alternative to the direct use of biopolymers for the preparation of biodegradable films (McHugh and Olsen, 2004; Sothornvit and Pitak, 2007; Azeredo et al., 2009; Du et al., 2011; Azeredo et al., 2012; Martelli et al., 2013). The first scientific research on the potential of fruits and vegetables for development of biodegradable packaging materials was reported in 1990s. This paved the way for fruits and vegetables purees, extracts and pulp to be used in biodegradable packaging (Du et al., 2008; Cerqueira et al., 2011; Ravishankar et al., 2012; Lorevice et al., 2012; Martelli et al., 2013; Torres et al., 2015; Orsuwan et al., 2016; Kadzińska et al., 2019). However, fruits and vegetables are the essential components of a healthy diet and their direct consumption in biodegradable films can lead to the issues of food security forever growing populations. Industrial processing of fruits and vegetables produces an enormous amount of wastes in the form of peels, pulp and seeds etc. (Campos et al., 2014; Ferreira et al., 2015). These waste materials are a rich source of biopolymers (polysaccharides and dietary fibers) and bioactive compounds (Babbar et al., 2011; Ajila and Rao, 2013). Owing to their low market value, most of these wastes are underutilized and are used as fertilizer and animal feed or rather discarded into soil representing not only an environmental issue but also wastage of potential nutrients and biopolymers (Park and Zhao, 2006). Proper handling and utilization of these materials in biodegradable film formation can provide new market for value added by products (Saravacos et al., 2011).

Potato is the major vegetable crop of Pakistan and is cultivated over and area of 177297 hectares with total annual production of 3974 thousand tons (Ahmed et al., 2017). It is processed into a variety of products including potato chips, French fries, potato protein, potato powder etc. About half of the potato solids are wasted during its industrial processing. Potato peel accounts for 15 to 40% of this potato waste. (Ali et al., 2015). Citrus (orange) is the major fruit produced in Pakistan and is cultivated on 192230 hectares with an annual production of 2344 thousand tones(Ahmed et al., 2017). Processing of citrus (into juices, essential oils and other products) produces massive amount of wastes in the form of pulp, peels and seeds. The resulting waste accounts for about 50% of the fresh fruit, comprising of approximately 10% seeds, 30-35% internal tissues and 60-65% peels. The waste is rich in pectin, cellulose and hemicellulose (Mannepula et al., 2015). Apple is the fourth major fruit of Pakistan regarding production and is cultivated on 96928 hectares. Its annual production is reported to be 620 thousand tons (Ahmed et al., 2017). Processing of apples generates peels, stems, seeds and soft tissues as by product which represents about 30% of the total input. This waste is rich in pectin and cellulose (Sablani et al., 2009). The present study was planned to explore the potential of locally generated fruit and vegetable wastes for development and characterization of biodegradable food packaging films.

**MATERIALS AND METHODS**

**Procurement and preparation of raw materials:** Fresh potatoes, citrus (orange) and apples were procured from fruits and vegetables market of Faisalabad. These fruits and vegetable were hand washed to remove the dust and other foreign matters. After washing, all the three commodities were manually peeled and subjected to juice extraction (apple and citrus only). The obtained peels and pomace were dried at 60°C for 6 to 8 hours in hot air oven. The dried residues (peels and pomace) were subjected to size reduction through grinding to get fine powder. This fine powder (residue flour) was then subjected to sieving through 40 (420 μm) mesh sieve to get residue flour of the uniform average particle sizes. Analytical grade glycerol, digestion tablets, sulfuric acid, sodium hydroxide, boric acid, petroleum ether, sodium azide, α and β amylase, pectinase, cellulase, methyl red and ethylene blue indicators were obtained from Sigma-Aldrich (Steinheim, Germany).

**Proximate composition of residue flours:** Proximate composition of residue flours was analyzed according to the method described in AACC (2000). The moisture content was measured by hot air oven at 105±5°C. Ash content was estimated by charring and complete burning in muffle furnace at 550 °C for 5-6 hours till greyish end point. Crude protein was determined by kjeldhal method in three stages consisting of digestion with acid, distillation with base and boric acid and titration with acid neutralization. Crude fat was determined by extraction with n-Hexane in Soxhlet apparatus. Crude fiber was measured by igniting the sample at 550-650 °C for 3-5 hours in the muffle furnace after digesting it with acid and base. Following equation was used to determine the nitrogen free extracts (NFE) of the residue flours.

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**Proximate Composition of Residue Flours**

- **Potato Peel Flours**
  - **Moisture:** 7.5% ± 0.2%
  - **Protein:** 6.2% ± 0.1%
  - **Fat:** 1.8% ± 0.1%
  - **Fiber:** 13.2% ± 0.3%
  - **Ash:** 2.1% ± 0.1%

- **Citrus Peels Flours**
  - **Moisture:** 8.0% ± 0.3%
  - **Protein:** 5.8% ± 0.2%
  - **Fat:** 2.0% ± 0.1%
  - **Fiber:** 14.3% ± 0.4%
  - **Ash:** 2.3% ± 0.2%

- **Apple Pomace Flours**
  - **Moisture:** 6.5% ± 0.2%
  - **Protein:** 6.0% ± 0.1%
  - **Fat:** 1.5% ± 0.1%
  - **Fiber:** 12.8% ± 0.3%
  - **Ash:** 2.5% ± 0.2%
Film preparation: Three g of each residue flour was dissolved in 97ml distilled water to prepare three different film forming solutions. The solutions were pre-homogenized by Ultra-Turrax homogenizer (T18 D, IKA-Germany) at 20,000rpm for 5 minutes (Sablani et al., 2009). The film forming solutions were subjected to stirring through magnetic stirrer for 1 hour to get the uniform mixture. Film forming mixtures were then subjected to sonication (400W) to disrupt the biopolymers in the solution. The said solutions were subjected to heating at 90°C for 30 minutes in water bath and cooled to 5°C (apple residue solution) or 20°C (potato and citrus residue solutions) in an ice box. A final homogenization of the film forming solutions was carried out at 20,000 rpm for 5 minutes after glycerol addition. Glycerol concentration was varied from 25% to 35% in all films. Finally, these solutions were subjected to degassing for the removal of air bubbles. The film forming solutions were cast on Teflon plates having 15.5 cm internal diameter. Approximately, 68 g of each solution was poured in the Teflon plates separately. After drying at 23 ± 2°C and 35 ± 5% relative humidity for 48 to 72 hours, the dried films were detached from the Teflon plates for further analyses (Kang and Min, 2010).

Film characterization

Film thickness: Each film sample was analyzed for its thickness at five different positions using digital micro-meter and average of these measurements were used for further analyses according the method described by Kang and Min (2010).

Water vapor permeability: The prepared films were sealed separately on permeation cells containing distilled water and were kept in the desiccator to measure the water vapor permeability. These permeation cells were weighed at regular intervals for 2-3 days to check the water vapor flux (Alves et al., 2010).

Opacity: Prepared film samples were cut into 1cm × 3cm size and absorbance spectrum between 400-800 nm were measured after placing these samples on the inner side of the spectrophotometer (Model: U2020, Serial- 20A Irmeo, Germany) cell. Area under curve was considered as the opacity of the films and expressed as the absorbance units × nano-meters (Mali et al., 2004).

Mechanical properties: Films were cut into 8 cm × 0.5 cm size and held between the grips of tensile tester (TST-01, Cell Instruments, China). Tensile strength, elastic modulus and elongation at break of the film samples were recorded by using 5mm/minute crosshead speed (Almasi et al., 2010). Moreover, film samples were cut into 40mm diameter and fixed on plate of tensile tester via adhesive tape to measure the puncture strength of the films. A cylindrical probe was moved perpendicularly at a constant speed of 1mm/s until the probe passed through the film. At rupture point, force-deformation curves were recorded (Mali et al., 2004).

Photo oxidation: The prepared film samples of specific dimensions were analyzed through Fourier Transform Infrared Spectroscopy (FTIR) to get the absorption spectra of the films. The interference of water vapors was reduced by equipping the instrument with dehumidifier (silica gel). An average of 20 scans were taken as spectra at a resolution of 4cm⁻¹ in the range of 400-4,000 cm⁻¹ through FTIR (Bruker Tensor 27) (Banisadr and Asempour, 2012).

RESULTS AND DISCUSSIONS

Proximate composition of residue flour: Proximate analysis of residue flours of potato, citrus and apple is summarized in Table 1. The highest moisture content was observed for the potato residue flour (13.33 ± 0.66%) followed by residue flours of citrus (10.0 ± 0.50%) and apple (3.3 ± 0.16%). Similarly, the highest protein contents were also recorded for the potato residue flour (15.31 ± 0.76%) and lower protein content was found in residue flour of citrus (4.37 ± 0.21%) and apple (8.33 ± 0.41%). Residue flours of citrus and apple were found to have 2 ± 0.1% fiber while potato residue flour exhibited 1.4 ± 0.07% fiber. In terms of the fat content in the residue flour of apple had the higher content (4.59 ± 0.22%) followed by citrus (4.46 ± 0.22%) and potato (1.4 ± 0.07%) residue flour. Ash content represents the mineral content of the sample. Highest ash contents were observed in potato residue flour (9.38 ± 0.46%) followed by residue flours of citrus (4.89 ± 0.24%) and apple (2.39 ± 0.11%). The protein contents of apple residue flour were greater than the value of 6.7% reported by Akpabio et al. (2012). Fat content of potato residue flour was comparable with findings of Bretón-Toral et al. (2016) who stated it to be 2.1%. Romelle et al. (2016) reported fat content of citrus and apple peel powders to be 8.70 ± 0.65% and 9.96 ± 1.52%, respectively. Nitrogen free extract (NFE) represents the carbohydrate content and was calculated from the aforementioned proximate composition on difference basis. According to Table 1, the highest NFE was found in residual flour of apple (79.39 ± 3.96%), which are similar with the outcomes of Akpabio et al. (2012). Lower NFE contents were observed in potato residue flour (57.97± 2.89%) followed by residue flour of citrus (74.28 ± 3.71%). Results of potato residue flour are comparable with the values of Bretón-Toral et al. (2016) who reported NFE contents in potato peel waste as 51.3%. The high content of NFE in the tested flours reflects their potential for preparation of biodegradable films. These results are similar to the findings of Romelle et al. (2016) who reported the chemical composition of different fruit peels (orange peel: 9.7 ± 0.6% crude protein, 14.19 ± 0.01% crude fiber, 5.17 ± 0.98% ash and 53.27 ± 0.10% carbohydrates) (apple peel: 2.80 ± 0.17% crude protein, 13.95 ± 0.10% crude fiber, 1.39 ± 0.14% ash and 59.96 ± 0.44% carbohydrates). Film thickness: Thickness of the biodegradable film plays a vital role in determining the other physical properties of film including mechanical and water vapor permeability (Laothakunjit and Noomhorm, 2004). At 25% glycerol
concentration, minimum thickness was observed in apple residue film followed by citrus residue film and potato residue film. The results showed that glycerol concentration had a highly significant effect on films thickness. The increase in glycerol concentration in film forming solutions caused an increase in the thickness of all the films irrespective of the raw material used (Table 2). Higher film thickness with greater glycerol concentration might be due to the presence of higher dry matter contents in film forming solutions (Nemeth et al., 2010). These results are similar to the findings of Kang and Min (2010) who developed potato peel based biodegradable films with 0.1mm thickness. Rezvani et al. (2013) reported that the thickness of the films prepared by using different concentrations of sodium caseinate and stearic acid ranged from 0.12 mm to 0.18 mm. The present results are also similar to the findings of Kechichian et al. (2010) who formulated cassava starch based biodegradable films incorporated with natural antimicrobial agents.

**Water vapor permeability:** Minimum WVP was observed in apple residue film followed by citrus and potato residue film. Lower WVP values of apple residue films has also been reported in previous study by Vargas et al. (2008) who explained that the higher WVP of potato residue films were due to the higher starch contents present in potato peels. Sablani et al. (2009) also reported the lower WVP of apple peel based edible films than potato and corn starch based edible films. The results showed that glycerol concentration had a significant effect on WVP of the films. The increase in glycerol concentration in film forming solutions caused an increase in WVP of all the films (Table 2). The increase in WVP might be due to the effect that the inclusion of glycerol molecules between polymer chains increased the molecular mobility in the film matrix by decreasing intermolecular attractions (increased inter-chain space). Resultantly, this increased mobility produced greater free volume in the matrix which allowed the movement of water vapor molecules through the film (Sothornvit and Krochta, 2000; Rodriguez et al., 2006). The increase in WVP could also be related with the high hydrophilicity of glycerol molecules which is favorable to the adsorption of water molecules (Kang and Min, 2010). A comparison of these results can be made with the conclusions of Sablani et al. (2009) who developed apple peels based edible films and reported that highest WVP values were observed at increased glycerol level (44%). Kang and Min (2010) also formulated potato peel based edible films and concluded that the increased glycerol concentration caused an increase in WVP of the films.

**Opacity:** Opacity of the films is the property of prime importance in food packaging and coating. It provides information about the dispersed particles in carbohydrate-based films. Particle sizes larger than visible wavelength cause hinderance in light and produce opaque or translucent films (Kampeeprapappun et al., 2007). Lower opacity values (Table 2) represent more transparency. Residue films prepared with 25% glycerol concentration showed greater transparency. Among the prepared films, apple residue film was more transparent followed by citrus and potato residue film. Greater transparency of apple and citrus residue films might be due to the presence of higher pectin contents in these residue films as also reported by Venkatesh and Sutariya (2019). Results showed that glycerol concentration had a highly significant effect on films opacity. The increase in glycerol concentration in film forming solutions caused a decrease in film transparency (Table 2). The results depicted

### Table 1. Proximate composition of potato, citrus and apple residue flours

<table>
<thead>
<tr>
<th>Film material</th>
<th>Moisture (%)</th>
<th>Ash (%)</th>
<th>Protein (%)</th>
<th>Fat (%)</th>
<th>Fiber (%)</th>
<th>NFE (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PRF</td>
<td>13.33±0.66</td>
<td>9.38±0.46</td>
<td>15.31±0.76</td>
<td>2.61±0.13</td>
<td>1.40±0.07</td>
<td>57.97±2.89</td>
</tr>
<tr>
<td>CRF</td>
<td>10.00±0.50</td>
<td>4.89±0.24</td>
<td>4.37±0.21</td>
<td>4.46±0.22</td>
<td>2.00±0.10</td>
<td>74.28±3.71</td>
</tr>
<tr>
<td>ARF</td>
<td>3.30±0.16</td>
<td>2.39±0.11</td>
<td>8.33±0.41</td>
<td>4.59±0.22</td>
<td>2.00±0.10</td>
<td>79.39±3.96</td>
</tr>
</tbody>
</table>

PRF: Potato residue flour; CRF: Citrus residue flour; ARF: Apple residue flour

### Table 2. Effect of glycerol on physical characteristics of the films prepared from potato, citrus and apple residue films

<table>
<thead>
<tr>
<th>Film material</th>
<th>Grams in 97ml film forming solution (w/w)</th>
<th>Glycerol (%)</th>
<th>Thickness (mm)</th>
<th>WVP (g mm/kPa h m²)</th>
<th>Opacity (AU mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PRF</td>
<td>3</td>
<td>25</td>
<td>0.07±0.0044&lt;sup&gt;ef&lt;/sup&gt;</td>
<td>2.81±0.33&lt;sup&gt;ab&lt;/sup&gt;</td>
<td>81.23±1.02&lt;sup&gt;d&lt;/sup&gt;</td>
</tr>
<tr>
<td>PRF</td>
<td>30</td>
<td>0.10±0.0078&lt;sup&gt;c&lt;/sup&gt;</td>
<td>3.29±0.38&lt;sup&gt;ab&lt;/sup&gt;</td>
<td>85.10±1.03&lt;sup&gt;c&lt;/sup&gt;</td>
<td></td>
</tr>
<tr>
<td>PRF</td>
<td>35</td>
<td>0.17±0.0045&lt;sup&gt;a&lt;/sup&gt;</td>
<td>3.97±0.60&lt;sup&gt;a&lt;/sup&gt;</td>
<td>97.45±1.36&lt;sup&gt;a&lt;/sup&gt;</td>
<td></td>
</tr>
<tr>
<td>CRF</td>
<td>25</td>
<td>0.06±0.0054&lt;sup&gt;fg&lt;/sup&gt;</td>
<td>2.75±0.35&lt;sup&gt;ab&lt;/sup&gt;</td>
<td>70.45±1.31&lt;sup&gt;e&lt;/sup&gt;</td>
<td></td>
</tr>
<tr>
<td>CRF</td>
<td>30</td>
<td>0.10±0.0065&lt;sup&gt;cd&lt;/sup&gt;</td>
<td>3.15±0.54&lt;sup&gt;ab&lt;/sup&gt;</td>
<td>78.12±1.15&lt;sup&gt;d&lt;/sup&gt;</td>
<td></td>
</tr>
<tr>
<td>CRF</td>
<td>35</td>
<td>0.13±0.0072&lt;sup&gt;b&lt;/sup&gt;</td>
<td>3.46±0.56&lt;sup&gt;ab&lt;/sup&gt;</td>
<td>89.45±1.19&lt;sup&gt;b&lt;/sup&gt;</td>
<td></td>
</tr>
<tr>
<td>ARF</td>
<td>25</td>
<td>0.05±0.0058&lt;sup&gt;gs&lt;/sup&gt;</td>
<td>2.48±0.41&lt;sup&gt;b&lt;/sup&gt;</td>
<td>63.03±1.11&lt;sup&gt;c&lt;/sup&gt;</td>
<td></td>
</tr>
<tr>
<td>ARF</td>
<td>30</td>
<td>0.08±0.0053&lt;sup&gt;de&lt;/sup&gt;</td>
<td>3.01±0.49&lt;sup&gt;ab&lt;/sup&gt;</td>
<td>72.11±1.04&lt;sup&gt;c&lt;/sup&gt;</td>
<td></td>
</tr>
<tr>
<td>ARF</td>
<td>35</td>
<td>0.10±0.0078&lt;sup&gt;c&lt;/sup&gt;</td>
<td>3.34±0.31&lt;sup&gt;ab&lt;/sup&gt;</td>
<td>85.76±1.01&lt;sup&gt;c&lt;/sup&gt;</td>
<td></td>
</tr>
</tbody>
</table>

PRF: Potato residue films; CRF: Citrus residue films; ARF: Apple residue films
that the thicker films showed greater opacity values compared to the films having lower thickness. Wu et al. (2009) explained this phenomenon as thickness of the films influenced the transmittance of light through the film resulting in the more opaque film. Mali et al. (2004) also demonstrated that opacity of the yam starch films also increased with the increase in film thickness. Transparency of the films is only required when the packaged food has to be visible. While, mechanical and barrier properties of the films depend upon the concentration of glycerol in film forming solutions. The increase in opacity with higher glycerol concentration might be due to the increased compaction of polymeric chain which modified the refractive index resulting in the restriction of passage of light passing through the film matrix (Saberi et al., 2016). Similar results were also reported by Paschoalick et al. (2003) and do Amaral Sobral et al. (2004). These authors related this phenomenon with the physical characteristics of the plasticizer and its dilution effect in the film forming solution. The results of the present study are in agreement with the findings of Shaw et al. (2002) who also found increased films opacity with increased glycerol concentrations.

**Mechanical properties:** Mechanical properties of films are required to sustain their integrity when applied to a food product. Tensile strength is the maximum tension which a film can support (Pereda et al., 2012). Among the prepared films, potato residue film showed greater tensile strength followed by citrus and apple residue film. Greater tensile strength of potato residue film was due to the presence of higher starch contents which formed a more compact and cohesive polymeric matrix (Ferreira et al., 2016). The results depicted that glycerol concentration had a highly significant effect on tensile strength of residue films. The increase in glycerol concentration in film forming solution caused a decrease in tensile strength of the films (Table 3). This decrease in tensile strength of the residue films might be explained by the plasticizing effect of glycerol that formed more homogeneous aggregation of biopolymer materials. This effect might increase the intermolecular spaces between the biopolymer molecules and produced a greater free volume resulting in the film flexibility (Sablani et al., 2009). Similar results were also observed by Sablani et al. (2009) who developed apple peel films and stated that increased glycerol concentration caused a decrease in tensile strength of the films. These findings are also in accordance with the results of Kang and Min (2010) who prepared potato peel films and reported that greater increase in glycerol concentration lessened the tensile strength of the films. Decrease in tensile strength of the films with the increase in glycerol concentration was also reported in previous studies (McHugh and Krochta, 1994; Park et al., 1994; Butler et al., 1996).

Elastic modulus of the film is its ability to resist its deformation against the stress applied. Citrus residue film showed greater elastic modulus followed by potato residue film and apple residue film. The difference in elastic modulus of these films could be attributed to the difference in the ratio of dietary fibers to total sugars (McHugh and Olsen, 2004). The results depicted that glycerol concentration had a highly significant effect on elastic modulus of residue films. The increase in glycerol concentration in film forming solution caused a decrease in elastic modulus of the films (Table 3). The decrease in elastic modulus of residue films with the increased glycerol concentration could be due to the plasticizing effect of glycerol. This effect increased the free volume by increasing intermolecular spaces between the biopolymer chains, which enhanced the film flexibility (Sablani et al., 2009). These results are similar to the findings of Kang and Min (2010) who prepared potato peel based biopolymer films with varying concentrations of glycerol and reported that the elastic modulus of the films decreased with the increasing concentration of glycerol. However, these values are lower than the values reported by Zhang and Whistler (2004) who prepared corn hull arabinoxylan based thin film. Sablani et al. (2009) formulated apple peel based edible films and described the similar trend of decreased elastic modulus with increased glycerol concentration.

The maximum variation in length of a film prior to breakage after being subjected to tension is called elongation at break (Pereda et al., 2012). Apple residue film showed higher elongation at break followed by citrus residue film and potato films prepared from potato, citrus and apple residue films.

### Table 3. Effect of glycerol on mechanical properties of the films prepared from potato, citrus and apple residue films

<table>
<thead>
<tr>
<th>Film material</th>
<th>Grams in 97ml film forming solution</th>
<th>Glycerol (%) (w/w)</th>
<th>Tensile strength (MPa)</th>
<th>Elastic modulus (MPa)</th>
<th>Elongation at break (%)</th>
<th>Puncture strength (N)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PRF</td>
<td>3</td>
<td>25</td>
<td>8.99±1.23&lt;sup&gt;a&lt;/sup&gt;</td>
<td>352.38±3.67&lt;sup&gt;d&lt;/sup&gt;</td>
<td>9.54±2.59&lt;sup&gt;b&lt;/sup&gt;</td>
<td>13.78±0.65&lt;sup&gt;c&lt;/sup&gt;</td>
</tr>
<tr>
<td>PRF</td>
<td>30</td>
<td>35</td>
<td>5.98±1.07&lt;sup&gt;ab&lt;/sup&gt;</td>
<td>237.88±4.58&lt;sup&gt;e&lt;/sup&gt;</td>
<td>10.33±3.61&lt;sup&gt;b&lt;/sup&gt;</td>
<td>12.01±0.21&lt;sup&gt;bc&lt;/sup&gt;</td>
</tr>
<tr>
<td>PRF</td>
<td>25</td>
<td>7.73±1.22&lt;sup&gt;a&lt;/sup&gt;</td>
<td>180.91±2.34&lt;sup&gt;d&lt;/sup&gt;</td>
<td>11.49±2.16&lt;sup&gt;ab&lt;/sup&gt;</td>
<td>10.54±0.43&lt;sup&gt;c&lt;/sup&gt;</td>
<td></td>
</tr>
<tr>
<td>CRF</td>
<td>25</td>
<td>1.75±1.55&lt;sup&gt;a&lt;/sup&gt;</td>
<td>720.01±2.90&lt;sup&gt;b&lt;/sup&gt;</td>
<td>10.34±3.24&lt;sup&gt;bc&lt;/sup&gt;</td>
<td>10.01±0.54&lt;sup&gt;d&lt;/sup&gt;</td>
<td></td>
</tr>
<tr>
<td>CRF</td>
<td>30</td>
<td>6.87±1.17&lt;sup&gt;a&lt;/sup&gt;</td>
<td>576.23±5.32&lt;sup&gt;b&lt;/sup&gt;</td>
<td>14.99±3.02&lt;sup&gt;abc&lt;/sup&gt;</td>
<td>8.88±0.33&lt;sup&gt;de&lt;/sup&gt;</td>
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<tr>
<td>CRF</td>
<td>35</td>
<td>5.61±0.15&lt;sup&gt;ab&lt;/sup&gt;</td>
<td>441.78±5.43&lt;sup&gt;c&lt;/sup&gt;</td>
<td>19.81±2.86&lt;sup&gt;e&lt;/sup&gt;</td>
<td>6.55±0.21&lt;sup&gt;f&lt;/sup&gt;</td>
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<tr>
<td>ARF</td>
<td>25</td>
<td>3.19±1.03&lt;sup&gt;bc&lt;/sup&gt;</td>
<td>40.85±6.79&lt;sup&gt;d&lt;/sup&gt;</td>
<td>11.87±3.55&lt;sup&gt;abc&lt;/sup&gt;</td>
<td>8.15±0.67&lt;sup&gt;ef&lt;/sup&gt;</td>
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<tr>
<td>ARF</td>
<td>30</td>
<td>2.89±1.01&lt;sup&gt;bc&lt;/sup&gt;</td>
<td>36.78±6.09&lt;sup&gt;e&lt;/sup&gt;</td>
<td>13.31±3.61&lt;sup&gt;abc&lt;/sup&gt;</td>
<td>7.12±0.25&lt;sup&gt;g&lt;/sup&gt;</td>
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<tr>
<td>ARF</td>
<td>35</td>
<td>1.99±0.12&lt;sup&gt;c&lt;/sup&gt;</td>
<td>33.67±4.54&lt;sup&gt;f&lt;/sup&gt;</td>
<td>16.03±2.34&lt;sup&gt;bc&lt;/sup&gt;</td>
<td>4.98±0.15&lt;sup&gt;bc&lt;/sup&gt;</td>
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</table>

PRF: Potato residue films; CRF: Citrus residue films; ARF: Apple residue films
The results depicted that glycerol concentration had a significant effect on elongation at break of residue films. The increase in glycerol concentration in film forming solution caused an increase in elongation at break of residue films (Table 3). The increase in elongation at break percentage might be due to the effect that glycerol is more hygroscopic in nature due to its lower molecular weight and thus, increased glycerol concentration caused an increase in film plasticization (Zhang and Whistler, 2004). Kang and Min (2010) prepared potato peel based biopolymer films and found a decrease in elastic modulus and increase in elongation at break of biopolymer film with increased glycerol concentration. Sablan et al. (2009) also demonstrated the similar results that with increased glycerol concentration, elastic modulus decreased and elongation at break of apple peel based edible film was found to increase. The percent elongation of these residue films were also comparable with the results of Fai et al. (2016) who prepared biodegradable films from fruits and vegetable residues and reported that the percent elongation of films were in the range of 13 to 17%. Potato residue film showed more puncture strength than citrus and apple residue film. The greater puncture strength of potato residue film might be due to the presence of higher starch contents in potato residue. The results depicted that glycerol concentration had a highly significant effect on puncture strength of the prepared films. The increase in glycerol concentration in film forming solution caused a decrease in puncture strength of the films (Table 3). Puncture strength of the films depends upon the film thickness as well as the concentration of glycerol in the film forming solutions. The puncture strength of the films increases with increasing film thickness. This effect of film thickness on puncture strength was observed by Sobral (2000) in protein films. The author utilized a higher mass of same film forming solution and developed thicker films. The decrease in puncture strength with increased glycerol concentration was also reported by Mali et al. (2004) who postulated that puncture strength of yam starch films was influenced by the glycerol concentration in the film forming solution, film thickness and concentration of the starch. The authors further suggested that higher starch concentration, higher film thickness with lower glycerol contents produced films with higher puncture strength. These results also confirmed the observations of Mali and Grossmann (2003) who reported that the intermolecular forces along polymer chains decreased with increased plasticizer resulting in the increased film flexibility and decreased barrier properties.

**Photo oxidation:** Fourier Transform Infrared Spectroscopy (FTIR) technique was used to measure the structure of organic compounds present in prepared biodegradable films. FTIR analysis also exhibited the bonding between glycerol and the carbohydrate contents present in the raw material used. FTIR spectra of the films prepared from potato, citrus and apple residues have been presented in Fig. 1, 2 and 3, respectively.

![Figure 1. FTIR spectra of potato residue film (a: 25%, b: 30% and c: 35% glycerol)](image1)

![Figure 2. FTIR spectra of citrus residue film (a: 25%, b: 30% and c: 35% glycerol)](image2)

![Figure 3. FTIR spectra of apple residue film (a: 25%, b: 30% and c: 35% glycerol)](image3)

FTIR spectra of potato residue films (Fig. 1) depicted that hydroxyl stretching vibration caused the formation of a broad band in the region of 3361 cm\(^{-1}\) and the band in the region of 2931 cm\(^{-1}\) was because of the presence of CH\(_2\) symmetric and
asymmetric stretching vibration (Demirgoz et al., 2000; Xu et al., 2004). CH$_2$ bending vibration formed a peak at 1435 cm$^{-1}$ (Kim et al., 2003). The higher moisture content in the film caused the appearance of water absorption wavelength at 1642 cm$^{-1}$. The spectra of the films prepared with lower glycerol concentrations in the film forming solutions showed decreased and narrower peaks indicating the hydrogen interaction between glycerol and starch. Additionally, it was also due to the lower plasticization effect as reported by Liu et al. (2014). The FTIR spectra of the current study are almost similar to the spectra obtained by Guohua et al. (2006) and Sharif et al. (2019). FTIR spectra of citrus residue films depicted the most intense band in the region of 3412 cm$^{-1}$ due to the presence of OH in carbohydrate contents of the citrus residue (Fig. 2). C-H stretching vibration and bending vibrations formed a distinctive band at 2978 cm$^{-1}$ and 1437 cm$^{-1}$, respectively. The appearance of the peak at 1698 cm$^{-1}$ might be due to the aliphatic and/or unsaturated aromatic compounds as also reported by Demirbas (2000) and McKendry (2002). Similar findings were also observed by Zapata et al. (2009) while conducting the thermo kinetics study of orange peel in air. Furthermore, the spectra of the current study also resemble with the spectra observed by Arslanoglu et al. (2008). The FTIR spectra of apple residue films also showed the peak close to the citrus residue films (Fig. 3). However, the absorbance was lower than that of citrus residue films. The absorbance peaks were observed in the region of 3382 cm$^{-1}$ and 2983 cm$^{-1}$. This might be due to the presence of OH bonding in the pectin structure of the apple residue and C-H stretching, respectively. The absorbance peak at 1673 cm$^{-1}$ might be due to the presence of high methoxly pectin (Gnanasambandam and Proctor, 2000). Similar spectra were also recorded by Urias-Orona et al. (2010). The spectra of the citrus and apple residue films prepared with lower glycerol concentrations in the film forming solutions showed decreased and narrower peaks indicating the hydrogen interaction between glycerol and pectin.

Conclusions: Among the three biodegradable films prepared from potato, citrus and apple residues, potato residue film exhibited optimum thickness, good resistance to water vapor transmission and mechanical properties compared to citrus and apple residue films. Opacity of potato residue films was slightly higher than other films. The varying concentration of glycerol significantly affected the properties of the prepared films. Increased glycerol concentration in film forming solutions resulted in an increase in thickness, opacity, elongation at break and water vapor permeability of all the films. However, increased glycerol concentration caused a decrease in tensile strength, elastic modulus and puncture strength of all the films.

REFERENCES


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