

Article

The Potential of Apple and Blackcurrant Pomace Powders as the Components of Pectin Packaging Films

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Abstract: This work aimed to investigate the possibility of using apple and blackcurrant pomace powders to produce biopolymer packaging films as one of the actions to minimize waste in a circular economy approach. The fragmented fruit powders of 10 μm did not form a continuous film structure, thus apple pectin at the concentration of 5% was used as a film-forming agent in combination with fruit pomace (10%) and glycerol as plasticizer (50% of pectin, 2.5 g). The pectin control films and those produced with the addition of fruit pomace differed in appearance and physical properties. The films with fruit pomace were characterized by a higher thickness and much darker color in comparison with transparent pectin films. Lightness (parameter L^*) decreased from 87.24 to 21.09, and the film opacity increased from 1.03 to 17.14 A/mm, indicating the capacity of light adsorption. Fruit powder addition also affected mechanical resistance, and the films showed higher tensile strength (3.11%–6.72%) with lower elongation at break (5.11%–6.07%). Sorption and wetting analyses showed that fruit pomace-containing films had a lower capacity to absorb water. The water contact angle increased from 50.69 to 70.89°. Scanning electron microscopy (SEM) allowed us to observe significant changes in the structure related to the film composition, which affected the surface roughness and obtained a more rigid film structure.

Keywords: edible films; fruit pomace; apple pectin; sorption; tensile properties



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1. Introduction

The dynamics of the developing food industry have contributed to the intensified creation of innovative solutions in the packaging industry [1]. Analyzing global packaging production, it was noted that more than half of the packaging produced is destined for the food industry. The main materials used for packaging are plastic, glass, wood, metal, and paper [2]. The selection of a suitable packaging material depends on the intended use of the packaging. A properly selected material should be characterized by barrier properties and should also protect the interior of the product from penetration by microorganisms from the external environment. The role of barrier properties is to maintain the high quality of the product throughout its shelf life [3]. This is achieved by limiting the access of oxygen and moisture to the packaged product, which contributes to inhibiting the rate of biochemical transformations occurring in food. In turn, antimicrobial properties are designed to ensure the packaged product's safety against harmful microflora growth [4]. The indicated properties are not the only ones that characterize packaging. The use of edible coatings and films allows the introduction of various substances into products [5]. This type of packaging can serve as a carrier for dyes, flavors, and antioxidants, as well as sweeteners and antimicrobial substances. These substances directly affect the attractiveness or improve the nutritional value of the product [6].

One of the most commonly used packaging materials is plastic. This type of material is characterized by high mechanical strength and susceptibility to forming different shapes.

Despite the valuable properties of plastic, its use in the production of packaging is associated with a threat to the planet. This is due to the long decomposition time of its polymers, which consequently affects the growing problem of managing the resulting waste and its negative impact on the environment [7]. An alternative to plastics is biopolymers [8]. The production of completely biodegradable packaging is possible using natural polymers of plant and animal origin. An example of the use of biopolymers in the production of food packaging is films and edible coatings, which can be applied to food as protective structures. The coatings are an integral part of the food, while the films are independent structures that are outside the food [9]. One of the methods of obtaining edible films is by bottling film-forming solutions in a given mold and then subjecting the solution to a drying process under controlled technological conditions. The use of films limits the migration of ingredients from food and reduces water evaporation, which maintains the consistent quality of the packaged product [10].

Research is currently underway to select suitable components for the production of edible films. The use of polymers such as starch, alginate, and gelatin improves the mechanical properties of films. Starch-based films, due to their high elasticity, high tensile strength, and colorlessness, are used for coating meat, poultry, and fish. Starch is not the only polysaccharide used in the production of edible films [11]. Pectin is also a good raw material for the production of edible films due to its low production cost, wide availability, and ability to form coatings [12]. The creation of edible films from natural composites contributes to the management of waste raw materials from the food industry [13]. An example of the use of agricultural by-products is the formation of edible films based on oilseed flours and soy protein isolate as gelling agents [14]. A good waste raw material that can be used in the production of edible films is fruit processing waste, such as fruit pomace [15]. Fruit pomace is obtained by pressing and crushing whole fruits or fruit fragments and extracting their juices [16]. The resulting pomace is a cheap and readily available source of active compounds. Apple and blackcurrant pomace have a high content of dietary fiber. In addition, they contain significant amounts of polyphenols, which exhibit antioxidant activity. In addition to its high polyphenol and fiber content, pomace has a high pectin content [17]. Other fruit pomaces can also be used to make edible films. Sushmita et al. [18] used pineapple pomace in their study. The film with the addition of pineapple pomace was characterized by good physicochemical properties, including moisture, swelling index, thickness, and opacity. On the other hand, Aloui et al. [19] used tomato pomace to produce edible packaging. This film was characterized by good flexibility and thermal stability. However, waste pomace often does not undergo further processing, and it is disposed of using landfilling or incineration, which generates an increase in greenhouse gas emissions. A method that addresses the negative environmental effects associated with the production of plastic packaging and the wasted high biological potential of fruit pomace is the production of edible films and coatings that act as food packaging [20]. Edible films used as packaging are enriched with bioactive components derived from plant raw materials including, among others, fruit pomace, and thanks to phenomena such as absorption, adsorption, and permeation, bioactive components react with food, affecting its sensory, organoleptic, microbiological, and physicochemical characteristics [21]. This study aimed to develop and characterize edible packaging films prepared from apple pectin incorporated with apple and blackcurrant fragmented pomace as one of the actions to minimize waste, which is in line with a circular economy approach. The pomace, which is waste, is reused, thus minimizing waste production and also helping to reduce the generation of new plastic packaging.

2. Materials and Methods

The materials consisted of control apple pectin films and films containing apple or blackcurrant pomace powders, as well as a combination of both pomaces. Apple pectin was purchased from ZPOW Pektowin S.A., (Jasło, Poland). Fragmented fruit pomaces of 10 µm were supplied by Greenfield Sp. z o.o. Sp. k., (Warsaw, Poland). Glycerol and other

reagents were of analytical grade and were purchased from Avantor Performance Materials Poland S.A. (Gliwice, Poland).

2.1. Film Preparation

Aqueous film-forming solutions of 5% pectin and 10% fruit pomace addition were heated to 60 °C and stirred at 250 rpm/min for 15 min using an RCT basic IKAMAG magnetic stirrer (IKA Poland, Warsaw, Poland). Glycerol as a plasticizer was added in an amount of 50% relative to pectin (2.5 g). Control films were produced without the addition of fruit pomace (control films (-)). Three variants were prepared with fruit pomace powders: with the addition of apple pomace (AP films (-)), blackcurrant pomace (BC Films), and a combination of both types of pomace powders in a ratio of 1:1 (AP+BC Films). The mixtures were poured in a constant volume of 9 mL onto Petri dishes of 90 mm diameter and dried at 50 °C for 6 h in a laboratory dryer (model SLW 115 SMART PRO (POL-EKO APARATURA S p.j., Wodzisław Śląski, Poland)). After drying, the samples were removed from the dishes and were conditioned in a climate chamber (model KBF 240 (Binder, Germany)) at a relative humidity of the environment of 50% and a temperature of 25 °C for 48 h. Before studying water vapor sorption isotherms to reduce moisture content, the samples were dried at the temperature of 30 °C and pressure of 1000 Pa for 48 h using a vacuum dryer (Memmert V0 500, Buechenbach, Germany). Then, the dried samples were kept in a desiccator over phosphorus pentoxide (P₂O₅).

2.2. Film Thickness

The thickness of the films was measured at 5 points with a ProGage thickness gauge (Thwing-Albert Instrument Company (West Berlin, NJ, USA)) with an accuracy of ±1 µm.

2.3. Water Content

For each film, 1 g was weighed using an AE240 analytical balance (METTLER TOLEDO, Warsaw, Poland) with an accuracy of ±0.0001 g, and the samples were dried for 24 h at 100 °C in a laboratory dryer (model SLW 115 SMART PRO (POL-EKO APARATURA S p.j., Wodzisław Śląski, Poland)) and then weighed again. Water content was determined as a mass change before and after drying and was analyzed in 3 replicates.

2.4. Water Solubility

The 20 × 20 mm film pieces were placed in weighing dishes and weighed using an AE240 analytical balance (METTLER TOLEDO, Warsaw, Poland) with an accuracy of ±0.0001 g and then dried in a SUP-65 WG universal dryer (Wamed, Warsaw, Poland) for 24 h at 105 °C. The dried films were immersed in 25 mL of distilled water at 25 °C for 24 h with stirring at 50 rpm. The samples were then removed from the water, drained, and dried again for 24 h at 105 °C. Water solubility was calculated based on the difference in film weight before and after storage in water [14]. The measurement was carried out in 3 replicates.

2.5. Swelling Index

The 20 × 20 mm film pieces were weighed using an AE240 analytical balance (METTLER TOLEDO, Warsaw, Poland) to the nearest ±0.0001 g and then immersed in 25 mL of distilled water at 25 °C for 1 min. The samples were then dried and weighed again. Swelling was performed in 3 replicates and calculated based on differences in film weight.

2.6. Optical Properties

2.6.1. Color

The color of the films was measured in 8 replicates with a Minolta CR-400 colorimeter (Tokyo, Japan) in the CIE $L^*a^*b^*$ color system, where L^* indicates the lightness of the color, a^* indicates the proportion of green (−) or red (+), and the parameter b^* indicates the proportion of blue (−) or yellow (+). The films were tested using a white standard with

fixed values of $L^* = 92.37$, $a^* = -0.5$, and $b^* = 0.69$. The total color difference between the values for the standard and for the film was calculated according to the following equation [11]:

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

where:

ΔE —total color difference; L^* , a^* , and b^* —parameters for the white standard; and L , a , and b —parameters for the films.

2.6.2. Film Opacity

The film opacity analysis was performed in 6 replicates using an EVOLUTION 220 UV-Visiblespectrometer (ThermoElectron Corporation, Waltham, MA, USA) equipped with Thermo INSIGHT software (latest version 2.5). The opacity of the film O (A/mm) was calculated using the formula [22]:

$$O = \frac{A_{600}}{l}$$

where:

A_{600} —absorbance at 600 (nm) and l —film thickness (mm).

2.7. Water Contact Angle

The wettability analysis was performed using the water droplet method for the measurement of the water contact angle using an OCA 25 Data goniometer (DataPhysics Instruments GmbH, Filderstadt, Germany). The contact angle was measured after applying a 10 μ L drop of distilled water at a rate of 10 μ L/s. The analysis was carried out in a minimum of 6 replicates, and the results were prepared using SCA software (latest version 4.1).

2.8. Water Vapor Sorption Isotherms

The water adsorption isotherms of the films were determined using a dynamic vapor sorption analyzer AQUADYNE DVS-2HT (Quantachrome Instruments by Anton Paar Sp. z o.o., Warsaw, Poland). Film samples of 20 ± 1 mg were loaded in a sample glass pan and exposed to a series of relative humidity (RH): 0%, 10%, 20%, 30%, 40%, 50%, 60%, and 75%, respectively, until the specimen reached equilibrium at 25 °C. The equilibrium criterion at each RH was the percentage rate of change in mass with time (dm/dt) $\leq 0.002\%$ min^{−1} within 10 min. Experimental data points were analyzed using Microsoft Excel 2019 together with aquaWIN Software (latest version Air3).

2.9. Mechanical Properties

2.9.1. Tensile Strength

Mechanical resistance as the tensile strength of the forms was analyzed in 6 replicates using a TA-XT2i texturometer equipped with the Texture Expert software (latest version 2.64) using the ASTM standard method D882-02 [23]. Films measuring 25×100 mm were placed between two measuring jaws spaced at a fixed width of 25 mm with a speed of 1 m/s. The following formula was used to calculate the tensile strength:

$$TS = \frac{M_S}{A}$$

where:

TS—tensile strength (MPa); M_S —maximum force at tensile (N); and A —initial cross-sectional area of the film (mm²).

2.9.2. Elongation at Break

Mechanical elasticity in the form of elongation at break was performed in 6 replicates using a TA-XT2i texturometer equipped with Texture Expert software. Films measuring 25×100 mm were placed between two measuring jaws spaced at a fixed width of 25 mm.

The measurement consisted of pulling the jaws apart at a speed of 1 m/s. The following formula was used to calculate the elongation at break (E):

$$E = \frac{\Delta L}{L} \cdot 100\%$$

where:

ΔL —elongation of the distance at rupture (mm) and L —the initial distance between handles (mm).

2.10. Microstructure

The microstructure of the film surface and cross sections were observed at a magnification of $1000\times$ using a scanning electron microscope (TM3000 Table Microscope (Hitachi High Tech, Tokyo, Japan)). Films measuring approximately $10\text{ mm} \times 10\text{ mm}$ were fixed on the metallic cylindrical using carbon paste PELCO with a diameter of 9 mm (Pik Instruments Sp. z o.o., Piaseczno, Poland).

2.11. Statistical Analysis

Statistical analyses were performed using Statistica 13 (StatSoft Inc., Tulsa, OK, USA). A one-way analysis of variance (ANOVA) with Tukey's test was used to determine the difference between the film samples at a 0.05 significance level. Mean values and standard deviations were calculated using Microsoft Excel 2019.

3. Results and Discussion

3.1. Film Characterization

The apple pectin and fruit pomace powders from apples and blackcurrants were used as raw materials for film formation (Figure 1). The fruit powders did not form a continuous film structure due to the low content of pectin and probably the very low fragmentation. Therefore, apple pectin was used as a gelling agent as it allowed for obtaining uniform structures. A similar strategy was used by Mikus et al. [14], who added soy protein isolate after determining that using oilseed flours as packaging film components did not form a continuous structure. The use of apple pectin was due to its good emulsifying properties and high gelling ability. Gels made with apple pectin are characterized by good hardness, gumminess, cohesiveness, elasticity, and adhesion [24]. The observations from the preliminary studies showed that adding too little pectin (in the range of 1%–4%) prolonged the drying time of the films and made it difficult to remove the films from the Petri dishes after the drying process was completed. On the other hand, higher concentrations of pectin (6%–10%) caused the mixtures to gel quickly, making it impossible to produce thin films with a uniform structure.



Figure 1. Raw materials used for film preparation: apple pectin and fruit pomace powders from apples (AP) and blackcurrants (BC).

Aqueous film-forming solutions were prepared without the addition of fruit pomace (control), which was slightly yellow in color, and with fruit pomace powders, as shown in Figure 2. In the preliminary studies, different concentrations of fruit pomace were used ranging from 10% to 50% for apple pectin, but the most suitable results, such as good miscibility and compatibility of ingredients followed by appropriate density, were achieved with the addition of fruit pomace at the concentration of 10% (2.5 g). After heating and stirring, the film-forming solutions remained liquid, which made it possible to measure a constant volume of the mixture before pouring it into the Petri dishes. Adding larger amounts of fruit pomace (from 15% to 30%) made it impossible to measure a constant volume of the solution, and the films produced were too brittle and characterized by inhomogeneity. This was attributed to the excessive density of the solutions and the rapid crosslinking of the mixture's molecules. Films to which a reduced amount of plasticizer was added in an amount less than 50% relative to pectin showed an increase in brittleness and deterioration in the mechanical properties. As a consequence, the resulting films were not very flexible and suitable for analyses. The addition of plasticizer is an important step, due to the reduction in the interactions between polymer chains and the increase in flexibility of the produced films [25].

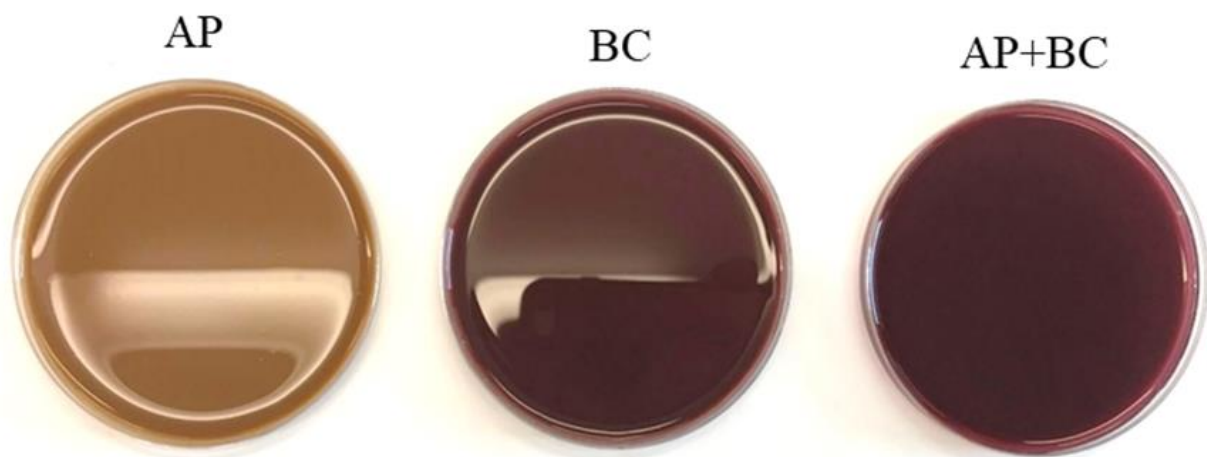


Figure 2. Photographs showing the film-forming solutions containing apple pomace (AP), blackcurrant pomace (BC), and a mixture of both powders (AP+BC).

The color of the mixtures produced depended on the color of the substances present in the pomace used. Therefore, mixtures with apple pomace were characterized by a significantly lighter color compared to the color of mixtures containing blackcurrant pomace and the combination of both types of fruit pomace. The color of the film-forming solution was similar to that of the films that were obtained after drying. The control pectin films produced and those containing fruit pomace are shown in Figure 3. The film produced without the addition of fruit pomace was thin and flexible and had a yellow tint, characteristic of apple pectin. The surface of the control film was smooth, homogeneous, and glossy, with no visible cracks or pores, characteristic of pectin films, which are often the base material for creating various types of films with functional properties [6]. Therefore, the introduction of fruit pomace affected the achievement of a similar structure and character of the films; however, the presence of both types of pomace and their combination affected the achievement of a slightly rough film surface. However, a continuous structure was obtained for all the films tested.

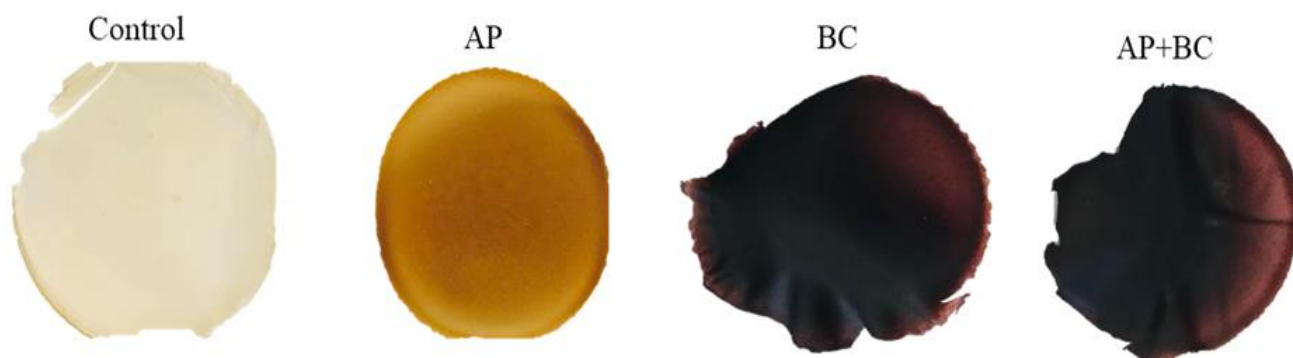


Figure 3. Photographs showing the obtained control films and those prepared with the addition of apple pomace (AP), blackcurrant pomace (BC), and a mixture of both powders (AP+BC).

Comparing the control films and those containing fruit pomace, a visual lack of transparency and differences in thickness were observed (Figure 3). The films with pomace were thicker and more rigid, which affected their susceptibility to deformation and fracture. For all types of films, the surface in contact with the Petri dish was smoother compared to the larger surface. This kind of observation is usually noticed in biopolymeric films by others, and the differences in both sides of the films affect their wettability [26]. The differences that occurred are related to the composition of the raw materials, as well as the drying process itself, i.e., the evaporation of the solvent (distilled water) and the formation of a continuous film structure during this time. In addition, the compatibility of the film components, as well as the interactions in the film-forming solution during preparation, bottling, and drying, may have played an important role [27].

3.2. The Effect of Fruit Pomace Addition on the Thickness of Pectin Films

During film preparation, the thickness was controlled using equal amounts of poured film-forming solutions; however, due to the nature of the raw materials used, differences in film thicknesses were obtained, as shown in Table 1. The thickness values of the tested films were in the range of 0.097–0.107 mm, which is similar to other biopolymer films [19]. The control pectin film had the lowest thickness, while the film containing a combination of apple pomace and blackcurrant pomace had the highest thickness of 0.107 mm. The thickness of a film may be depended on the presence of solids in the polymer matrix. Examples of solids present in the matrix include seeds and stalks [28]. In addition, the type and quantity of phenolic compounds contained increase the thickness of the film coating. It has been observed that the high content of phenolic compounds in blackcurrant pomace affects the thickness of the produced pectic films, which is caused by the formation of bridges by these compounds and a decrease in intermolecular distances [29]. There were no statistically significant differences ($p < 0.05$) between samples of the control films and films containing blackcurrant pomace, probably due to the lower amount of pectin. Films produced with the addition of apple pomace and films produced with the addition of a mixture of apple pomace and blackcurrant pomace were statistically significantly different ($p < 0.05$) from the other film samples.

Table 1. The thickness, water content, swelling index, and water contact angle for the control pectin films and films with the addition of apple pomace (AP), blackcurrant pomace (BC), and a mixture of both powders (AP+BC).

| Film | Thickness (mm) | Water Content (%) | Swelling Index (%) | Water Contact Angle (°) |
|---------|------------------------|---------------------|---------------------|-------------------------|
| Control | 0.097 ± 0.011^a | 1.425 ± 0.035^b | - | 50.69 ± 2.62^a |
| AP | 0.102 ± 0.005^{ab} | 1.305 ± 0.078^a | 117.70 ± 0.08^a | 53.32 ± 3.33^a |

Table 1. Cont.

| Film | Thickness (mm) | Water Content (%) | Swelling Index (%) | Water Contact Angle (°) |
|-------|----------------------------|----------------------------|----------------------------|---------------------------|
| BC | 0.098 ± 0.011 ^a | 1.330 ± 0.014 ^a | 181.13 ± 0.10 ^c | 50.90 ± 9.01 ^a |
| AP+BC | 0.107 ± 0.002 ^b | 1.318 ± 0.039 ^a | 154.17 ± 0.03 ^b | 70.89 ± 4.45 ^b |

Mean values ± standard deviations. Different superscript letters (^{a–c}) within the same column indicate significant differences between the films ($p < 0.05$).

3.3. The Effect of Fruit Pomace on the Water Content of Pectin Films

Research on the sorption properties of biopolymer films is extremely important for explaining their moisture-absorbing capacity. Scientific studies show that these are water vapor-sensitive materials, and their greatest application can be observed in products with low water content [6]. Many studies have focused on modifying the raw material composition by introducing substances that reduce the hygroscopicity of biopolymer films, including fats [19]. The water content of the tested films is shown in Table 1. The values were in the range of 1.305%–1.425%, which is relatively low and characteristic of dried materials. The highest value was observed for the control film, while the lowest value was observed for the film with the addition of apple pomace. The lower water content of films produced from apple pectin and the addition of apple pomace relative to the other samples is due to the content of hydrophilic molecules, which are responsible for binding water molecules via hydrogen bonds to the film material matrix. The reduction in water content in films with the addition of fruit pomace may be due to the high content of phenolic compounds, which limited access to water molecules in the film structure. The less hydrophobic the structure of the film, including the nature of the ingredients from which it is made, the higher the water content of the film [30]. The differences between the control samples and the samples of films with the addition of fruit pomace were statistically significant ($p < 0.05$). The films produced with the addition of fruit pomace were not statistically significantly different ($p < 0.05$). The water content of films produced with the addition of fruit pomace was similar.

3.4. The Effect of Fruit Pomace on the Water Solubility of Pectin Films

The analyzed pectin films of the control and those containing fruit pomace were inserted into bottles of distilled water after drying, and moments after being placed in water, the films began to solubilize and lose their integrity. Figure 4 presents a photo showing the solubility analysis, where the visible change in the color of the distilled water associated with the dissolution of the tested films can be observed. The degradation of the films in water indicates their potential for easy degradation and the degree of release of active compounds from the films. The control films produced without the addition of fruit pomace degraded the fastest when placed in distilled water. This may be due to the good dissolving properties of the control films. Control films with the addition of blackcurrant pomace and apple pomace did not disintegrate immediately after being placed in water; rather, the film samples gradually disintegrated, losing homogeneity. The films produced with the addition of a mixture of blackcurrant and apple pomace water retained their integrity the longest after being placed in water and did not disintegrate. The addition of fruit pomace reduces the ability of the pectin film structure to disintegrate in an aquatic environment. The apparent color change in the distilled water to purple was due to the release of anthocyanins from the fruit pomace. The color of the solution is also influenced by pH. In solutions with a more acidic pH, the color is purple. In solutions with an alkaline pH, the color is blue due to the formation of an echinoid base. In solutions with a pH close to 4.5, the solution is colorless [31].

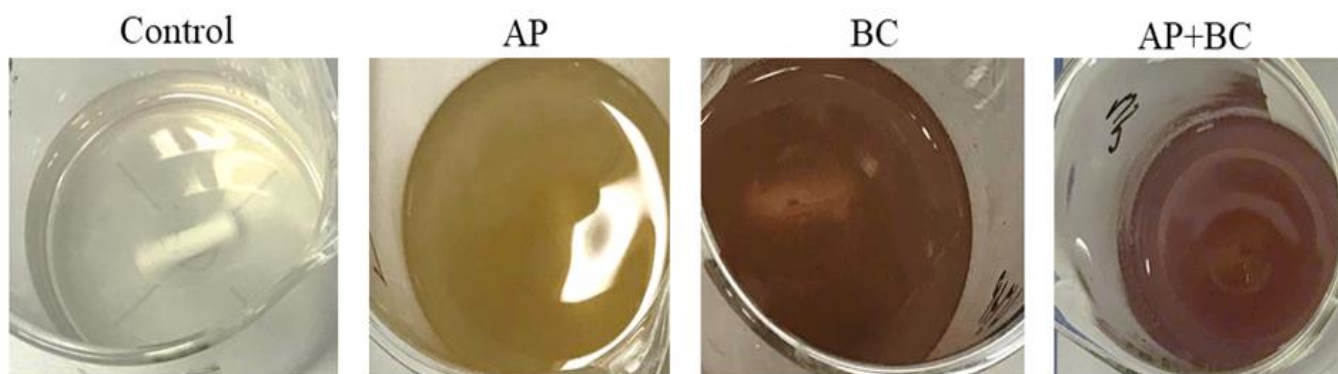


Figure 4. Photographs showing beakers with distilled water after the immersion of the analyzed pectin control films and those prepared with the addition of apple pomace (AP), blackcurrant pomace (BC), and a mixture of both powders (AP+BC).

The solubility of pectin films with and without fruit pomace added varied to different degrees, making it impossible to weigh the samples again. Blackcurrant pomace probably has a higher organic acid content than apple pomace, resulting in a more acidic pH, and thus the color of the resulting solution was purple. This is due to the predominance of the flat vinyl cation form in the resulting solution. The pectin film solution without the addition of fruit pomace had a clear color, indicating the presence of colorless structures (chalcone). Sutharsan et al. [29] pointed out the relationship between the presence of hydroxyl groups in flavonoid molecules, showing that the higher their content, the greater the binding of the film surface to water molecules, thus increasing the solubility of the film. This means that the higher the content of flavonoid molecules in the produced pectin films, the faster the films dissolve in water.

3.5. The Effect of Fruit Pomace on the Swelling Index of Pectin Films

The control film made from apple pectin without the addition of fruit pomace disintegrated when placed in water, making it impossible to weigh it again and determine the swelling index and showing that it has a very good ability to adsorb water above its weight, resulting in solubilization. The highest swelling index in water among the films containing pomace was found for the film containing blackcurrant pomace (181.13%), while the lowest was found for the film with apple pomace (117.70%). A value of 154.17% was obtained for the film produced from a combination of both types of pomace, which reflects the mutual miscibility of the film components. The differences in swelling in water between the shaken film samples were statistically significant ($p < 0.05$). Films produced using the pomace from blackcurrant, due to their highest swelling capacity in water, are most prone to disintegration in a damp environment. The addition of fruit pomace influenced the reduction in water absorption and retention in the film structure, which affects the better mechanical properties of the film. The addition of fruit pomace can affect the thickening properties and improve the structure of the produced films due to the presence of dietary fiber in the pomace.

3.6. The Effect of Fruit Pomace on the Optical Properties of Pectin Films

To estimate the color of the produced pectin-based films, the parameters L^* , a^* , and b^* were measured, and for a better interpretation of the results, the total color difference between the film and the white standard (ΔE) was calculated. Table 2 shows the results of color performance measurement for the analyzed pectin films. The parameter L^* denotes lightness and is measured from black to white, the parameter a^* is measured from green to red, and the parameter b^* is measured from blue to yellow [32]. The total color difference with which the difference between the test material and the reference can be quantitatively shown indicates the difference between a given color and a specific standard.

Table 2. Color parameters L^* , a^* , and b^* and the total color difference (ΔE) for the control pectin films and films with the addition of apple pomace (AP), blackcurrant pomace (BC), and a mixture of both powders (AP+BC).

| Film | L^* | a^* | b^* | ΔE | Film Opacity (A/mm) |
|---------|--------------------|----------------------|----------------------|--------------------|---------------------|
| Control | 87.24 ± 0.68^d | $(-1.15) \pm 0.04^a$ | 13.75 ± 1.90^c | 14.05 ± 2.01^a | 1.03 ± 0.18^a |
| AP | 62.22 ± 3.89^c | 9.78 ± 2.13^c | 39.08 ± 1.56^d | 49.95 ± 3.92^b | 10.64 ± 0.56^b |
| BC | 28.69 ± 3.69^b | 13.51 ± 3.18^d | 7.12 ± 1.95^b | 65.62 ± 2.69^c | 17.14 ± 0.87^c |
| AP+BC | 21.09 ± 0.70^a | 2.44 ± 0.87^b | $(-0.03) \pm 0.21^a$ | 71.34 ± 0.67^d | 14.79 ± 3.62^c |

Mean values \pm standard deviations. Different superscript letters (^{a–d}) within the same column indicate significant differences between the films ($p < 0.05$).

The higher the parameter, the greater the color discrepancy, while the lower the parameter, the closer the color is to the reference standard [33]. The control film had the highest value of the L^* parameter (87.24), meaning it was the lightest. In addition, the a^* and b^* parameters for this film relate to the green–yellow tones of the film. The lowest value of the L^* parameter was found for a pectin film produced with the addition of blackcurrant pomace (21.06). This film had the lowest lightness coefficient, meaning that its color was darkest relative to the other comparative films. The color of the film produced with the addition of blackcurrant pomace affected the color of the film, resulting in shades of green and blue. The addition of apple pomace during the manufacturing of the pectin films had the least effect on the L^* parameter. These films remained bright; however, a significant reduction in brightness was observed (62.22), and the a^* and b^* parameters indicated a color with red–yellow shades. Films produced with the addition of a mixture of apple and blackcurrant pomace were dark; the value of the L^* parameter indicated similarity concerning films produced with the addition of blackcurrant pomace (28.69), and these films were comparatively dark. The a^* and b^* parameters of these films indicate dominant shades of red and green.

The total color difference (ΔE) for the test films was in the spectrum of 14.05–65.62 and showed significant color differences between the sample films and the standard. This parameter can be interpreted using the following indications: ΔE less than 1 indicates no discernible difference in color, ΔE between 1 and 2 indicates a difference in color discernible by a person properly trained to do so, while ΔE between 2 and 3.5 indicates a discernible difference in color by a person not trained to do so. A value of ΔE above 5 indicates a significant color difference [34]. The most similar films in terms of color were pectin films with the addition of blackcurrant pomace and pectin films with the addition of a mixture of blackcurrant pomace and apple pomace. The ΔE value was lower for the film produced with the addition of a mixture of blackcurrant pomace and apple pomace. The most deviating film in terms of color difference was the film produced without the addition of fruit pomace (14.05). The color differences between the samples were statistically significant ($p < 0.05$). The addition of apple pomace affected the color difference between the film produced with apple pomace and the film produced without pomace.

Opacity is the inverse of translucency and can be understood as a parameter describing opacity and the degree of light retention by a given coating. Table 2 shows the opacity values for different types of pectin films with and without apple pomace and blackcurrant pomace. The opacity at 600 nm assumed a range from 1.030 to 17.14 A/mm. Films produced without the addition of fruit pomace had the lowest opacity (1.030 A/mm) and were the most transparent compared with the other produced films. In contrast, films containing blackcurrant pomace had the highest opacity (17.14 A/mm). Films produced with the addition of apple pomace and blackcurrant pomace had a higher opacity value. These films had an intense and uniform red–purple color. The opacity of the films increased with the addition of fruit pomace, which may be related to the presence of color compounds, including flavonoid compounds and anthocyanins. The higher the concentration of color

compounds in fruit pomace, the higher the opacity of the resulting films, which means higher the opacity of the coatings. The opacity of the films is also affected by the thickness of the film and the degree of distribution of the fruit pomace and other components of the mixture. The formation of thickening due to poor mixing of the solution and the presence of impurities in the pomace can distort the obtained results. The differences between the produced film samples were statistically significant ($p < 0.05$), except for films produced with the addition of blackcurrant pomace and films produced with the addition of a mixture of apple pomace and blackcurrant pomace.

3.7. The Effect of the Fruit Pomace on the Water Contact Angle of Pectin Films

The degree of wettability of the surface can be determined by analyzing the contact angle of the applied water droplet, which indicates the character of the surface as being either hydrophobic or hydrophilic. The higher the angle, the higher the hydrophobicity of the film [35]. Figure 5 shows photographs of applied water droplets on the tested pectin films. It was observed that the greatest difference in the shape of the water droplets was obtained for the film containing a combination of apple and blackcurrant pomace.

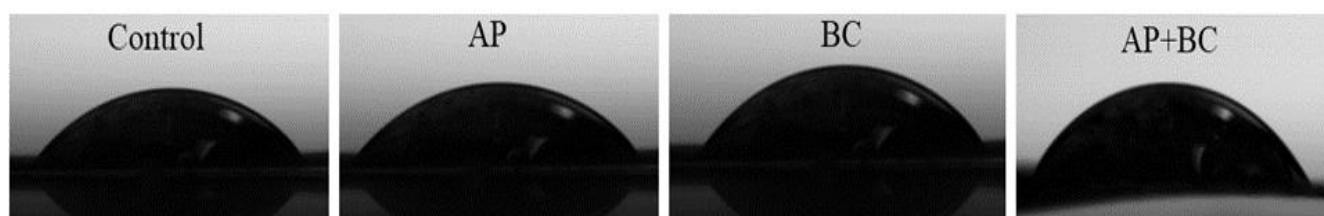


Figure 5. The shape of the initial water droplet deposited to the control pectin films and those with the addition of apple pomace (AP), blackcurrant pomace (BC), and a mixture of both powders (AP+BC).

Analyzing the obtained results for the contact angle of the tested pectin films, which are shown in Table 1, it can be observed that the addition of fruit pomace increased hydrophobicity. The results obtained ranged from 50.69° to 70.89° . The statistical analysis showed that the addition of blackcurrant pomace resulted in statistically significant differences in the wettability of the films relative to the other samples ($p < 0.05$). For these films, the angle value was the highest (70.89°), while the control film had the lowest value (50.69°). The film produced with the addition of apple pomace had a significantly lower wettability angle compared to the film produced with the addition of blackcurrant pomace. The above may be indicative of the hydrophobic structure of the film produced with the addition of blackcurrant pomace. The type and amount of fruit pomace added to pectin films can affect the degree of repulsion of water particles from the film surface. When the two types of pomace were combined, the wetting angle was lower than for films produced without combining them. In addition, the contact angle can be influenced by homogeneity in the structure of the tested films, the presence of irregularities on the film surface, the presence and size of impurities such as stalks, leaves, and stems, as well as the degree of fineness of the fruit pomace. Films produced with blackcurrant pomace had the highest hydrophobicity, while the other films had lower hydrophobicity at a similar level.

3.8. The Effect of Fruit Pomace Addition on the Sorption Isotherms of Pectin Films

Figure 6 shows the water vapor sorption isotherm plots for the analyzed films, where the influence of the fruit pomace addition on the mass change can be observed. All curves have a similar shape, as it is common for dry food to absorb a relatively small amount of water at low and a large amount of water at high water activity. This shape of isotherms is characteristic of most biopolymer materials based on polysaccharides [36]. The control films showed the highest mass changes in comparison with the films containing fruit pomaces. The most notable differences were observed at higher water activity values, which is also related to easier mobility of water molecules when the material becomes softer.

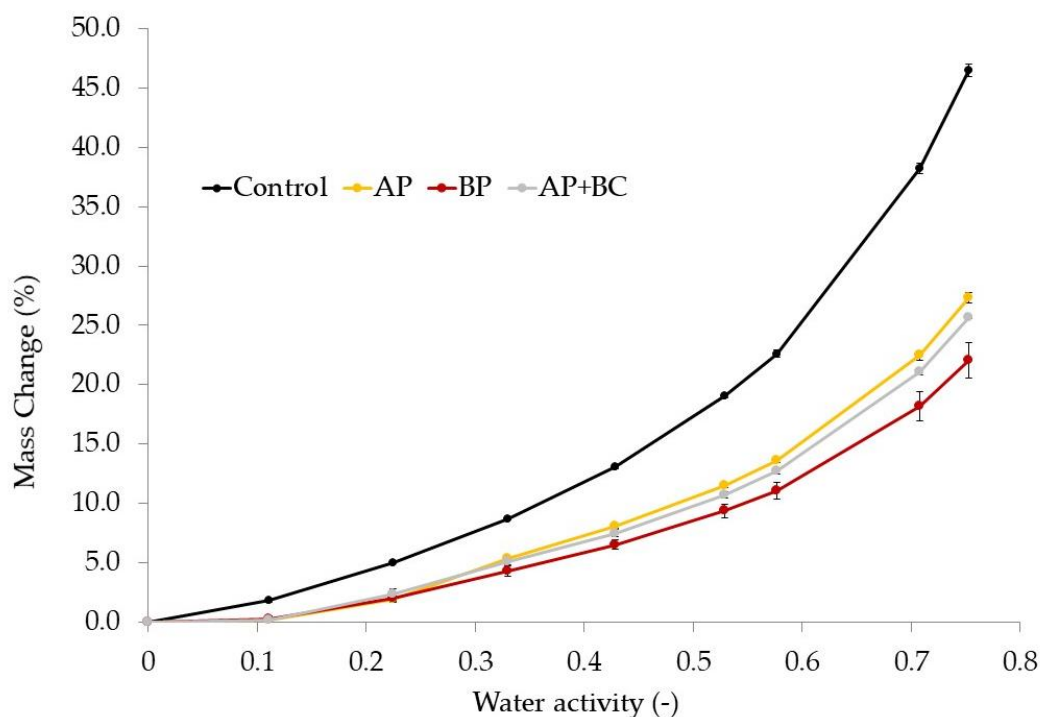


Figure 6. Water vapor sorption isotherms for the control pectin films with the addition of apple pomace (AP), blackcurrant pomace (BC), and a mixture of both powders (AP+BC).

In addition, the control pectin films had the lowest film thickness (Table 1), which could induce higher water vapor adsorption. Taking into account all films containing fruit pomace, those with apple pomace showed higher mass changes, and those with blackcurrant pomace showed the lowest mass change values. The mixture of both fruit pomaces affected the modified adsorption rate, which was similar to apple pomace-containing films. This can be attributed to many factors, such as water content, film thickness, and microstructure. However, a study conducted by Reißner [37] showed that the structure and chemical composition had an impact on the water vapor sorption properties. Their samples consisted of black and redcurrant, chokeberry, gooseberry, and rowan pomace, and the results showed that the chokeberry pomace, which contained the most carbohydrates, had the best hygroscopic properties.

3.9. The Effect of Fruit Pomace on the Mechanical Properties of Pectin Films

To determine the mechanical strength of the tested pectin films, the tensile strength and elongation at the break of the obtained films were investigated. The results are shown in Table 3. The breaking strength of the produced pectin films ranged from 2.97 MPa for the control films to 6.72 MPa for films containing a combination of apple pomace and blackcurrant pomace. This parameter determines the tensile stress at which the film breaks [38]. The introduction of fruit pomace increased the mechanical strength of the pectin films. The greatest effect was achieved with a combination of apple and blackcurrant pomace and with apple pomace alone. The increase in breaking strength was inversely proportional to the elasticity of the pectin films. Statistical analysis of the films indicated statistically significant differences in the parameter studied ($p < 0.05$).

A significant reduction in elongation at break was observed for films with the addition of fruit pomace compared with the control films (10.16%) (Table 3). Elongation at break is a parameter that determines the increase in film area that occurred during stretching until breakage. The lowest values were obtained for films containing apple pomace (5.11%), but in general, films with the addition of fruit pomace had similar elongation values ranging from 5.11% to 6.07%. The lower elongation gain during stretching may be influenced by the presence of impurities in the films, film thickness, and elasticity. The presence of

contaminants in the film can degrade the uniformity of the film structure and affect film brittleness and breakage. The obtained values for elongation at break were relatively low compared to other biopolymer films [39].

Table 3. Tensile strength and elongation at break for the control pectin films and films with the addition of apple pomace (AP), blackcurrant pomace (BC), and a mixture of both powders (AP+BC).

| Film | Tensile Strength (MPa) | Elongation at Break (%) |
|---------|------------------------|-------------------------|
| Control | 2.97 ± 1.19^a | 10.16 ± 2.02^b |
| AP | 5.71 ± 1.71^{bc} | 5.11 ± 1.22^a |
| BC | 3.11 ± 1.41^a | 6.07 ± 1.45^a |
| AP+BC | 6.72 ± 1.14^c | 5.76 ± 1.35^a |

Mean values \pm standard deviations. Different superscript letters (^{a-c}) within the same column indicate significant differences between the films ($p < 0.05$).

3.10. The Effect of the Fruit Pomace on the Microstructure of Pectin Films

The structures of the film surface and cross-sections of the analyzed films with and without the addition of fruit pomace were investigated using a scanning electron microscope. During the observations, attention was paid to changes in the surface of the films resulting from the addition of fruit pomace, as well as the uniformity and smoothness of the surface of the produced films. The surface structures of the tested films are shown in Figure 7, where the apparent difference between the films can be observed. The surface of the control films produced with pectin without the addition of fruit pomace was smooth and devoid of pores and roughness. This was due to the even distribution of the ingredients in the film-forming mixture and the low amount of impurities. The lack of pores and smooth surfaces favorably affected the barrier and mechanical properties of the produced films. The structure of films produced with the addition of pomace was rougher and more porous.

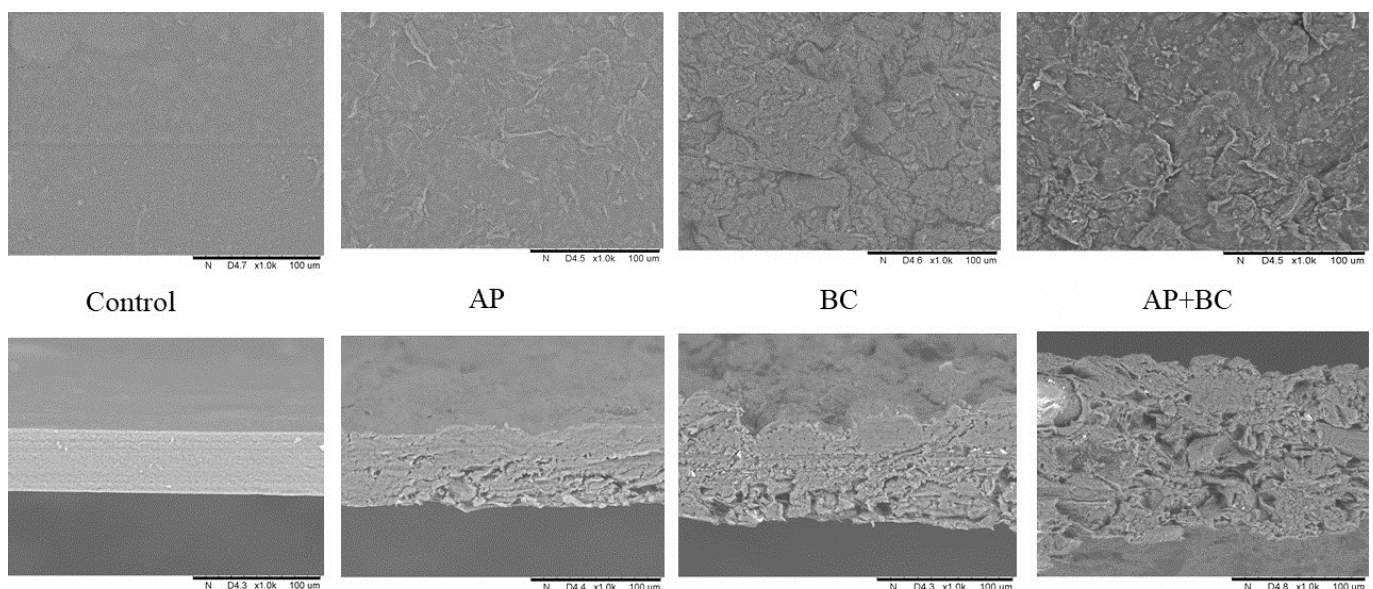


Figure 7. The film surface (**upper**) and cross-section (**lower**) structure of the control pectin films and those with the addition of apple pomace (AP), blackcurrant pomace (BC), and a mixture of both powders (AP+BC). Magnification 1000 \times .

The addition of apple pomace made the resulting films crisper, slightly wrinkled, and more porous. In addition, they had a more stiff and less flexible structure compared with the control films. The structure of pectin films produced with the addition of blackcurrant pomace had a much higher amount of roughness compared to control films and pectin films produced with the addition of apple pomace. The film had an uneven, wrinkled surface. Free spaces could be observed in the film, which could have been formed due to uneven mixing of the mixture components and due to the presence of impurities from fruit pomace. At the same time, the rough surface of the produced films may depend on the degree of grinding of the fruit pomace. Pectin films produced with the addition of a mixture of blackcurrant pomace and apple pomace had a surface texture similar to the films produced with the addition of blackcurrant pomace from blackcurrants. The addition of fruit pomace affects the structure of the produced films. Films with the addition of pomace are uneven and wrinkled with visible impurities. This above may affect the textural properties of the produced films and have important implications for other performance characteristics, which should be taken into account for practical applications.

The cross-sections of the tested pectin films without the addition of fruit pomace and films produced with the addition of blackcurrant and apple pomace indicated that the film surface varied depending on the fruit pomace used. The control film had a smooth surface, and its cross-section showed the absence of cracks and damage to its internal structure. The pectin film produced with the addition of apple pomace was smooth, and its cross-section showed small cracks. The film produced with the addition of blackcurrant pomace had a larger number of pores and cracks compared with the film produced with the addition of apple pomace; however, it was also noticeably thicker than the film produced with the addition of apple pomace. The pectin film produced with the addition of a mixture of pomace from blackcurrants and apple pomace was the thickest, and its cross-section showed the presence of numerous cracks and corrugations. The surface of pectin films without the addition of blackcurrant pomace and apple pomace was the smoothest and thinnest. The film produced with the addition of apple pomace had the smoothest surface compared with the films produced with the addition of blackcurrant pomace. The surface of the film with the addition of the mixture of blackcurrant pomace and apple pomace had an irregular, rough surface, which could be due to uneven dissolution and mixing of the mixture components [40].

4. Conclusions

The use of apple and blackcurrant pomace powders, as well as the mixture of both types of fruit pomace, made it possible to produce pectin films with different physical properties that can be evaluated for use in food packaging. The addition of fruit pomace significantly increased the thickness and the mechanical strength of pectin films, while water vapor adsorption was reduced. All the films analyzed showed very good solubility in water, indicating the potential for good degradation of materials in aqueous conditions. The introduction of fruit pomace into the matrix of pectin films reduced the water content, as well as swelling in water. The structure of the analyzed pectin films varied and depended on the presence of fruit pomace. However, more research is needed to analyze the thermal stability and the other functional capacities during the practical applications of the analyzed films as packaging components or trays to carry different food products.

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