Research Square

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Fabrication and characterization of cellulose acetate film containing falcaria vulgaris extract

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Abstract

Nowadays, the use of packaging materials based on natural polymers reduces the use of synthetic materials that cause environmental pollution. For this reason, in the present study, the possibility of preparing biodegradable films from cellulose acetate incorporated with Falcaria vulgaris extract was investigated. Cellulose acetate films were produced by casting method and Falcaria vulgaris extract was added in concentrations of 0.1, 0.2 and 0.3%. Mechanical properties, permeability to water vapor and oxygen, solubility, color, opacity, contact angle, morphology, antioxidant and antimicrobial properties of cellulose acetate films were evaluated. The results showed that the tensile strength, modulus of elasticity, vapor permeability, water absorption, solubility, swelling percentage and percentage of water content of the resulting films decreased with the increase of Falcaria vulgaris extract concentration. The contact angle percentage of cellulose acetate film and cellulose acetate film with 0.3% extract were 59.32 ± 0.97, 65.47 ± 1.16, respectively. Cellulose acetate films containing Falcaria vulgaris extract showed good antioxidant properties, namely the amount of DPPH radical trapping of cellulose acetate film with 0.3% extract equals 35.31% compared to cellulose acetate film (0.15%), which showed a significant increase. Furthermore, the produced films showed good antimicrobial properties against *Escherichia coli*, *Staphylococcus aureus*, *Candida albicans* and Candida glabrata. In general, this study showed that cellulose acetate film containing extract could be used for food packaging that increasing the shelf life of food.

Introduction

In the last decade, the tendency of consumers towards food products with better quality and fresher has increased. Microbial growth as well as oxidative and chemical changes are the most important factors reducing the quality of food products during storage (Roy & Rhim, 2020). The most important method used to deal with spoilage and pathogenic factors in food is to use chemical preservatives. Despite several types of approved and commercial antimicrobials, their use faces limitations. In this respect, it is now known that some cancers may be related to chemical preservatives and their toxic residues (Assis et al., 2020). All these cases have caused the awareness of consumers and their demand for foods preserved with natural ingredients to be increased, and researchers are looking for new natural and non-chemical preservative compounds and methods in food. One of the groups of natural antimicrobial substances of medicinal plants are essential oils and plant extracts. In addition to being harmless, these compounds have shown a wide range of antimicrobial and antioxidant effects in various studies (Meshkani et al., 2013). Therefore, essential oils and plant extracts can be used as an alternative method to control pathogenic and spoilage factors and increase the shelf life of food (Adilah et al., 2018). The direct use of these compounds in the food itself faces some limitations, so researchers are looking for ways to improve their activity. One of the methods to use these natural materials in food packaging and coating is called active packaging. These biocompatible coatings based on edible films, which are mainly made of polysaccharides, proteins, fats or a combination of them, are of special importance day after

day due to their natural materials, renewable ability and non-environmental pollution (Hanani et al., 2019).

In recent years, essential oils and plant extracts have been widely used as additives in biodegradable edible films. Due to their fatty nature, these compounds are expected to reduce vapor permeability in hydrophilic films. It has been proven that these materials can affect some properties of edible films and coatings in addition to providing antioxidant and antimicrobial properties. In this regard, in a research, red cabbage extract was added to cellulose acetate films and their thermal, physical, permeability, color, and microscopic properties were investigated. The light permeability of the edible film with red cabbage extract increased and the mechanical properties of the edible film changed due to the softening properties of the extract. The appearance became non-uniform and disordered because of the extract, and this problem increased with the increase in the concentration of the extract (Freitas et al., 2020). Gonçalves et al. (2020) investigated the functional and antimicrobial properties of cellulose acetate films containing fennel essential oil and different amounts of plasticizer (5, 10, 20 and 30% glycerol). The results showed that increasing the amount of plasticizer increased the thickness, water vapor transfer rate and tensile strength. In another research, the essential oil of cinnamon and oregano was added to cellulose acetate film and the improvement of antibacterial activity and mechanical properties of the film was resulted (Zhang et al., 2017). Cellulose acetate is a polymer with various characteristics such as strength, biodegradability, suitable separation as a membrane and a relatively low price, is very hydrophilic, and is often used to reduce sedimentation in membranes. This polymer is used to prepare food packages and in Pharmaceuticals have been used to prepare tablets and coating materials to control drug release. Moreover, this type of polymer is widely used for preparing films and membranes (Carvalho et al., 2017, Abedini et al., 2011).

Falcaria vulgaris (locally called as ghazyaghi/poghazeh), is a member of the Apiaceae family, which is known as a weed in fields and grows wildly near agricultural lands. This plant is used in some mountainous areas of Iran in the spring as a vegetable and flavoring in all kinds of foods (Khazaei and Salehi, 2006). The presence of compounds such as spatolnol, carvacrol and alphafin as well as compounds with antioxidant properties such as anthraquinone, alkaloids, phenol, tannin, saponin, steroids and flavonoids in this plant has given it unique medicinal properties. In traditional medicine, Falcaria vulgaris is used as a treatment for skin wounds, stomach problems, liver disease, and blood purification (Jaberian et al., 2013). The objective of this article is to produce biodegradable films and investigate the physicochemical properties of cellulose acetate film incorporated with Falcaria vulgaris extract.

Materials And Methods

Preparation of Falcaria vulgaris extract

In the process of preparation for extraction, the aerial part of Falcaria vulgaris was ground after initial cleaning (3–5 minutes) and passed through a mobile sieve system with a hole size of 0.4 mm (Taylor,

USA) and mixed with solvent at a ratio of 1:10. 50 ml of Falcaria vulgaris and solvent mixture was placed in the treatment chamber of the Ohmic heating system, and then by placing the electrodes, the extraction process was carried out depending on the treatment in a certain voltage gradient, temperature and specific extraction time. In this method, four variables including voltage gradient (10, 15, 20, 25 and 30 V/m), extraction time (10, 15, 20, 25 and 30 minutes), extraction temperature (30, 37.5, 45, 5 52 and 60°C) and the solvent ratio of water to ethanol (0, 25, 50 and 100%) were investigated. After extracting, the solvent was separated from the plant using Whatman No. 1 filter paper, and the final extract was concentrated using a rotary evaporator at a temperature of 40°C and a rotation speed of 95 rpm. After that, the extract was placed in a 40°C oven until it reached a constant weight. The final extract was poured into a dark, airtight glass vial and kept at 4°C until the tests were performed (Tunç and Koca 2019).

Preparation Of Cellulose Acetate Film

Cellulose acetate film was prepared using casting method. Cellulose acetate in acetone with a weight percentage of 3%, the extract with dilutions of 0, 2, 4 and 6% and a fixed amount of glycerol of 0.325 grams as a plasticizer were poured and sealed inside the capped bottles and mixed for 5 minutes at 1500 rpm by a homogenizer. Then the mixture was molded on glass plates in sterile conditions for 5 ml and finally the films were separated from the casting plates. Before performing analysis and additional tests, for a period of 48 hours at a temperature of 25°C and a relative humidity of 40%, it was placed in a desiccator containing saturated solutions of magnesium nitrate until the moisture balance was reached. The films were kept at a temperature of 4°C. As a control sample, the film was prepared by the above method without the presence of extract (Fiorntini et al., 2017).

Evaluation Of Film

Mechanical properties

ASTM D882 standard method was used to obtain the actual stress-strain curves. The elastic modulus (EM), tensile strength (TS) and elongation at break (EB) of the films were calculated from these curves. The mechanical properties of the film were determined through a tensile test using a texture tester (STM5, Iran). Rectangular samples (10 mm x 100 mm) were cut from the film and equilibrated for 48 h at 51% relative humidity using a saturated magnesium nitrate solution. Then the film was placed between two clamps of the device and they were stretched at a speed of 50 mm per minute. The percentage of elongation to tear was defined as the percentage change in the sample length to the initial length between two clamps. Tensile strength was calculated through the following formula (Rezaee et al. 2018).

$$\mathrm{Tensilestrength\ (TS)} = \frac{themaximumforceattearmoment}{filmwidth \times filmthickness}$$

1

$$\mathrm{Elasticmodulus} = \frac{\mathrm{tension}}{\mathrm{strain}}$$

2

$$strain = \frac{Finallength - Initiallengthbeforestretching}{\mathrm{Initiallengthbeforestretching}}$$

3

Color

The color analysis of the films was measured using a manual colorimeter, and the color characteristics and color values were measured based on L* index or brightness (0 equals black and 100 equals white), a* index (-60 equals green and + 60 equals red) and b* index (-60 equals blue and + 60 equals yellow). The device was calibrated using a standard tile (L*= 94.56, a* =-1/575 and b*= -3/428). All measurements were calculated considering three points and in five repetitions. Total color difference (ΔE) and whiteness index (WI) were calculated from equations 4 and 5, respectively (Rodrigues et al., 2018).

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

4

$$WI = 100 - \sqrt{\left(100 - \mathrm{L}^{*}\right)^{2} + \left(\mathrm{a}^{*}\right)^{2} + \left(\mathrm{b}^{*}\right)^{2}}$$

5

Light Transmission And Transparency

The light transmission of the film was measured with a spectrophotometer (SPECORD 250, Germany) at a wavelength between 200 and 600 nm, and a wavelength of 560 nm was used to calculate the transparency of the film. Rectangular film samples (20 mm x 40 mm) were placed inside a desiccator containing magnesium nitrate (51% relative humidity) at room temperature. Afterward, the thickness of the film samples was measured and the film samples were attached to the tube with transparent tape and placed inside the device to read and record the number. An empty cuvette was used as a reference (Karami et al., 2014).

$$opacity = \frac{\mathrm{Theabsorptionrateofwavelength600nm}}{\mathrm{Filmthickness}}$$

6
Thickness

The thickness of the film was measured with a digital micrometer (country model) with an accuracy of 0.01 mm in ten repetitions, and the average of at least 10 different measurements was calculated (Rezaee et al. 2018).

Vapor Permeability

5 grams of calcium chloride without water was poured into a certain container (small cup) and then the surface of the container was covered with a film using melted paraffin. In this way, 0% humidity was created inside the container. The small cups were placed in a desiccator containing saturated sodium chloride solution. A saturated solution of sodium chloride at 25°C created a humidity of 75%. The difference in humidity on both sides of the film at a temperature of 25°C created a vapor pressure gradient equal to 1753.55 Pa. The change in the weight of the container was recorded using a digital scale (LS 120A scs, Switzerland) with an accuracy of 0.0001 and its graph was drawn as a function of time. A container without sodium chloride, covered by film, was considered as a control (Alizadeh, et al. 2017).

$$\mathrm{watervaporpermeability}\,(WVP) = \frac{y * WVTR}{(P_1 - P_2)}$$

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$$WVTR = \frac{\mathrm{Theslopeofthemoisturepenetrationcurve}}{\mathrm{Filmsurfacearea}}$$

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The water vapor permeability was calculated from Eq. 7, where y is the average film thickness in meters, WVTR is the rate of weight loss (Eq. 8), and P_1-P_2 is the partial pressure gradient of water vapor on both sides of the film at 25°C (equal to 179 3.0 kPa).

Solubility

To determine the solubility, first, a 2 cm square area of each film sample was cut and placed in an oven (K.M.85, Iran) at a temperature of 100 ± 5°C for 24 hours to be dried and reached a constant weight. The obtained weight is known as initial dry weight (W_0). Then, the samples of the films were immersed in 50 ml of distilled water and periodically stirred. After 24 hours of storage at a temperature of 23 ± 2°C, they were dried again at a temperature of 100 ± 5°C for 24 hours to obtain the final dry weight (W_f). The solubility of films was calculated using Eq. 9 (Rodrigues et al., 2018).

$$\text{Solubility}\ (\%) = \frac{w_o - w_f}{w_o} \times 100 \quad (9)$$

Contact Angle

The Sessile drop method was used to measure the contact angle of the films. For this purpose, 2 microliters of double distilled water was placed on the film sample using a sampler (precision syringe). This method is based on image processing and curve fitting to calculate the contact angle. For this purpose, imaging was done with the help of a digital microscope Dinolight (Edge 3-Taiwan model) in controlled light conditions and at an early time, and the contact angle was done with the help of the droplet analysis section (contact angle) of image j software (version v52/1). At least five measurements were taken in each film and the contact angle on both sides of the drop was measured. The average value was used as the contact angle. All the tests were performed at an ambient temperature of 25 ± 2°C and a relative humidity of 50 ± 1% (Chiaoprakobkij et al., 2019).

Moisture Content

First, a surface of 2 2 cm was cut from each film sample and their weights were recorded separately as the initial weight (W_0). Then, the cut films were placed in an oven at 100 ± 5°C for 24 hours to be completely dried and reached a constant weight. After removing the films from the oven and placing them in the desiccator, the obtained weights were recorded as the final weight (W_f) each. Finally, the moisture content (MC) of the films was calculated using Eq. 10 (Wu et al., 2017).

$$\text{Moisturecontent}\ (\text{MC}) = \frac{W_0 - W_f}{W_0} \times 100$$

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Swelling

To determine swelling, first, a 2 2 cm surface was cut from each film sample and their weights were measured. Then the cut films were placed in an oven at 100 ± 5°C for 24 hours to be dried and reached a constant weight. The obtained weights were recorded as initial dry weight (W_0). Then, the film samples were immersed in 30 ml of distilled water, and at specific time intervals every 50 minutes, the films were removed from the distilled water, and after removing the surface water, it was weighed (W_f), and this process continued until the films reached a constant weight (Türe 2019).

$$\text{Sweling}\ (\%) = \frac{W_0 - W_f}{W_0} \times 100$$

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Oxygen Permeability

The oxygen permeability of the films was measured at a temperature of 23 and a relative humidity of 0% using an oxygen permeability measurement device (Mocon, Germany). The films were purged with nitrogen for 16 hours and then subjected to oxygen flow at a rate of 10 mL/min. The oxygen permeability coefficient is proportional to the amount of oxygen passing through the thickness. And the amount of oxygen passage was calculated from eauation 12 (Rajeswari et al., 2020).

$$\text{Oxigen tranmision rate (OTR) (\%)} = \frac{Dry\ film\ weight - Wet\ film\ weight}{Wet\ film\ weight} \times 100$$

12
Degradability Test

The moisture content of films with regular shapes was taken and their initial weight was measured. Then they were placed in a chamber with dense atmosphere inside the soil for 75 days. The temperature of the upper part of the chamber was fixed at room temperature by regularly adding water. The condition of the upper part was checked by weighing. The films embedded in the soil were cleaned and dried and then weighed. The average percentage of weight loss was calculated from the following formula (Rajeswari et al., 2020).

$$WL\,(\%) = \frac{Wo - Wt}{Wo} \times 100$$

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Where WL is the percentage of weight loss, W_0 is the initial weight of the film and W_t is the final weight of the film.

Antioxidant Properties

25 mg of the film sample was immersed in 3 ml of distilled water and the resulting solution was measured using DPPH method. According to the DPPH method (2,2-diphenyl-1-picrylhydrazyl), 3 ml of solution was added to 1 ml of 0.1 mm methanol DPPH solution, after 30 minutes of incubation in the dark at room temperature, the absorbance was measured at 517 nm (Li et al., 2018).

Total Phenol Content (Tpc)

The amount of total phenolic compounds was checked according to the method of de Oliveira et al., 2019. For this purpose, 25 mg of the film sample was immersed in 3 ml of distilled water. Then, 0.3 ml of the resulting solution was mixed with 2.5 ml of Folin-Cicaltio reagent (10% by volume/volume) and finally, 2 ml of sodium carbonate (7.5% by weight/volume) was added to it and before measuring the

amount of absorbance at 760 nm, the mixture was incubated at 50°C for 5 minutes. Gallic acid solutions (ppm1000-0) were used to obtain the standard curve. The result was expressed as micrograms equivalent to Gallic acid per gram of film (µg gae/g film) (Li et al., 2018).

Antimicrobial Properties

Agar diffusion disk method was used to determine the antimicrobial activity of the films. Test bacteria included gram-negative Escherichiacly (PTCC = 1217), Staphylococcal gram-positive bacteria (PTCC = 10536), Candida Galabrata (PTCC = 5297) and Candida Albicans (PTCC = 5012). The medium of the Muller Hinton Aggar (MHA) was sterilized in autoclave and cooled 80 mm to 45°C. Then, 100 microliters of the prepared suspensions were poured on the surface of Mueller Hinton agar petri dishes. Finally, before placing the discs on the surface of the culture medium, the surface culture was performed with the help of a sterile cotton swab using 100 microliters of the culture medium with a content of approximately 10^8 CFU/mL of bacteria, mold and yeast. The agar plates were incubated at 25 degrees Celsius for 15 minutes until the microbial coating dried. Then, the film samples were prepared into 6 mm diameter disks using a sterile punch. The discs were placed on Mueller Hinton agar culture medium in completely sterile conditions and kept in a greenhouse at 37°C for 24 hours. The diameter of the created nimbus of the inhibition zone was measured by a digital caliper in millimeters and the results were recorded. The difference in the diameter of the halos formed from the diameter of the discs was considered as an indicator of the antimicrobial activity of the films (Alizadeh et al., 2017).

Scanning electron microscope (SEM)

The surface morphology of the films was observed using a VEGA\TESCAN-LMU scanning electron microscope with an acceleration voltage of 7 kW and a magnification range of 75000 – 1000. For this purpose, a small piece of the film sample was dried before observation. After fixing on the corresponding surface of the scanning electron microscope and coating with gold (8 nm), imaging was done. Three samples per film treatment were analyzed (Rajeswari et al., 2020).

Statistical analysis

For the film tests, a factorial design with a completely randomized basic experimental design and at least three repetitions was used. Analysis of optimization data was done with Design Expert software (version 52, USA) and comparison of other obtained data was done using one way ANOVA with the help of SPSS software (version 28, USA). Duncan's post hoc test was used to compare the means.

Result And Discussion

Mechanical properties and thickness

The thickness of the film plays an important role in its physical properties such as barrier properties and tensile strength. As shown in Fig. 1, the thickness of the films prepared with the concentrations of 0.1 and 0.2 percent of the extract had an approximate average thickness of 0.04 ± 0.005 mm and did not show a significant difference with the cellulose acetate film without the extract ($P < 0.05$). This indicates that the extract is completely placed in the cellulose acetate polymer matrix. The thickness of the cellulose acetate film with 0.3% extract showed a decrease compared to other cellulose acetate films prepared in this study (0.03 ± 0.005 mm).

Previous research showed that the change in the thickness of the active films is mainly due to the increase in the total insoluble solid content in the film-forming emulsion containing essential oil, and the interesting thing is that the thickness of the films is not affected by the type of essential oil used (Xue et al. 2019). The thickness of the final film varies with changing the concentration and surface tension of the solution, the evaporation rate of the solvent, room temperature, humidity, the amount of dissolved solids and how the extract is placed inside the film, etc. (Adilah et al 2017). This result is in agreement with another study that the combination of three essential oils with three different types of polyphenols (tea polyphenols, wax flavones and grape proanthocyanidins) reduced the thickness of chitosan-zein active films, which was attributed to stronger intermolecular interactions and compaction resulted from it (Xue et al., 2021). Mechanical properties such as elasticity, strength and flexibility are very important factors in the development of food packaging. Tensile strength (TS) and elongation at break (EB) are used to indicate film strength and flexibility, which are strongly related to the chemical structure of films (Acevedo-Fani et al., 2015). The results related to mechanical tests (elastic modulus (EM), tensile strength (TS) and elongation to tear of cellulose acetate films containing different concentrations of Falcaria vulgaris extract and ellulose acetate film were presented in Figs. 1 and 2. The results of Fig. 1 showed that addition of Falcaria vulgaris extract to the film increased the elongation to tear compared to cellulose acetate films. Moreover, increase in the concentration of the extract caused the percentage of elongation at the tear point to be increased significantly (17% of the cellulose acetate film against 22% of the 0.3% cellulose acetate film). An increase in elongation at break means a softer film. The linear and simpler the biopolymer structure is, the higher the tensile strength will be, and vice versa. By increasing the intermolecular and intramolecular bonds between the side chains of the biopolymer, which makes its structure more complex, the tenacity of the film will increase and its tensile strength will decrease. Adding Falcaria vulgaris extract to cellulose acetate polymer probably reduces the interaction between cellulose monomers and prevents the interaction between cellulose acetate polymer chains, thus reducing the tensile strength and increasing the elongation at break point. In fact, the plant extract acts as a softener and allows the polymer to increase in length before tearing. These results were consistent with the results of other researchers who added different concentrations of essential oils and extracts to the edible film (Kanmani et al., 2014; Rocha Bastos et al., 2016).

Figure 2 shows that adding Falcaria vulgaris extract to the cellulose acetate film decreased the tensile strength compared to the cellulose acetate film. In addition, with the increase in the concentration of Falcaria vulgaris extract, the tensile strength showed a downward trend, which is due to the destruction of the film network because of adding the extract. The tensile strength of cellulose acetate film was 48.23

MPa and 0.1, 0.2 and 0.3 cellulose acetate films were 46.71, 42.21 and 39.31 MPa, respectively. The tensile strength of the films is related to the chemical structure of the molecules or to the interconnection of the polymer chains in the sheet matrix of the film. Adding Falcaria vulgaris extract to the cellulose acetate film causes inter-strand interaction in cellulose acetate and may prevent running counter effect between the chains of cellulose acetate. In other words, replacing the extract in the polymer matrix causes the interaction between the polar groups of the polymer to be stronger than the interaction between the polar groups of the polymer and the non-polar lipidized groups, and since the extract has a lipid nature, the tensile strength has decreased. These results were consistent with Adilah et al., (2017) who studied the effect of increasing the concentration of mango kernel extract on soy protein isolate film and gelatin.

Figure 2 shows that adding Falcaria vulgaris extract to cellulose acetate film reduces the modulus of elasticity. The modulus of elasticity of cellulose acetate film with 0.3, 0.2, 0.1 and 0% of Falcaria vulgaris extract was 1390, 1500, 1800 and 1900 MPa, respectively. A downward trend was observed with increasing concentration of Falcaria vulgaris extract. The presence of non-polar fat molecules caused a partial break between the polymer chains and finally the softening property of fats decreases (Carvalho et al., 2019). These results are consistent with the section of tensile strength and elongation to tear. This fact has been proven by using the electron microscope of the samples that the addition of the extract was created irregularities in the microstructure of the film and this phenomenon led to a decrease in the mechanical parameters. Another research studied Chitosan films containing essential oils of basil or thyme and found that the induction of close polymer-oil contact with homogenization under high pressure increased the fragility of the films during stretching, which was due to the weakening effect on the Chitosan matrix during the induction of chain reactions. These findings are consistent with the results of the present research (Bonilla et al., 2012). Other researchers suggested in their study that bioactive compounds can decrease tensile strength and increase flexibility by forming hydrogen bonds with chitosan chains and increasing the free volume of the film network (Saberi et al., 2017). The results of this research are completely consistent with previous studies.

Permeability To Water Vapor

Permeability to water vapor in edible films has the ability to prevent the transfer of moisture between the food and the surrounding environment, which is one of the most important practical properties of packaging. In general, the lower value of permeability to water vapor has a good ability to reduce the effective role of water in destructive, chemical, enzymatic reactions and microbial growth in the materials of food film packaging (Polaa et al., 2016). Figure 3 shows that the water vapor permeability of the cellulose acetate film is 188 units and the increase in the concentration of the extract led to a decrease in the vapor permeability of the produced films compared to the cellulose acetate film without extract. Furthermore, the water vapor permeability of the cellulose acetate film with 0.3% of the extract, was 163 units, which can be due to the hydrophobic nature of the phenolic compounds present in the Falcaria vulgaris extract that acts as a barrier inside the film and reduces the rate of water vapor from the film. The mechanism by which water vapor penetrates through packaging films depends on the ratio of

hydrophobic to hydrophilic components of the packaging film. Essential oils are known as a hydrophobic compound, reduce this ratio, and lead to a decrease in permeability to water vapor (Polaa et al., 2016). In addition, the reduction in permeability to water vapor can be explained by the combination between phenolic and the polymer network through the cross-linking of hydrogen bonds or hydrophobic interactions (Ho et al., 2020). Other researchers reported that the combination of tamarind essential oil in a higher concentration increased the water vapor permeability of the films, but the lemon grass essential oil led to a decrease in the water vapor permeability of the film. This may be due to the difference in the hygroscopic nature of the essential oils used, which causes different ability to absorb water into the film network (Ahmed et al., 2012). These results were reported by the researches including Gonçalves et al. (2013); Biswas et al. (2020) and Fawal et al. (2019). Furthermore, the permeability to water vapor is influenced by various factors such as the type of polymer, the amount and composition of the added extract, and the reaction between the added extract and the packaging film (Sanchez-Gonzalez et al., 2011).

Color

The color of the film is one of the important characteristics in the acceptance of the product by the consumer. Table 1 shows that all the produced films had a high brightness index (L), which indicates the transparency of all the samples, and the highest level of this index was related to the cellulose acetate film (90.32 ± 0.86) and with the increase in the concentration of Falcaria vulgaris extract in cellulose acetate film had a reduced brightness index. In fact, the addition of the extract caused a significant decrease in the L index and as a result, an increase in the darkness of the produced cellulose acetate film. As a result, the cellulose film with 0.3% extract showed L index of 86.12 ± 0.65. The produced cellulose acetate films had a significant difference in terms of color index a*, and the highest index was related to the cellulose film (2.53 ± 0.24) and the addition of 0.3% of Falcaria vulgaris extract decreased the index significantly (0.78 ± 0.15). In other words, the amount of red color in the cellulose acetate film decreased with the addition of extract, which was related to the type and color of the added extract. The lowest amount of color index b* was observed in the cellulose film (-5.23 ± 0.22), while the highest amount of color index b* was observed in cellulose acetate films with Falcaria vulgaris extract which indicates an increase in the amount of yellow color in samples with extract.

Table 1
Color index of cellulose acetate films with Falcaria vulgaris extract

Film	*b**	*a**	L	ΔE	WI
cellulose acetate 0	0.22^{a}± -5.23	0.24^{a} ± 2.53	0.86^{a} ± 90.32	0.22^{a} ± 2.72	0.98^{a} ± 88.71
cellulose acetate 0.1	0.18^{a}± -5.11	0.19^{ab} ± 2.11	0.75^{ab} ± 89.95	0.19^{a} ± 2.59	0.85^{a} ± 88.52
cellulose acetate 0.2	0.72^{a}± -4.98	0.22^{b} ± 1.98	0.91^{b} ± 88.32	0.21^{a} ± 2.39	0.72^{a} ± 87.14
cellulose acetate 0.3	0.32^{a}± -5.02	0.15^{c} ± 1.78	0.65^{c} ± 86.12	0.18^{b} ± 3.18	0.54^{b} ± 85.12

* The numbers with same letters in each column are not significantly different (p 0.05).

The produced films were different from each other in terms of the overall color difference, and the biggest color difference was observed in the cellulose acetate film with 0.3% Falcaria vulgaris extract (3.18 ± 0.18). The whiteness index of cellulose film had the highest value (88.71 ± 0.98) and this index decreased with the increase in the concentration of Falcaria vulgaris extract. Therefore, the sample with 0.3% extract showed the lowest whiteness index (85.21 ± 0.54).

The appearance of the produced films is shown in Fig. 4. As can be seen, with the increase in the concentration of Falcaria vulgaris extract, the appearance of the produced films can be seen to decrease the whiteness and increase the amount of yellowness. The lack of significant change in some color indicators can be attributed to the nature of the Falcaria vulgaris extract (brighter color), the lower concentration of the extract used, and the coating effect of cellulose acetate. These results are consistent with Atarz et al. (2010). The researchers investigated the changes in the color index of carrageenan films with different concentrations of grapefruit seed extract. They observed that increasing the concentration of the extract caused a decrease in whiteness, a decrease in brightness, and an increase in yellowness in the tested films. Changes in color index can be affected by the type and amount of extract added to the film (Kanmani et al., 2014). Changes in color index can also be affected by the type and amount of extract added to the film (Kanmani et al., 2014). Other researchers reached similar results and found that adding the concentration of savory essential oil in kefir-carboxymethyl cellulose bio-composite films significantly decreases the L* and a* indexes and increases the b* index compared to the control film (Hasheminya et al., 2019). These results are consistent with the results of the present study.

Opacity

The average opacity of cellulose acetate films with different concentrations of Falcaria vulgaris extract is shown in Fig. 5. With the increase in the concentration of the extract, the opacity of the cellulose acetate films increased and the highest opacity was related to the cellulose acetate film with 0.3% extract (89.54

± 0.65), which in comparison with the cellulose film (85.45 ± 0.95) showed a significant increase, although this decrease in film transparency is not visually visible. Opacity is an important factor related to the amount of light contact on food and has a direct effect on the appearance of the packaged product, which affects customer acceptance. The customer accepts the transparency of food packaging because the product inside the package will be visible. For this reason, the materials should be used in the preparation of edible films that do not significantly increase the amount of opacity (Chen et al 2009).

Other researchers emphasized that the presence of the dispersed and insoluble phase of the essential oil or extract increases the amount of turbidity due to the difference in the refractive index of the phases, the size and concentration of the dispersed particles. These differences are related to the reaction between the extract and the molecules forming the edible film, which reduces the amount of light passing through the film (Mohsenabadi et al., 2018; Monedero et al., 2009). In another study, it was stated that, in general, films containing eugenol or ginger essential oil are more opaque than films without essential oil. This was probably due to the effect of essential oil droplets dispersed in the network of biopolymers, which can change the light transmission properties of the film (Bonilla et al., 2018). The combined effect of the film's different thickness, distribution of essential oils in films and surface and internal structure of films can ultimately affect their light scattering characteristics (Acevedo-Fani et al., 2018).

Solubility Percentage, Swelling, Contact Angle And Water Absorption

Solubility in water is one of the characteristics of edible films and coatings. Normally, the property of low solubility in water is essential in increasing the overall integrity of the film in wet environment (Ojagh et al., 2010). Table 2 reports the solubility of the produced cellulose acetate films. The solubility of films with Falcaria vulgaris extract was decreased compared to cellulose acetate film. The solubility percentage of cellulose acetate film containing 0.3% extract was reported as 30.09 ± 0.93, which significantly decreased compared to cellulose acetate film with 36.21 ± 1.08. The decrease in solubility of cellulose acetate film with 0.1 and 0.2 percent was significantly different from cellulose acetate film by 34.097 ± 97 and 33.1 ± 14.33, respectively ($p < 0.05$).

Table 2
Contact angle, swelling, solubility and water content of cellulose acetate film containing different Falcaria vulgaris extract concentration (0, 0.1, 0.2 and 0.3%).

Film type	Contact angle (CA, º)	Water content (WC, %)	The degree of swelling (SW) (%)	Water solubility (WS, %)
cellulose acetate 0	59.32 ± 0.97 [a]	23.45 ± 0.22 [a]	0.320 ± 0.09 [a]	36.21 ± 1.08 [a]
cellulose acetate 0.1	60.21 ± 1.07 [ab]	22.16 ± 0.31 [b]	0.302 ± 0.04 [b]	34.97 ± 0.89 [ab]
cellulose acetate 0.2	62.11 ± 1.24 [b]	21.98 ± 0.28[b]	0.300 ± 0.03 [b]	33.14 ± 1.13 [b]
cellulose acetate 0.3	65.47 ± 1.16 [c]	20.17 ± 0.25 [c]	0.276 ± 0.07 [c]	30.09 ± 0.93 [c]

* The numbers with same letters in each column are not significantly different (p 0.05).

Cellulose acetate is very sensitive to water, which can be the main factor explaining the relatively high solubility of control films without extract. As mentioned in the contact angle section of the present study, cellulose acetate film has a hydrophilic property, which is mostly related to its cellulose. However, since the extracts were a hydrophobic compound, by being placed in the film structure and creating a new arrangement with biopolymers, they reduced the solubility of the film. According to Table 2, the solubility of water in the films was reduced by adding Falcaria vulgaris extract with different concentrations. Other researchers reported that increasing the concentration of clove essential oil from 0.5–1.5% in pectin film reduced the moisture content and solubility of the films. The decrease in film solubility is due to the hydrophobic nature of the essential oil and the interaction between essential oil components and hydroxyl groups (Yuan et al., 2015). The observations from this research are also consistent with the data reported by other researchers (Vahedikia et al., 2016; Nisar et al., 2018).

Moisture content of biodegradable packaging is an important physical property of film and plays a fundamental role in the shelf life of packaged materials or coatings. Moisture content is a variable relating to the total free volume occupied by water molecules in the network of films (Roy & Rhim, 2020). Table 2 also shows the percentage of water content of cellulose acetate films prepared with Falcaria vulgaris extract. As can be seen, the addition of Falcaria vulgaris extract reduces the amount of water absorption in cellulose acetate films, which is consistent with the results of the solubility percentage. The percentage of water content of cellulose acetate film is equal to 23.45, which showed a significant decrease compared to cellulose acetate film 20.17 with 0.3% of Falcaria vulgaris extract. This decrease is mainly due to the hydrophobic properties of the essential oils of the extracts and the interaction of the oil components with the hydroxyl groups of the films, which can limit the interaction of the hydroxyl groups with water molecules due to their less accessibility (Nisar et al., 2018). The difference in moisture content may be related to the microstructure of cellulose acetate films with Falcaria vulgaris extract and oil solubility (Wu et al., 2019). In previous studies, it was stated that the increase in the moisture content may

be related to the loss of the film network, which increases the amount of water molecules between the polymer chains (Haghighi et al., 2019). Therefore, it can be concluded that by increasing the concentration of Falcaria vulgaris extract, the network of cellulose acetate film is formed more coherently and densely, which can be confirmed by the results of scanning electron microscope. Other researchers attributed the reason for the decrease in the moisture content of chitosan films with cinnamon essential oil to the compact structure of the film as a result of the formation of covalent bonds between the functional groups of the chitosan chain (Ojagh et al., 2010). In another study, researchers reported that adding clove essential oil to pectin film reduces the amount of moisture content due to the hydrophobic properties of the essential oil and the interaction of the oily components with the hydroxyl groups of the pectin film (Nisar et al., 2018). Other researchers also obtained similar results on triticale protein film containing oregano essential oil (Aguirre et al., 2013).

The contact angle is used as a parameter to estimate the film's water resistance and determine the hydrophobicity or hydrophilicity of the film surface (Pereda et al., 2010). When the drop is placed on the surface, attraction is established between the water molecule and the polymer surface. The strength of this attraction depends on the characteristics of the liquid and the surface. A smaller contact angle between the droplet and the polymer surface means that there is a strong attraction between the water and the polymer surface and therefore they can stick together better. In general, a low contact angle (less than 65°) indicates a hydrophilic surface, while a high contact angle (greater than 65°) characterizes a hydrophobic surface (Sharma et al., 2020). The results of the contact angle of the film using water as a polar solvent are shown in Table 2. Despite the hydrophilic nature of cellulose acetate, the percentage of contact angle was relatively high (59.0 ± 32.97). The contact angle percentage increased significantly with the addition of Falcaria vulgaris extract, which is due to the hydrophobicity of the extract ($p < 0.05$). The contact angle of cellulose acetate films with 0.1, 0.2 and 0.3 percent of the extract was (60. 1 ± 21.07, 62. 11 ± 1.24 and 65.47 ± 1.16), respectively. Cellulose acetate film with 0.3% extract shows the highest contact angle, the lowest swelling rate and the lowest water vapor permeability all of which were confirmed in the current research. The lower contact angle of cellulose acetate film without extract can be attributed to the binding capacity of water with glycerol and active groups of cellulose acetate. The increase in contact angle is due to the coverage of functional and hydrophilic groups in cellulose acetate, which is consistent with the results of chitosan edible film containing cinnamon and caseiate film containing polished oil (Ojagh et al., 2010). The increase in contact angle may also be due to the more uniform distribution of the oil phase and greater affinity between the compounds of Falcaria vulgaris extract and cellulose acetate film, which leads to the formation of tighter and denser films. In general, a smaller contact angle allows the film to spread on the substrate and absorb more water, and a larger contact angle increases the hydrophobic nature of the film. An increase in the contact angle of cellulose acetate film with 0.3% extract indicates an increase in the hydrophobicity of cellulose acetate film, which plays an important role in increasing the quality of packaging (Xue et al., 2021).

The percentage of swelling can be attributed to the hydrophilicity of polymer films, which usually shows the film's ability to absorb water and refers to maintaining quality during packaging and storage of food. Table 2 also shows the swelling percentage of cellulose acetate films. The swelling percentage was

significantly affected by the addition of the extract ($p < 0.05$). According to the obtained results, the cellulose acetate film has the highest percentage of swelling, which is related to the hydrophilic nature of cellulose acetate that increases its ability to absorb water. Increasing the concentration of Falcaria vulgaris extract from 0.1–0.3% has reduced the swelling ratio from 0.302 to 0.276. It should be noted that there is no significant difference between 0.1% and 0.2% cellulose acetate film. The decrease in swelling percentage with the increase in the concentration of the extract is related to its hydrophobic nature. The hydrophobic parts of cellulose acetate interact with the extract through hydrophobic reactions, and as a result, the surface interaction between cellulose and the extract is improved. Falcaria vulgaris extract has a filling role here and saturates the network of the cellulose acetate film, and water molecules cannot penetrate the network of the cellulose acetate film, and the amount of swelling decreases. The decrease in the percentage of swelling indicates a higher resistance to the penetration (absorption) of water than other films, which is probably due to the increase in crosslink density through the interaction between cellulose acetate and Falcaria vulgaris extract, which is consistent with the results of other researchers (Huerta et al., 2019; Torabi et al., 2018; Archana et al., 2013).

Biodegradation Test

Degradation of cellulose acetate films with different concentrations of Falcaria vulgaris extract for 75 days is shown in Fig. 6 for each film as a percentage of weight loss versus time. Based on the results shown, the cellulose acetate film was destroyed within 75 days (25% by weight) and in the same time, cellulose acetate films containing 0.1, 0.2 and 0.3% of Falcaria vulgaris extract were destroyed amount to 65, 80 and 95%, respectively. According to the results shown, the cellulose acetate film with 0.3% extract completely degraded and decomposed in the soil within 75 days and showed the highest level of biodegradability compared to the cellulose acetate film without extract. The reason for this can be attributed to the changes of microscopic structure, as mentioned in the electron microscope section. In fact, the addition of the extract causes irregularities in the structure of the cellulose acetate film, and this accelerates the rate of decomposition of the cellulose acetate film with Falcaria vulgaris extract, which is consistent with the results of other researchers who investigated the biodegradability of polyvinyl alcohol and starch composite films (Negim et al., 2014).

During degradation, the polymer was first converted into its monomers and then these monomers were decomposed into their constituent minerals. Biodegradability depends on various factors including polymer properties (mobility rate, molecular weight, type of functional groups and crystallization, etc.), type of organism and nature of pre-process. The rate of degradation depends on temperature (50–70), humidity and type of microbes. Decomposition will be fast if all three factors are present (Emadian et al., 2016). In another study, the biodegradability of cellulose acetate, carrageenan and alginate composite film was investigated within 70 days, and its biodegradability was reported in three months (Rajeswari et al., 2020).

Antioxidant Properties

Oxidation is one of the most important causes of food spoilage. This reaction occurs during the processing and storage of food products. The mechanism of antioxidant activity of plant extracts in combination with edible films and coatings has been described as improving the property of the barrier capacity against oxygen and involving oxygen or the release of plant compounds inside the coated product (Hashemia et al., 2017). The antioxidant activity of Falcaria vulgaris extract is largely related to the presence of phenolic groups in it. Comparing the results of total phenol content and DPPH free radical inhibitory activity in the research, this direct relationship can also be seen, so that the structure of phenolic compounds enables the extract to have the ability trap DPPH free radicals and act as antioxidants (Lopez et al., 2003). The phenolic compounds of Falcaria vulgaris extract were estimated based on the reduction of Folin Ciacalto reagent and the results are shown in Fig. 7 The results show that cellulose acetate had a very low total phenol content (0.11 mg of Gallic acid per dry weight of the film) and with increasing the concentration of the extract in the cellulose acetate film, the amount of total phenol increased significantly ($p < 0.05$). In this respect, the cellulose acetate film with 0.3% extract showed the highest total phenol content (19.93 mg of Gallic acid per dry weight of the film). In a study conducted by other researchers regarding soy protein isolate films containing thyme essential oil, they reached a similar result that due to the high total phenol content of thyme essential oil, the antioxidant property of soy protein isolate film increased with the increase in the concentration of essential oil (Gholizadeh et al., 2018). Other researchers also reported that essential oil and extract are rich sources of phenolic content and the antioxidant power of films depends on the concentration of essential oil added to them (Hasheminya et al., 2019). The report of this research confirmd the results of the current research. As a result, by increasing the concentration of Falcaria vulgaris extract in active cellulose acetate film, the content of total phenol and finally the antioxidant property of the film increased.

The results related to the amount of antioxidant activity in the form of ability to trap DPPH radical of cellulose acetate films and different concentrations of Falcaria vulgaris extract are presented in Fig. 7. As can be seen, with the increase in the concentration of Falcaria vulgaris extract, the ability to trap DPPH radical increases, which indicates the increase in antioxidant power with the increase in the concentration of the extract. In this regard, the DPPH radical trapping rate of cellulose acetate film with 0.3% extract was 35.31% compared to cellulose acetate film with 0.1% (16.21%), cellulose acetate film with 0.2% and cellulose acetate film (0.15%) showed a significant increase ($p < 0.05$). Cellulose acetate film showed weak antioxidant activity due to the presence of polysaccharide hydroxyl groups (Hashemia et al., 2017). The presence of phenolic compounds in Falcaria vulgaris extract increased the antioxidant activity of cellulose acetate film, which is due to the increase in the reaction of free radicals with free amine groups (NH), which form stable radical macromolecules. In other words, amine groups can create ammonium groups (NH_4) by absorbing hydrogen ions from the solution. These compounds break the chain reactions of free radicals, the hydroxyl groups of phenolic compounds act as hydrogen donors. The connection between antioxidant and cellulose acetate matrix was created by hydrogen bonds, especially during film formation (Gholizadeh et al., 2018). Another study investigated the effect of aloe Vera and rosemary essential oil on the physicochemical properties of cellulose acetate film and showed that the addition of essential oils to the film significantly increased their free radical scavenging properties (Fawal et al.,

2019). In another study, the researchers found that adding eugenol essential oil to gelatin-chitosan films increased the antioxidant activity of these films, and suggested to use gelatin-chitosan films containing eugenol essential oil as active packaging for food products with high oxidation rates due to increasing the shelf life of these products. (Bonilla et al., 2018). Other researchers also reached a similar result. They reported that the addition of bay leaf essential oil to edible films made from carboxymethyl cellulose increased its antioxidant properties due to the phenolic content of bay leaf essential oil (Rincón et al., 2019).

Morphological Features And Microscopic Structure Of Activated Cellulose Acetate Films

Microscopic images obtained from the cross-section of cellulose acetate films with different concentrations of Falcaria vulgaris extract are shown in Fig. 8. As can be seen, there are pores in the transverse structure of the cellulose acetate film, which is due to the high volatility of acetone and its evaporation during the drying process of the films.

As can be seen, the places where the extract is present are marked with a darker color compared to the other parts, and it shows that there is a weak reaction between the extract, which is predominantly hydrophobic, and the hydrophilic polymer matrix of cellulose acetate. In all cases, the liquid nature of the extract increased the creaminess of the drops on the surface of the film after drying and filling the irregular parts of the surface of the cellulose acetate matrix and made it softer. With the increase in extract concentration (0.3%), the roughness shows a tendency to increase, which is related to the process of fat accumulation and creaming during the drying stage, and causes some irregularities to be seen on the surface of the film. Therefore, the polymer-extract interaction affects the final molecular structure of the film. Not only the nature of the seed extract, but also the homogenization method determined the mechanisms that affected the molecular structure of the films (Freitas et al., 2020; Atarés et al. 2010).

Oxygen Permeability

The composition of gases in the upper space of the packaging is one of the most important aspects of packaging for the storage of agricultural products that are minimally processed. Increasing the amount of oxygen inside the packaging leads to undesirable reactions such as oxidation and browning, and the low amount of oxygen accelerates the growth of anaerobic bacteria. Therefore, it is necessary to maintain the proper ratio of oxygen to carbon dioxide in food packaging. An oxygen level of less than 2% promotes the growth of anaerobic bacteria such as clostridium botulinum and semi-aerobic bacteria such as Listeria species and lactic acid bacteria. The characteristics of oxygen permeability is an important factor in determining the compounds present in the upper space of the package. Oxygen permeability is reported as the oxygen permeability coefficient, which is the amount of oxygen passing through a certain part of the package at a certain time. Therefore, packaging that has a lower oxygen permeability

increases the shelf life of the product (Abdul Khalil et al., 2018). Degradable films usually have higher oxygen permeability, which limits their use in the food industry.

As seen in Fig. 9, the oxygen permeability of cellulose acetate films increases with the addition of Falcaria vulgaris extract and increasing its concentration from 0.1 to 0.3%. in this respect, the OTR of cellulose acetate film with 0.3% extract was equal to 800 units, while the cellulose acetate film without extract had OTR equal to 650 units. The softening effect of the extract observed in this part is consistent with the results of the water vapor permeability analysis and the mechanical properties section in the previous part. The extract moves the cellulose acetate polymer chain, weakens the intermolecular reactions, and increases the oxygen permeability process. In addition, the permeability to oxygen due to the high solubility of oxygen in the hydrophobic phase distributed in the cellulose acetate polymer matrix was also beneficial (Ma et al., 2018).

In another study, Rojas-Graü et al. (2006) added different essential oils to the edible film to cover apple puree and observed that the oxygen permeability of the produced film increased. Other researchers added oregano essential oil to cellulose acetate film and obtained similar results, both of which confirm our hypothesis (Polaa et al., 2016).

Antimicrobial Assay

Despite the use of preservatives to control microorganisms that cause food spoilage and infection, food safety is an important challenge in the industry. Currently, the use of natural preservatives is a healthy and effective alternative to chemicals (Shafaghat, 2011). Evaluation of antibacterial activity is necessary to determine the activity of edible film against bacterial pathogens for different applications. Antimicrobial properties of cellulose acetate film and cellulose acetate films with different concentrations of Falcaria vulgaris extract against two bacteria Escherichia coli and Staphylococcus aureus and two fungi Candida albicans and Candida glabrata were presented in Table 3. Antimicrobial activity was determined using the disk penetration method. The diameter of the formed halos was considered as an indicator of the antimicrobial activity of the films. In cases where a halo was not formed, that is, there was no antimicrobial activity, and instead of the initial disc diameter, its value was reported as ND. The diameters of the halos obtained from the disk penetration test for films with different concentrations of extract are shown in Table 3. When antimicrobial substances were added to the film, these compounds penetrated into the agar and showed antimicrobial properties. The results showed that the cellulose acetate film had no antimicrobial activity, which is consistent with the results of other researchers (Stoja et al., 2017). Adding different concentrations of extract to cellulose acetate films caused an antimicrobial property in the film and caused a significant decrease in the population of studied microbes, and by increasing the concentration of the extract from 0.1 to 0.3% in the prepared cellulose acetate film, the antimicrobial property of the film increased in agar medium. The results of Table 3 show the effect of films containing Falcaria vulgaris extract on gram-negative bacteria (Escherichia coli) and gram-positive bacteria (Staphylococcus aureus). As can be seen, the diameter of the halo for cellulose acetate film with

0.1% extract against Escherichia coli and Staphylococcus aureus bacteria was reported to be 7.02 and 5.98 mm, respectively, and increasing the concentration of the extract to 0.3%, the diameter of the halo was 11.35 and increased by 10.42 mm.

Table 3
Antibacterial and antifungal effect of cellulose acetate film containing different Falcaria vulgaris extract concentration (0, 0.1, 0.2 and 0.3%).

Treatment	**The diameter of the inhibition zone (mm)**			
	Candida albicans	*Candida glabrata*	*Staphylococcus aureus*	*Escherichia coli*
cellulose acetate 0	ND	ND	ND	ND
cellulose acetate 0.1	10.18 ± 0.89^{c}	5.98 ± 0.29^{c}	7.02 ± 0.67^{c}	12.93 ± 0.94^{c}
cellulose acetate 0.2	13.21 ± 0.61^{b}	8.15 ± 0.33^{b}	9.22 ± 0.76^{b}	16.39 ± 0.75^{b}
cellulose acetate 0.3	16.65 ± 0.73^{a}	10.42 ± 0.51^{a}	11.35 ± 0.83^{a}	17.94 ± 0.81^{a}

* The numbers with same letters in each column are not significantly different (p 0.05).

Monoterpenes, sesquiterpenes and their oxygenated derivatives are the main compounds of plant extracts that show antimicrobial activity. The phenolic compounds in Falcaria vulgaris extract exert their antibacterial activity by forming a complex with bacterial membrane proteins. Furthermore, the antimicrobial activity increases with the increase in the concentration of the extract because a greater amount of phenolic compounds will pass through the cell membrane, and it is predicted that this can lead to increasing the physical disturbance in phospholipid stability and reducing the number of viable cells. These results were consistent with the researchers' results regarding the use of rosemary, aloe Vera, thyme and clove essential oils in edible film against Escherichia coli, Staphylococcus aureus and Bacillus subtilus bacteria (Sharma et al., 2020, Fawal et al., 2019). Another researcher investigated the antibacterial activity of edible film containing clove essential oil against Salmonella Typhimurium and Listeria monocytogenes (Mulla et al., 2017).

Plant extracts affect bacteria through structural damage and cell membrane changes. Plant extracts penetrate into the mitochondrial membrane, leading to greater organelle permeability and potassium ion leakage. The release of potassium ions out of the cell is a clear sign of damage to the cell membrane and its death. In general, gram-negative bacteria are less sensitive to antimicrobial substances of plant origin compared to gram-positive bacteria, which is due to the outer lipopolysaccharide membrane of gram-negative bacteria that limits the release of hydrophobic compounds. Nevertheless, this does not mean that gram-positive bacteria are always more sensitive to antimicrobial compounds. These plant substances affect the cell wall of microorganisms through various mechanisms, which include attacking the phospholipids of the two layers of the cell membrane, destroying enzyme systems, destroying the

genetic material of bacteria, and forming fatty acid hydro peroxidase, which causes the oxygenation of unsaturated fatty acids. The effectiveness of herbal antimicrobial compounds depends on various factors. They include the method of extracting essential oils or extracts from plant materials; the amount of inoculated microorganisms, the growth phase of microorganisms, the culture medium used, the internal and external characteristics of food such as the amount of fat, protein, water and antioxidants; preservatives; pH; temperature and duration of greenhouse storage; packaging method and physical structure of food (Francisco et al., 2018; Gonçalves et al., 2013, Fawal et al., 2019).

The results of the antifungal effect of cellulose acetate films containing different concentrations of Falcaria vulgaris extract were also presented in Table 3. According to the results, the halo diameter for cellulose acetate film with 0.1% extract against Candida albicans and Candida glabrata fungi was reported to be 10.18 and 12.93 mm, respectively, and increasing the concentration of the extract to 0.3%, halo diameter increased to 16.65 and 17.94 mm, respectively. In 2019, researchers prepared chitosan films with oil essence of oregano, pepper, and thyme and examined their antifungal effect. The known mechanism for the antifungal activity of plant extracts is called degradation mechanism. Due to its hydrophobic structure, the plant extract can attack the cell membrane and destroy the cell membrane, which leads to the releasing of cell contents, affecting the enzyme system, hindering the respiratory system, and causing cell death. Since plant extracts are composed of various compounds (terpenoids, esters, aldehydes, ketones, acids, and alcohols), it is difficult to relate antifungal activity to a specific compound (Noshirvani et al., 2017). In fact, the antifungal activity of plant extracts is the result of the intensifying activity of various compounds present in the plant extract. Studies have shown that there are effective compounds such as α-Terpiny acetate, α -Caryophyllene and α -Pinene in the essential oil and extract of Falcaria vulgaris plant, which have antimicrobial activity (Shafaghat 2010, Shafaghat 2011). These compounds have an inhibitory effect on the enzymes forming the microbial cell wall. In addition, they cause the structural and functional destruction of the cytoplasm, mitochondrial membrane and cell membrane and the destruction of the order of the microbial cell wall. Another antimicrobial mechanism of these compounds is to create changes in the permeability of the cell membrane and cytoplasmic membrane, which leads to the leakage of the cytoplasmic content of the cell. (Hossain et al., 2012, Hyldgaard et al., 2012). In general, the difference in the antimicrobial activity of plant extracts depends on the ratio of polymer to extract in the polymer, the composition and nature of the extract, and the reaction between the active compounds in the extract and the polymer that affects the penetration of the extract in the culture medium (Sanchez-Gonzalez et al., 2011).

Conclusion

Due to the problems of petroleum polymers in environmental pollution, biodegradable films have the ability to replace many of these packages. The results of the evaluations showed that the strength, tensile strength and modulus of elasticity of the produced cellulose acetate films decreased with the increase of Falcaria vulgaris extract. The amount of elongation increased to the extent of breaking stage of films containing Falcaria vulgaris extract. In other words, the resulting films became weaker, which can be related to the intensification of the softening effect of Falcaria vulgaris extract. Increasing the

concentration of the extract led to a decrease in vapor permeability, water absorption, solubility, swelling percentage and water content percentage, due to its hydrophobic properties. Furthermore, it led to an increase in the contact angle of the produced films compared to the cellulose acetate film without extract, all of which are effective in maintaining the quality and increasing the durability of the packaged food material. Increasing the concentration of Falcaria vulgaris extract caused changes in the appearance of the produced films, and the increase in turbidity was one of these changes. The presence of phenolic compounds in Falcaria vulgaris extract increased the amount of antioxidant activity and DPPH active radical inhibition of cellulose acetate film and these compounds broke the chain reactions of free radicals. Cellulose acetate films with different concentrations of the extract showed higher oxygen permeability than cellulose acetate, the softening effect of the extract observed in this part is consistent with the results of the water vapor permeability analysis and the mechanical properties section. The results showed that active films with different concentrations of Falcaria vulgaris extract had appropriate antimicrobial properties against two bacteria, Escherichia coli and Staphylococcus aureus, and two fungi, Candida albicans and Candida glabrata. Therefore, it can be concluded that the use of these films for food can be useful.

Declarations

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Consent to participate

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Availability of data and material

The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

Code availability

Not applicable

Author Contribution

Zeinab Hassanloofard: Conceptualization (lead); writing –original draft (lead); formal analysis (lead). **Mehdi Gharekhani**: Software (lead); writing –original draft (supporting); writing –review and editing (equal); **Mohsen Zandi**: Software (lead); writing –original draft (supporting); writing –review and editing (equal); methodology (lead); conceptualization (lead); **Ali Ganjloo**: review and editing (equal); conceptualization (supporting). **Leila Roufegarinejad**: review and editing (equal); conceptualization (supporting).

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Figures

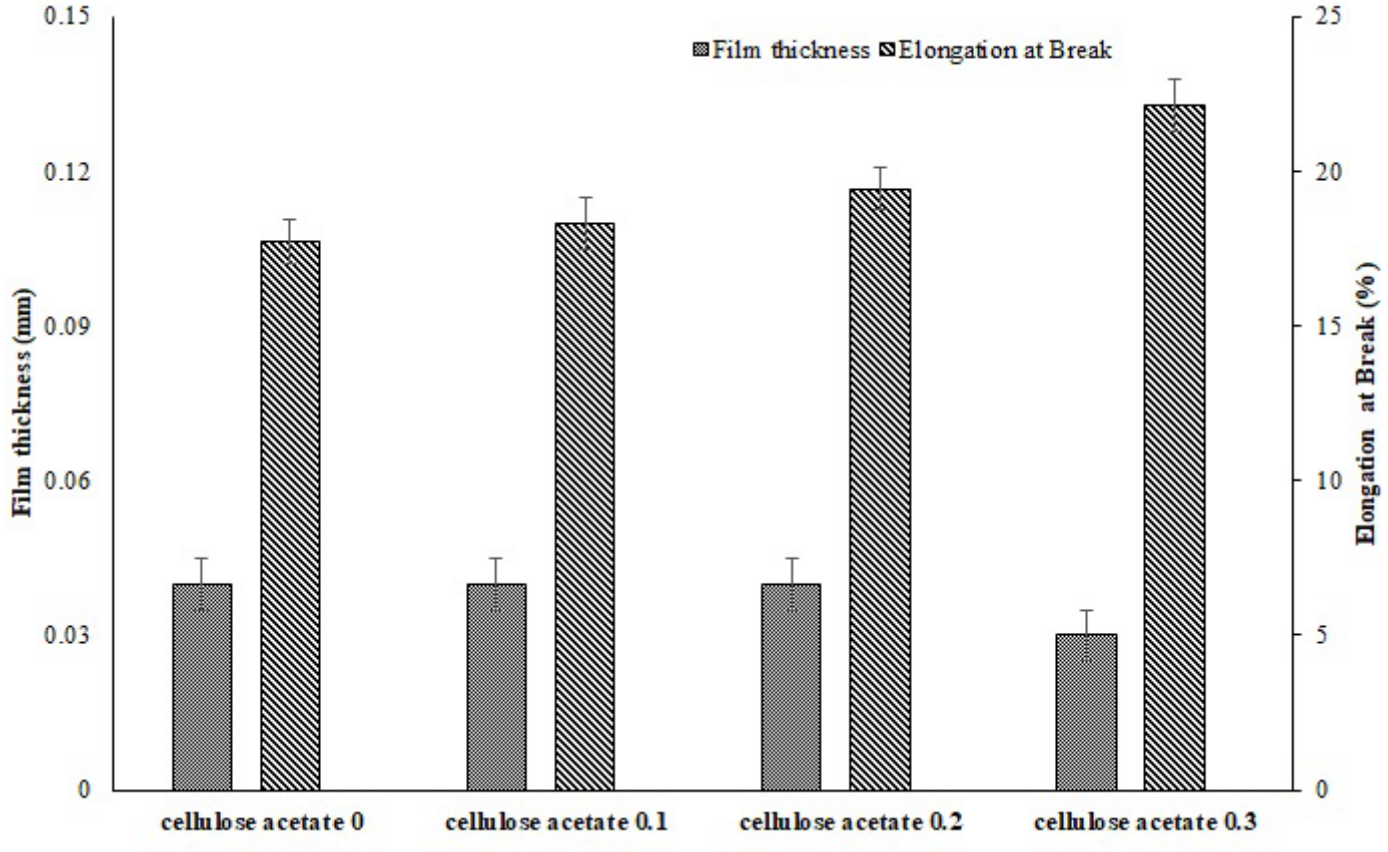


Figure 1

Thickness and elongation at break of cellulose acetate film containing different Falcaria vulgaris extract concentration (0, 0.1, 0.2 and 0.3%).

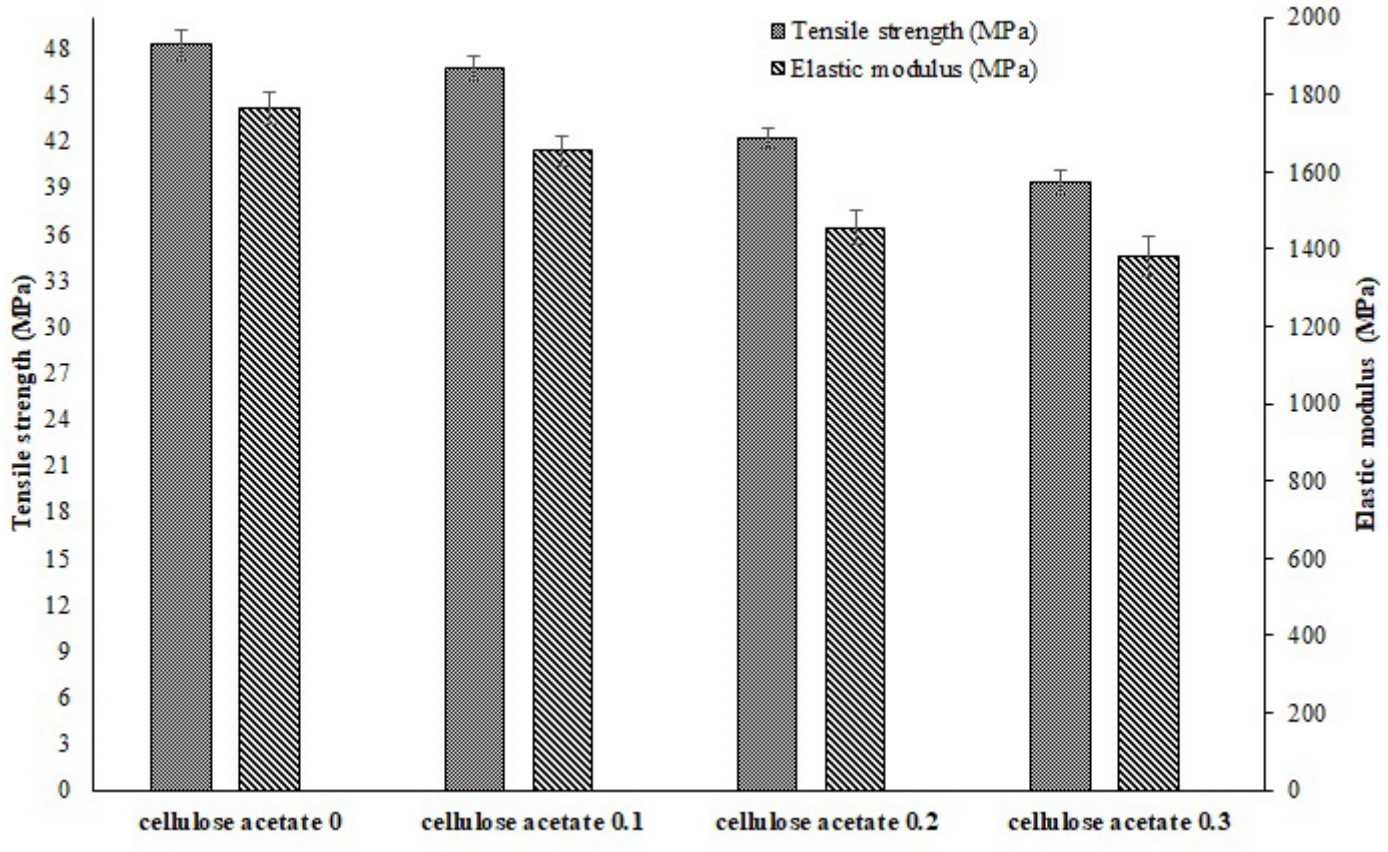


Figure 2

Tensile strength and elastic modulus of cellulose acetate film containing different Falcaria vulgaris extract concentration (0, 0.1, 0.2 and 0.3%).

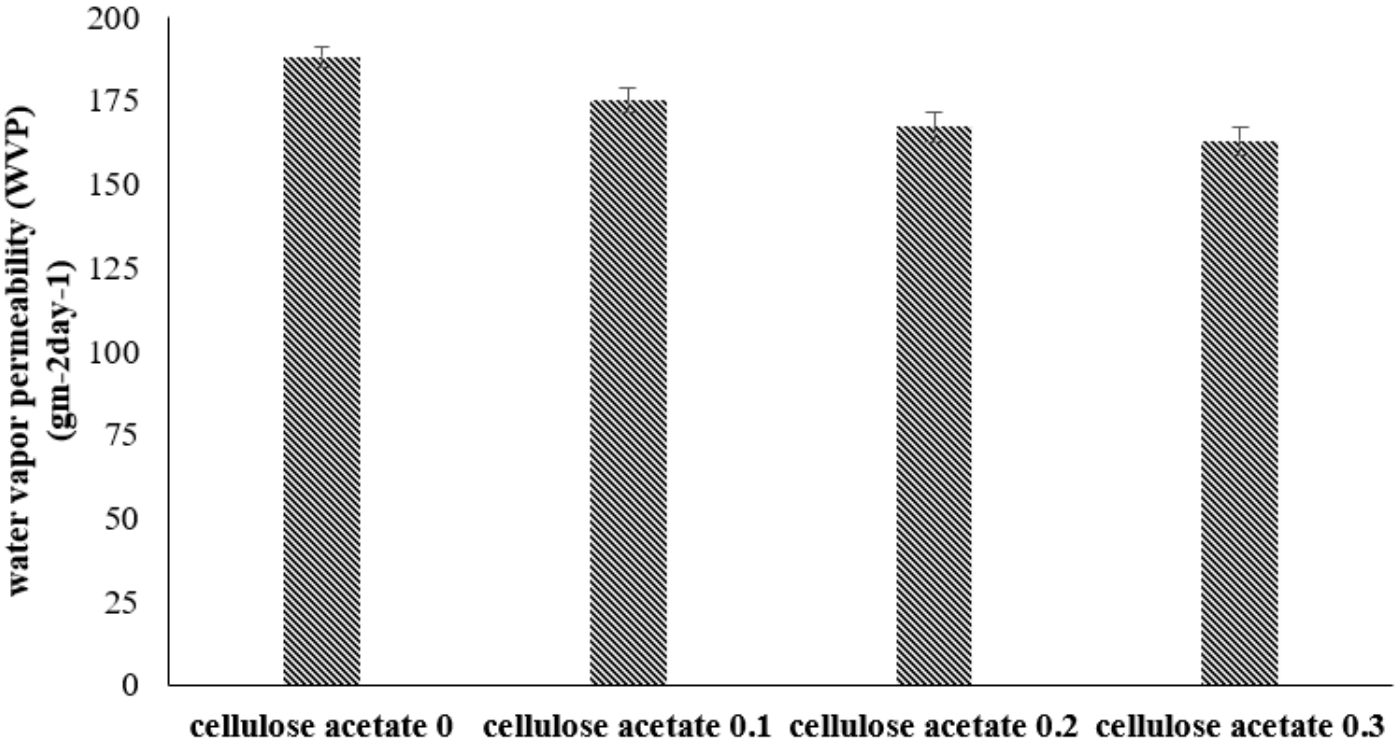


Figure 3

Water vapor permeability of cellulose acetate film containing different Falcaria vulgaris extract concentration (0, 0.1, 0.2 and 0.3%).

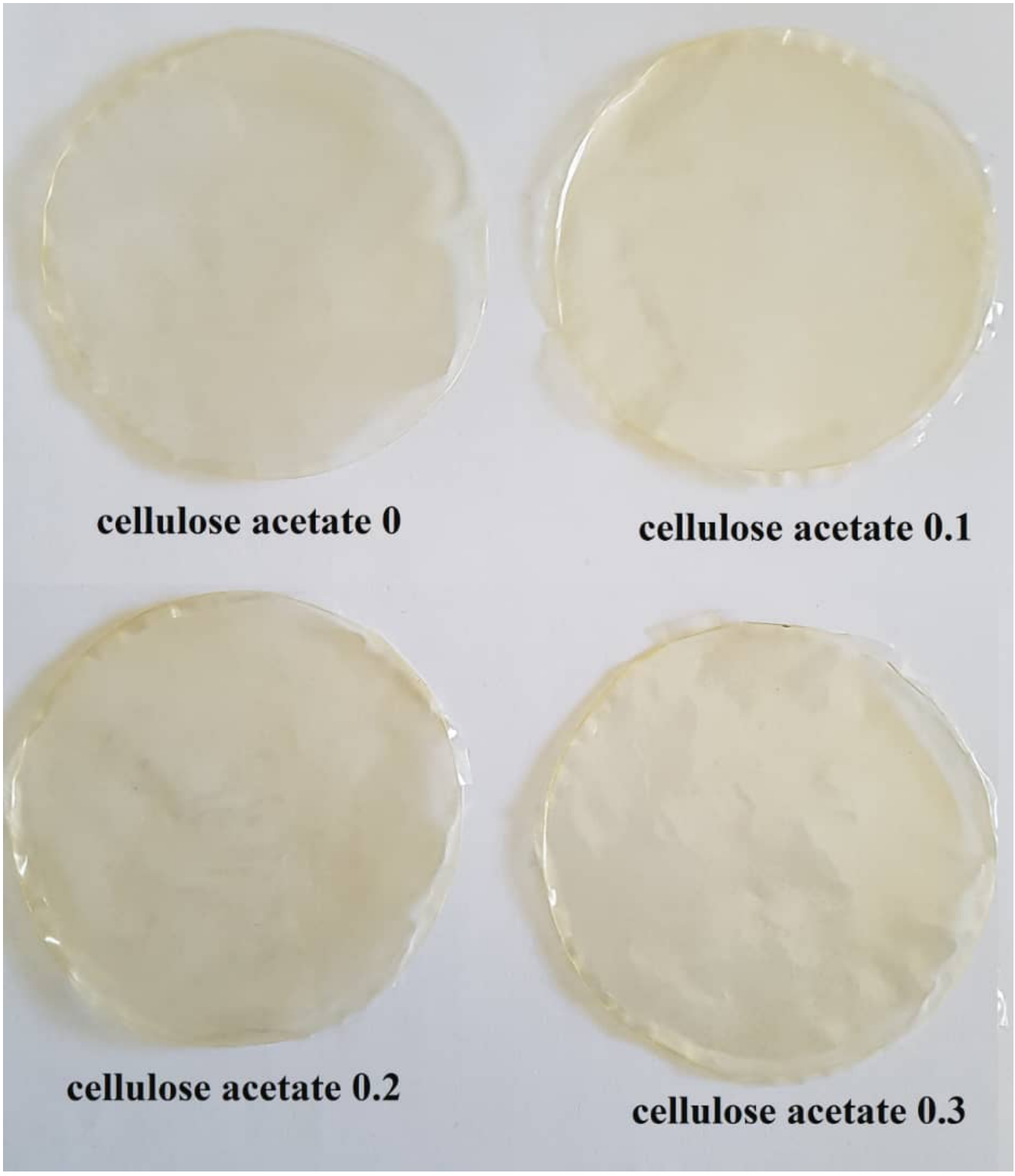


Figure 4

Cellulose acetate film containing different Falcaria vulgaris extract concentration (0, 0.1, 0.2 and 0.3%).

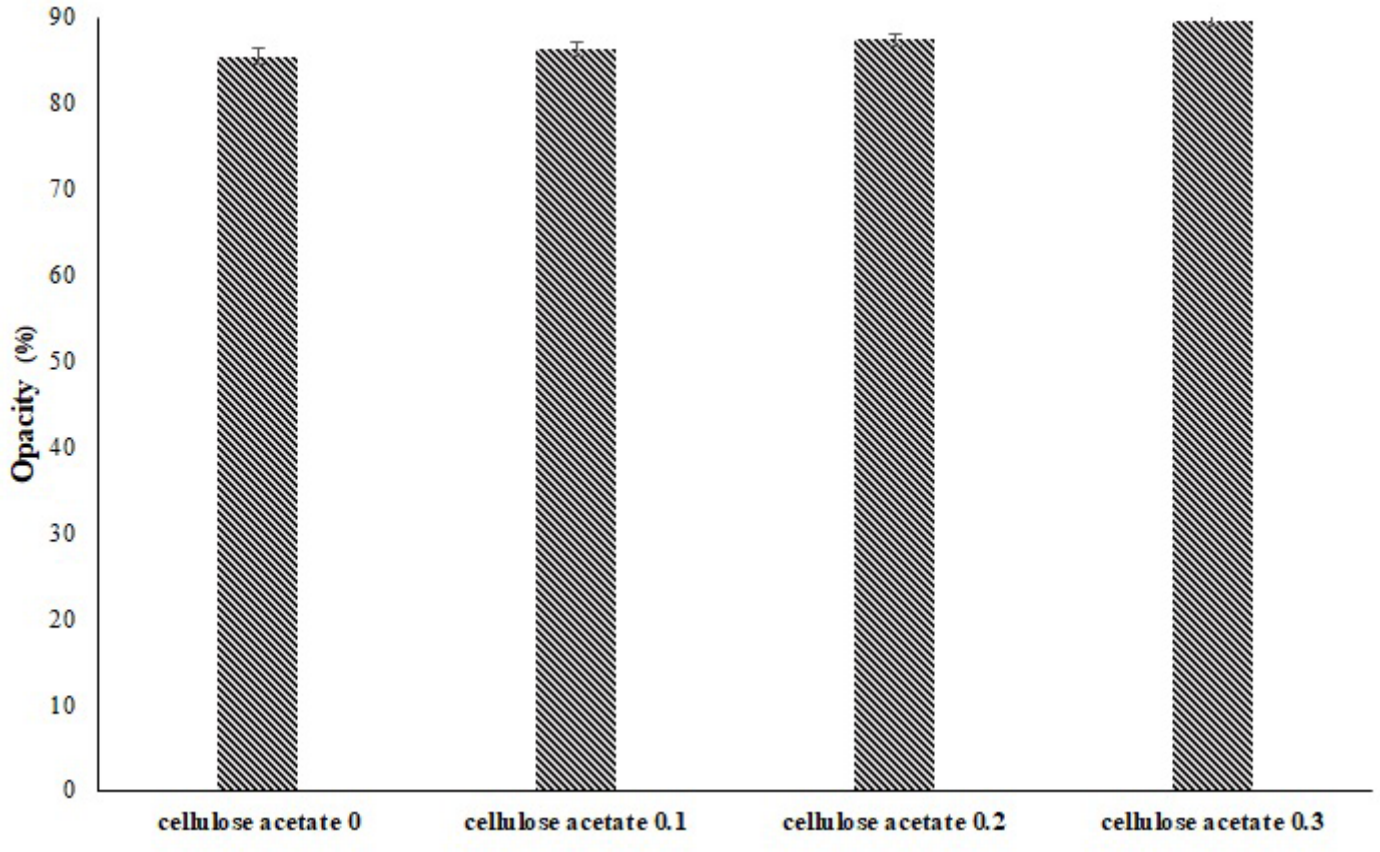


Figure 5

Opacity of cellulose acetate film containing different Falcaria vulgaris extract concentration (0, 0.1, 0.2 and 0.3%).

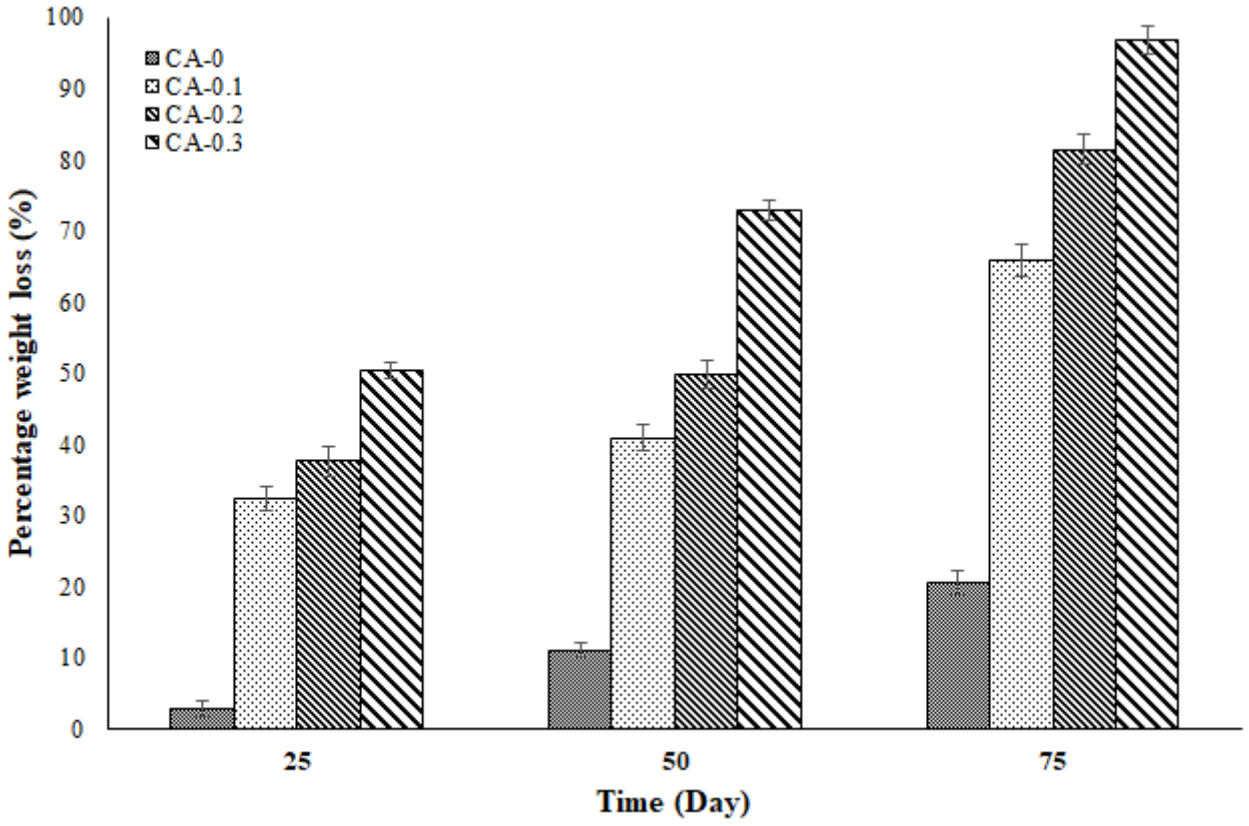


Figure 6

Weight loss of cellulose acetate film containing different Falcaria vulgaris extract concentration (0, 0.1, 0.2 and 0.3%).

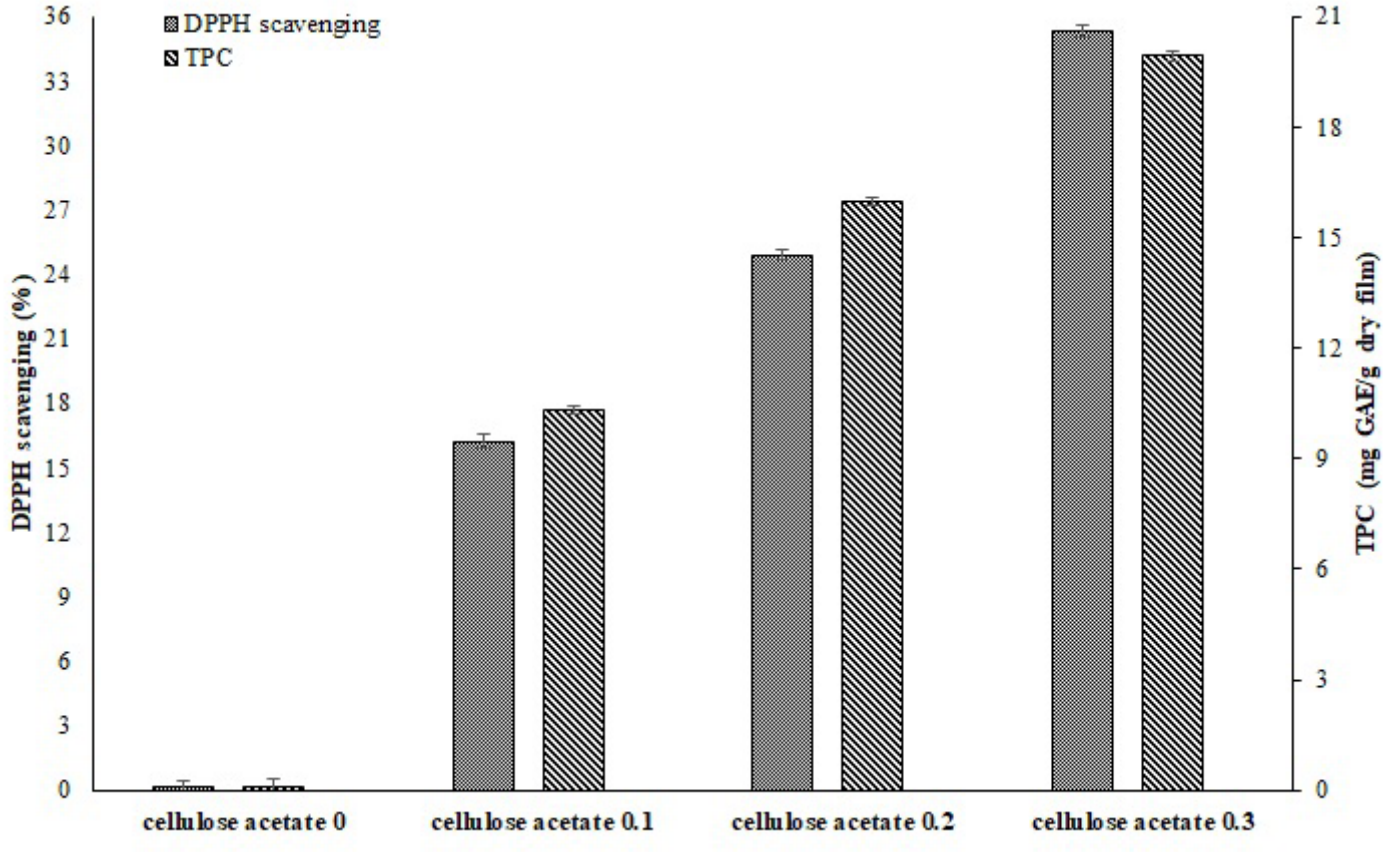


Figure 7

Total phenol content and antioxidant activity of cellulose acetate film containing different Falcaria vulgaris extract concentration (0, 0.1, 0.2 and 0.3%).

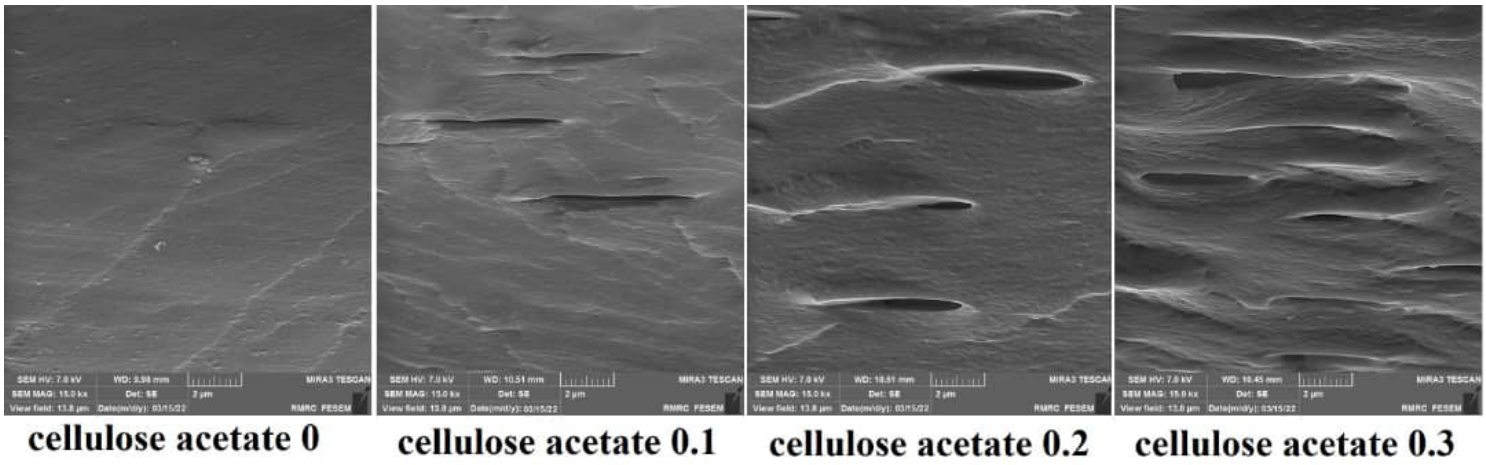


Figure 8

Microscopic structure of cellulose acetate film containing different Falcaria vulgaris extract concentration (0, 0.1, 0.2 and 0.3%).

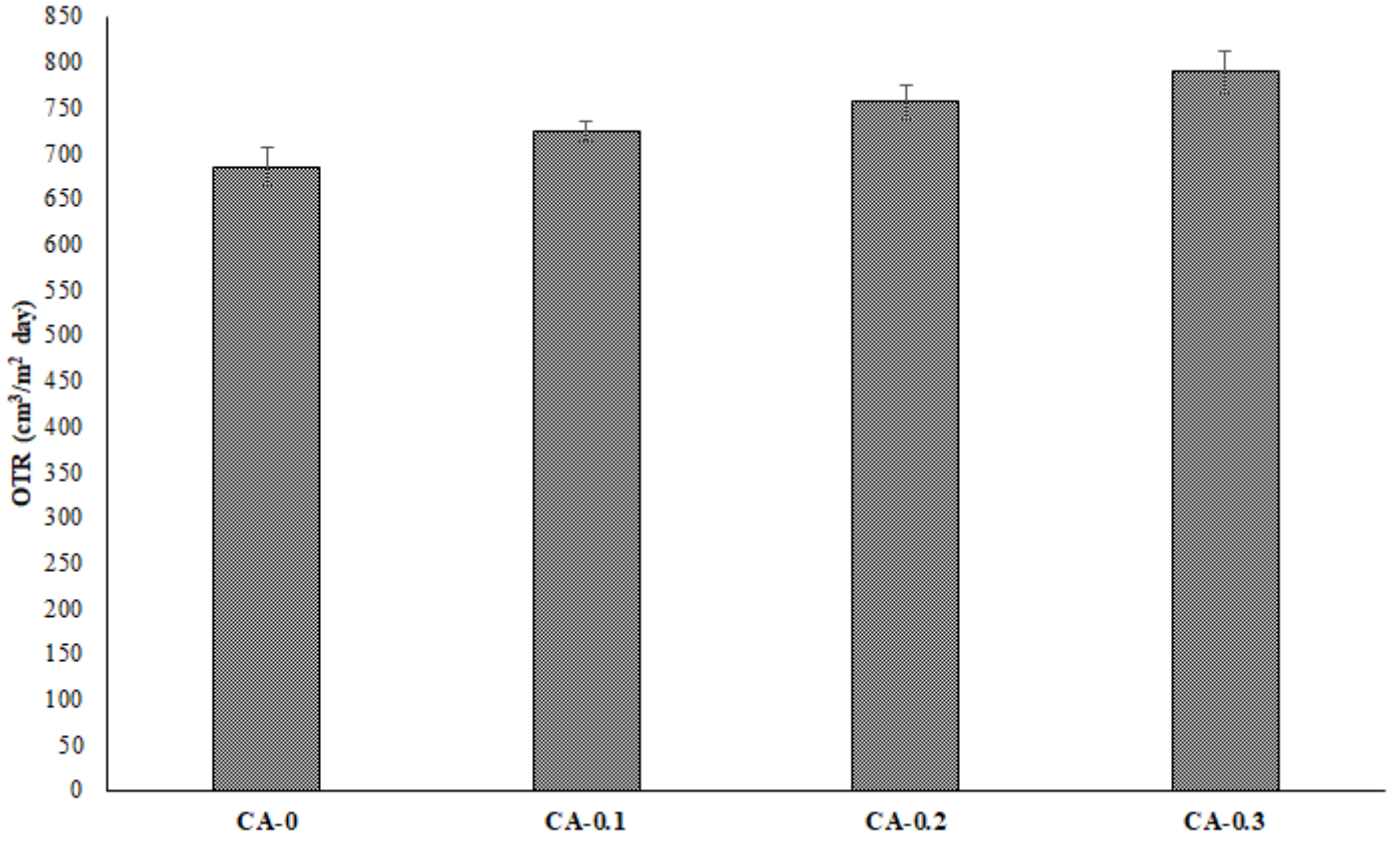


Figure 9

Oxygen permeability of cellulose acetate film containing different Falcaria vulgaris extract concentration (0, 0.1, 0.2 and 0.3%).