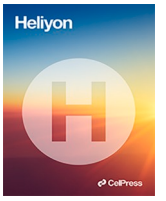
Heliyon 10 (2024) e36895

Contents lists available at ScienceDirect

Heliyon

journal homepage: www.cell.com/heliyon



Research article

Examining the potential of peppermint essential oil-infused pectin and kappa-carrageenan composite films for sustainable food packaging

Saurabh Bhatia [a,b,**,1], Aysha Salim Alhadhrami [a,1], Yasir Abbas Shah [a], Tuba Esatbeyoglu [c], Esra Koca [d], Levent Yurdaer Aydemir [d], Ahmed Al-Harrasi [a,***], Syam Mohan [e,*], Asim Najmi [f], Asaad Khalid [g,****]

[a] *Natural and Medical Sciences Research Center, University of Nizwa, P.O. Box 33, Birkat Al Mauz, 616, Oman*
[b] *School of Health Science, University of Petroleum and Energy Studies, Dehradun, 248007, India*
[c] *Department of Molecular Food Chemistry and Food Development, Institute of Food and One Health, Gottfried Wilhelm Leibniz University Hannover, 30167, Hannover, Germany*
[d] *Department of Food Engineering, Faculty of Engineering, Adana Alparslan Turkes Science and Technology University, Adana, 01250, Turkey*
[e] *Center for Global Health Research, Saveetha Medical College, and Hospitals, Saveetha Institute of Medical and Technical Sciences, Saveetha University, India*
[f] *Department of Pharmaceutical Chemistry and Pharmacognosy, College of Pharmacy, Jazan University, Jazan, 45142, Saudi Arabia*
[g] *Health Research Centre, Jazan University, Jazan, 45142, Saudi Arabia*

ARTICLE INFO

Keywords:
Biopolymer films
Essential oil
Antioxidant
Food packaging

ABSTRACT

Essential oils are key ingredients in the development of edible films and provide a diverse approach to improving food preservation, as well as sensory qualities. The pectin and kappa-carrageenan composite films were obtained by adding peppermint essential oil in different quantities. The films after their fabrication were thoroughly evaluated for their attributes, which included mechanical, barrier, optical, chemical, thermal, and antioxidant properties. The visual assessment of the films demonstrated that PEO-loaded films showed a uniform, homogenous, and slightly yellowish appearance. There was an increase in the thickness (0.045 ± 0.006 to 0.060 ± 0.008 mm), elongation at break (12.73 ± 0.74 to 25.05 ± 1.33 %), and water vapor permeability (0.447 ± 0.014 to 0.643 ± 0.014 (g*mm)/(m^2*h*kPa)) was observed with the addition of PEO. However, tensile strength (45.84 ± 3.69 to 29.80 ± 2.10 MPa) and moisture content (25.83 ± 0.046 to 21.82 ± 0.23 %) decreased with the incorporation of PEO. Furthermore, thermal and antioxidant properties were enhanced by the inclusion of PEO. The presented investigation can be employed to synthesize food packaging material with antioxidant properties with potential applications in food packaging.

* Corresponding author.
** Corresponding author.
*** Corresponding authors.
**** Corresponding author.
E-mail addresses: sbsaurabhbhatia@gmail.com (S. Bhatia), aharrasi@unizwa.edu.om (A. Al-Harrasi), syammohanm@yahoo.com (S. Mohan), drasaad@gmail.com (A. Khalid).
[1] These authors contributed equally to this work.

https://doi.org/10.1016/j.heliyon.2024.e36895
Received 23 May 2024; Received in revised form 23 August 2024; Accepted 23 August 2024
Available online 24 August 2024

1. Introduction

Plastic-based films such as stretch, cling, shrink, and silage films are commonly used in food and agricultural industries [1]. Natural polymers have emerged as promising alternatives to synthetic plastics in the development of edible films, which offer improved biodegradability and minimize plastic waste accumulation. Polysaccharides, proteins, lipids, and their composites have gained attention as suitable materials for these environmentally friendly films due to their natural origin and ease of degradation [2]. Numerous studies have explored the combination of pectin with other polysaccharides to enhance the characteristics of pectin-based edible films [3–5]. Blending pectin with another natural polymer can lead to improved physical and mechanical properties. κ-Carrageenan is particularly promising when blended with pectin, offering the potential for superior film quality and performance [6].

Pectin and κ-carrageenan have been known for their excellent film-forming properties. This composite material has been reported to develop transparent material [7]. Furthermore, pectin contains a high concentration of carboxyl groups, while carrageenan is rich in hydroxyl groups. This combination of functional groups can lead to interactions that potentially enhance the properties of films made from both materials, making them a promising blend for edible film development [8,9]. Thus, this can result in the formation of hydrogen and ionic bonds between two polymers resulting in the formation of stable crosslinking networks with less intermolecular spacing [9]. Such polymeric networks with less intermolecular spacing can show more compactness. Thus, this approach can address the limitation of the low mechanical strength in carrageenan gel [9].

The constant exposure of food to oxygen often triggers degradation reactions resulting in fat and oil rancidity, microbial growth, nutritional loss, and changes in the color, odor, and texture of the food. Several packaging are designed in a way to protect the food from undesirable oxidation reactions [10]. In contrast, to maintain the freshness of living tissues including fresh fruits and vegetables permeability to oxygen and carbon dioxide is critical. Therefore, films that offer an optimum (moderate) barrier with a regulated respiratory exchange rate are more suitable [10]. Natural polysaccharides generally possess less antioxidant properties. Thus, the addition of essential oil is expected to improve antioxidants with improvement in physical and mechanical properties. Recently, several essential oils have been investigated for their antimicrobial activities [11] and potential applications in food packaging [12]. Their natural antimicrobial and antioxidant properties make them attractive candidates for enhancing the safety and shelf-life of packaged foods, particularly when incorporated into biodegradable films. Peppermint oil is derived from the hydro distillation of *Mentha piperita*, containing monoterpenes and mainly menthol and menthone. Peppermint oil contains a high level of phenolic compounds and thus it is considered as a potential natural antioxidant [13,14]. However, its utilization in food packaging has only been investigated in a few studies. Therefore, this study is mainly designed to simultaneously improve the physico-chemical properties of pectin-k-carrageenan films by using peppermint oil as a strong natural antioxidant agent.

2. Materials and methodology

2.1. Fabrication of film samples

Pectin, kappa-carrageenan, and Tween 80 were purchased from Sisco Research Laboratories Pvt. Ltd. (SRL) in India. Glycerol, with a 99.0 % purity, was obtained from BDH Laboratory Supplies (UK). Pectin (1.5 %) and κ-carrageenan (1.5 %) solutions were individually prepared by continuously stirring for 1 h at room temperature. Subsequently, both solutions were mixed in a beaker, and glycerol (0.5 %) was added to the film-forming solution as a plasticizer and mixed continuously for another 30 min at room temperature to ensure uniform dispersion of all the ingredients. After mixing, the solution containing pectin and κ-carrageenan was divided into four labeled beakers. Peppermint essential oil (PEO), identified with Batch No: NNIMPEO/155/0721, was procured from Nature Natural (India). Different concentrations of PEO and tween 80 were added to each beaker according to Table 1. After preparing the solution, 20 mL was transferred into plastic petri plates and left to dry at room temperature for 24 h for the fabrication of films. Once fully dried, the film samples were carefully removed from the plates and subjected to further analysis.

2.2. Microstructure analysis of the films

SEM images were obtained using a JSM-6510 LA instrument (Jeol, Japan) at an accelerating voltage of 20 kV, following the method outlined by Bhatia et al. [15].

Table 1
The ratios of components used for the development of different film samples.

Sample Codes	Pectin (*w/v*)	κ-Carrageenan (*w/v*)	Glycerol (*v/v*)	Peppermint EO (*v/v*)	Tween 80 (*v/v*)
PKO1	1.5 %	1.5 %	0.5 %	–	–
PKO2	1.5 %	1.5 %	0.5 %	0.1 %	0.1 %
PKO3	1.5 %	1.5 %	0.5 %	0.2 %	0.2 %
PKO4	1.5 %	1.5 %	0.5 %	0.3 %	0.3 %

*PKO1 (control film sample without the addition of PEO), PKO2 (loaded with 0.1 % of PEO), PKO3 (loaded with 0.2 % of PEO), and PKO4 (loaded with 0.3 % of PEO).

2.3. Analysis of the film thickness and mechanical properties

The thickness of the film samples was carefully measured using a digital micrometer (model 2046F, Kawasaki, Japan). This was done following the procedure described in the work of Bhatia, Shah [16]. Accurate thickness measurements are crucial in evaluating the uniformity and structural properties of the films. Ten random measurements were taken to calculate the average thickness of each film, in millimeters.

The mechanical attributes of the film samples (PKO1-PKO4) were evaluated following the ASTM D882 standard procedure [17]. The ASTM D882 standard procedure provides a consistent framework for comparing mechanical characteristics across different film formulations. Rectangular strips measuring 70 mm in length and 7 mm in width were securely held between the clamps of a texture analyzer (TA. XT plus, Stable Micro Systems, Godalming, England) and the grip distance was 60 mm. The tensile strength (TS) and elongation at break (EAB) were determined using the Exponent Connect software platform. Higher tensile strength suggests that the film can endure greater force without tearing, making it suitable for packaging products that require more robust protection. A higher elongation at break signifies better flexibility, ideal for wrapping irregularly shaped foods.

2.4. Barrier properties and moisture content analysis

Following the methodology described by Erdem et al. [18], the WVP of the films was determined. The samples underwent a conditioning process within a desiccator for 24 h, during which the humidity was regulated to a constant 50 %. RH of the measurement systems were adjusted using water (RH = 100 %) and silica gel (RH = 0 %). The WVP (measured in g mm m^{-2} h^{-1} kPa^{-1}) was subsequently determined using Equation (1).

$$3.\ WVP = \frac{\Delta m}{\Delta t \times \Delta p \times A} \times d \tag{1}$$

$\Delta m/\Delta t$: This represents the rate of moisture gain over time, measured in grams per day.
Δp: Denotes the difference in water vapor pressure across the film, measured in kilopascals.
A: Represents the surface area of the film, measured in square meters.
d: Indicates the thickness of the film, measured in millimeters.

Furthermore, the gravimetric method was used to determine the moisture content (MC) of the film samples. This method allowed accurate assessment of the moisture content by comparing the weight before and after drying, providing a reliable indicator of the moisture retained in the film samples.

$$MC = \frac{W1 - W2}{W1} \times 100 \tag{2}$$

2.5. Opacity measurement and color properties

The opacity of the films was assessed following the methodology outlined by Zhao et al. [19]. This involved spectrophotometric analysis using an ONDA V-10 Plus spectrophotometer (Italy). Additionally, the color characteristics of the film samples were evaluated using a spectrophotometer CM-5 (1328). The combination of these tools allowed for accurate and detailed measurement of both opacity and color in the film samples.

2.6. Antioxidant analysis

The antioxidant properties of the composite films were evaluated by both DPPH and ABTS assays, as described by Brand-Williams, Cuvelier [20], and Re, Pellegrini [21], respectively. 30 mg film for ABTS and 100 mg film for DPPH analysis were used. The results of both assays were expressed as the percentage inhibition of $ABTS^{+}$ or DPPH radicals, based on the average of three replicates. This approach provided a reliable measure of the antioxidant properties of the films by quantifying their capacity to neutralize free radicals.

2.7. XRD analysis

The X-ray diffraction (XRD) examination was conducted using a Bruker D8 Discover instrument operating at 40 kV. The edible films underwent scanning at a speed of 0.500 s/point within the 2θ range of 5–55°.

2.8. FT-IR spectroscopy analysis

The FT-IR spectra of the films were collected using an InfraRed spectrometer (Tensor 37, Bruker, Ettlingen, Germany). At a resolution of 4 cm^{-1}, the measurements were obtained within the range of 4000 to 400 cm^{-1}. This technique enabled a detailed analysis of the film's molecular composition by identifying the characteristic absorption bands within the specified spectral range.

2.9. Thermal properties

Thermogravimetric analysis (TGA) was performed using a TA Instruments SDTQ600 thermal analyzer (New Castle, DE, USA). The film samples were sealed in an aluminum pan. Each sample, weighing 10 mg, was heated at a rate of 10 °C/min from 25 to 600 °C in a nitrogen-rich atmosphere. This method allowed for the assessment of thermal stability and decomposition patterns of the film samples over a broad temperature range.

2.10. Statistical analysis

The results were shown as mean ± standard deviation (SD) from triplicate findings. One-way analysis of variance (ANOVA) was conducted between means using Duncan's test by SPSS software (V 17.0, IBM Company, Chicago, IL, USA) at the 5 % level of significance.

3. Results and discussion

3.1. Visual appearance

Films were visually observed under an illuminated background to check the presence of any droplets or particles formed while preparing the FFS, casting the FFS, and drying process. Visual inspection of the composite films, both with and without oil, revealed smooth surfaces that were easy to peel off, consistent with the findings of Bhatia et al. [22]. The films that incorporated PEO exhibited a uniform, homogenous, and slightly yellowish appearance with high opacity, as shown in Fig. 1. Films with higher concentrations of PEO showed soft, flexible, more stretchable, and less resistant to breakage. Films without oil (PKO1) were relatively more transparent

Fig. 1. Visual analysis of the film samples PKO1 (control film sample without the addition of PEO), PKO2 (loaded with 0.1 % of PEO), PKO3 (loaded with 0.2 % of PEO), and PKO4 (loaded with 0.3 % of PEO).

than films with oil (PKO2-PKO4).

3.2. Microstructural analysis

The microstructural analysis using SEM, as presented in Fig. 2, displays the surface and cross-sectional structures of all the films. All films demonstrated smooth and homogenous surface structural properties without cracks and pores. The homogenous surface structural properties represent the compatibility between polymers, glycerol, and PEO at the molecular level. However, a slight decrease in the surface roughness was observed in the micrographs of the films with increasing PEO concentration. Nevertheless, no signs of droplet separation from the polymeric matrix were observed. This indicates that PEO dispersed uniformly in the film matrix as observed in the previous studies [23,24]. Film with the highest concentration of PEO showed a more regular, continuous, and compact structure than other films. This is contradictory to the studies where the incorporation of oil causes phase separation, development of heterogeneous structure, holey structure, and increase in the surface coarseness due to the migration of oil droplets towards the surface due to volatile nature [25]. The formation of a more compact, smooth, and regular structure at higher concentrations of PEO may result from the crosslinking effect of the phenolic components present in PEO [26]. These components can enhance the structural integrity of the films, leading to improved uniformity and organization.

3.3. Film thickness, mechanical, and barrier properties

The effect of incorporating PEO on the film thickness is shown in Table 2. The film thickness varied between 0.045 and 0.060 mm, significantly increasing ($p \leq 0.05$) as the PEO concentration rose. This increase in thickness could be attributed to the higher solid content, as observed in previous studies [27,28].

Table 2 also represents values for TS and EAB of the films obtained from the κ-carrageenan and pectin polymer infused with various concentrations of PEO. As per the findings obtained, the incorporation of PEO reduced the TS of films, whereas EAB increased significantly ($p < 0.05$). These results were in line with the visual assessment where films with the highest concentration of PEO were soft, flexible, more stretchable, and less resistant to breakage (Fig. 1). This effect may be due to PEO's role in reducing the intra- and intermolecular interactions. The addition of oil resulted in the incomplete replacement of the stronger intermolecular polymer interactions with weaker polymer-oil interactions in the film. This change affected the structural integrity of the film, potentially leading to variations in its properties [29,30]. The current findings were in line with the reported studies and results obtained from XRD analysis that showed a decrease in the crystallinity with an increase in the concentration of PEO (Fig. 3) [31–33]. Moreover, this mechanical behavior of the films could be due to the plasticization effect of PEO that may have improved polymer chains mobility as well as the flexibility of the film, by this means improving the EAB of the films [34].

The results of water vapor permeability of all the films varied between 0.447 and 0.643 (g*mm)/(m^2*h*kPa) (Table 2). The incorporation of PEO significantly increased the permeability of films against water vapors. This behavior could be due to changes in structure or hydrophilic-hydrophobic ratio of film components that affected the water vapor transfer process. WVP of the films is dependent on several factors such as the nature of the additive and its concentration, its distribution, microstructural arrangements, and crystallinity of the films [35]. As it was noted in previous research, adding oil to the films made them more hydrophobic, which raised the amount of free space between the polymeric chains and decreased the crystallinity of the films. In our studies, the crystallinity of the films decreased with the addition of oil (Fig. 3). Therefore, the overall impact of oil incorporation on the films could have been nullified. Studies by Di Giuseppe et al. [35] found similar results for films made from chitosan, sodium caseinate, and

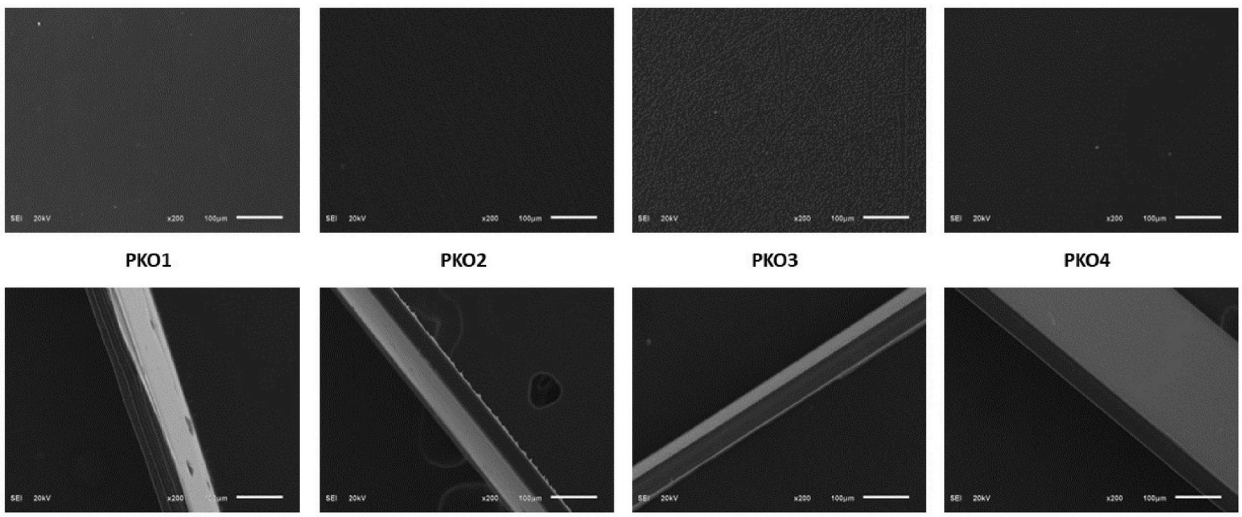


Fig. 2. Micrographs of the films loaded with and without PEO (scale 100 μm).

Table 2
Thickness, mechanical properties, water permeation, and moisture percentage mean values of film samples. The ± sign means standard deviations.

Film samples	Thickness (mm)	EAB (%)	TS (MPa)	WVP ((g*mm)/(m2*h*kPa))	MC (%)
PKO1	0.045 ± 0.006[a]	12.73 ± 0.74[a]	45.84 ± 3.69[a]	0.447 ± 0.014[a]	25.83 ± 0.046[a]
PKO2	0.048 ± 0.005[a]	19.48 ± 1.40[b]	35.04 ± 2.48[b]	0.486 ± 0.007[b]	25.22 ± 0.31[a]
PKO3	0.058 ± 0.010[a]	19.99 ± 1.12[b]	29.74 ± 1.83[c]	0.550 ± 0.014[c]	21.99 ± 1.11[b]
PKO4	0.060 ± 0.008[a]	25.05 ± 1.33[c]	29.80 ± 2.10[c]	0.643 ± 0.014[d]	21.82 ± 0.23[b]

* Values represented by distinct letters (e.g., a, b, c, and d) within a column signify notable differences ($p < 0.05$).

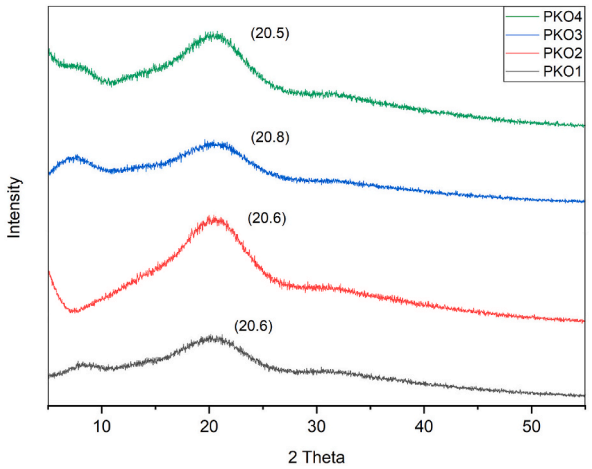


Fig. 3. XRD analysis of the film samples loaded with and without PEO. PKO1 (control), PKO2 (loaded with 0.1 % of PEO), PKO3 (loaded with 0.2 % of PEO) and PKO4 (loaded with 0.3 % of PEO).

rosemary essential oil. They discovered that the permeability of these materials was not just influenced by the addition of lipophilic components but also by the microstructure of the films. This was significantly impacted by the physical state of the essential oil and how it was distributed in the polymer matrix [36,37]. The state of the EO and its dispersion affects the structural integrity and barrier properties of the film. Thus, grasping the interplay between essential oils and polymers is crucial to refining film formulations, as microstructural differences can alter permeability, mechanical strength, and overall functionality. This increase in WVP values with an increase in oil concentration could be due to the discontinuation caused in the polymeric structure by lipid droplets, triggering a loss in film cohesion. This could also be due to the changes in structure or hydrophilic-hydrophobic ratio of film components [38].

3.4. Moisture content

The sensitivity of the films against water vapors was examined by moisture content. Table 2 presents the moisture content levels in all the films. It was noticed that as the PEO concentration increased, the films' moisture content reduced. This could be due to the increase in the interaction between phenolic components of PEO and polymer, thus reducing the availability of polar amino as well as hydroxyl groups. This chemical interaction reduced hydrogen bond interactions between the polymer and the water, resulting in a decrease in the moisture content of the films. The findings obtained in the current study align with the reported results in the literature [39,40].

3.5. Color attributes and opacity of the fabricated films

Optical analysis is a critical factor influencing consumer acceptance of edible films. Table 3 provides optical data for all the films.

Table 3
Mean values of the transparency and color attributes of the films. Mean (n = 3) ± sign standard deviation.

Film samples	Opacity	L	a*	b*	ΔE
PKO1	1.38 ± 0.07[a]	96.09 ± 0.02[a]	−0.01 ± 0.00[a]	0.54 ± 0.03[a]	0.48 ± 0.02[a]
PKO2	4.67 ± 0.32[b]	97.52 ± 0.11[b]	0.24 ± 0.06[b]	0.98 ± 0.09[b]	1.88 ± 0.12[b]
PKO3	11.50 ± 0.24[c]	97.25 ± 0.31[b]	0.23 ± 0.15[b]	0.91 ± 0.28[b]	1.62 ± 0.43[bc]
PKO4	13.26 ± 0.86[d]	97.31 ± 0.58[b]	0.62 ± 0.04[c]	1.64 ± 0.11[c]	2.25 ± 0.24[c]

In each column, the letters a, b, c, and d correspond to certain values that indicate statistically significant differences ($p < 0.05$). L stands for lightness, a for green-red, b* for blue-yellow, and ΔE* for total color variation.

Films containing PEO exhibited elevated b values and ΔE, which increased as the PEO concentration rose. This phenomenon may be attributed to the phenolic components in PEO, as detailed in a previous study [41]. The opacity results indicated significant differences across the films, ranging from 1.38 to 13.26 (as shown in Table 3). Films with higher PEO concentrations appeared opaque, likely due to increased light scattering by oil droplets within the film matrix. A higher oil concentration can lead to an increase in both the size and number of droplets, resulting in more opaque films [42].

3.6. Antioxidant properties of the fabricated films

Table 4 shows the antioxidant properties of films that were assessed by $ABTS^{\bullet+}$ and DPPH radical scavenging assays. This assessment helped in determining the capacity to scavenge free radicals. The control film showed antioxidant effects probably due to the presence of κ-carrageenans [43]. Our findings indicated that the DPPH and ABTS scavenging activities of the films exhibited a significant increase ($p < 0.05$) with the rising PEO concentrations as shown in Table 4. The previous study suggested that the antioxidant effects of essential oils could be attributed to their phenolic content [44]. The increase in the antioxidant effect of essential oil-loaded films was expected due to the presence of menthol, menthone, 1,8-cineole, and neo-menthol as reported in the previous study [13]. When compared to the control film, the antioxidant capacity of the films improved with an increase in PEO content, indicating the effectiveness of PEO as a natural antioxidant. These results are consistent with the earlier research [45].

3.7. XRD analysis

Various studies have reported the plasticization effect of essential oil, however crosslinking effect of the oil has been also reported [22,26,34]. The crystallinity of the films directly impacts the mechanical strength of the polymeric films. Thus, it is important to understand the effect of essential oil on the crystallinity of the films. Fig. 3 illustrates the XRD diffractograms of all fabricated film samples. All films did not show any characteristic crystalline peak due to the amorphous nature of pectin and κ-carrageenan present in the film. All films with and without oil showed broad diffraction peaks at 7° and 20°, which could be attributed to kappa carrageenan and pectin as observed in the previous study [46]. The addition of oil showed variations in the intensities and changes in the peak positions which could be due to the addition of PEO.

According to the XRD curves, incorporating PEO decreases the crystallinity of the films, as shown in Fig. 3. This behavior aligns with findings reported in the literature [47]. The reduction in the peak intensities after PEO incorporation could be due to the decrease in hydrogen bond formation, resulting in a decrease in regular domains, compactness, and film crystallinity. These results were in accordance with mechanical studies where TS was reduced and EAB was increased with the addition of oil (Table 2). This plasticization effect of oil increased the free volume and chain mobility by reducing hydrogen bonding. In the earlier studies due to the plasticization effect of oil, a drop in crystallinity of the films was observed at higher concentrations of oil in the films, resulting in an increase in the broadness of the peaks, decrease in the intensity and shifting/disappearance of the peaks [24,48,49]. However essential oil components also act as crosslinkers by improving hydrogen bonding increasing the crystallinity of the films as observed in the previous studies [34,50,51].

3.8. FTIR analysis

FTIR analysis identifies chemical compounds by measuring their infrared absorption spectra [52]. The FTIR analysis was conducted to investigate the chemical interactions among the functional groups of the film-forming components. Fig. 4 illustrates the FTIR spectrum, displaying characteristic peaks. Peaks were observed at 844, 917, 1034, 1250, 1735, 2354, 2923, and 3343 cm−1. The peaks at 844 and 917 cm−1 suggest the presence of C=C bending in alkene. Furthermore, the peak at 1034 cm−1 is indicative of the glycosidic bond. Peaks at 1735 cm−1 suggest stretching of the carbonyl group (C=O) in carboxylic acids, while those at 2360 cm−1 indicate stretching of carbon dioxide (O=C=O). The peak observed at 2924 cm−1 signifies the stretching of C-H bonds (alkanes). The broad band observed at 3343 cm^{-1} corresponds to the O–H stretching, likely originating from hydroxyl groups present in biopolymers and water molecules [53]. The FTIR analysis indicates that the interactions between peppermint essential oil and the films involve primarily hydrogen bonding. The phenolic compounds in peppermint oil likely form hydrogen bonds with the hydroxyl groups of pectin and carrageenan, enhancing or modifying the film properties. A slight difference in peak intensity was noted, which was probably caused by the addition of PEO, however as the PEO concentrations within the film matrix varied, the intensity changed.

Table 4
Free radical scavenging activity of the film samples. The ± sign means standard deviations.

Film samples	DPPH (% inhibition)	ABTS (% inhibition)
PKO1	13.92 ± 0.26^{a}	33.29 ± 0.47^{a}
PKO2	22.56 ± 0.21^{b}	38.51 ± 0.33^{b}
PKO3	27.72 ± 0.30^{c}	39.98 ± 0.41^{c}
PKO4	38.15 ± 0.53^{d}	44.35 ± 0.72^{d}

* Values represented by distinct letters (e.g., a, b, c, and d) within a column signify notable differences ($p < 0.05$).

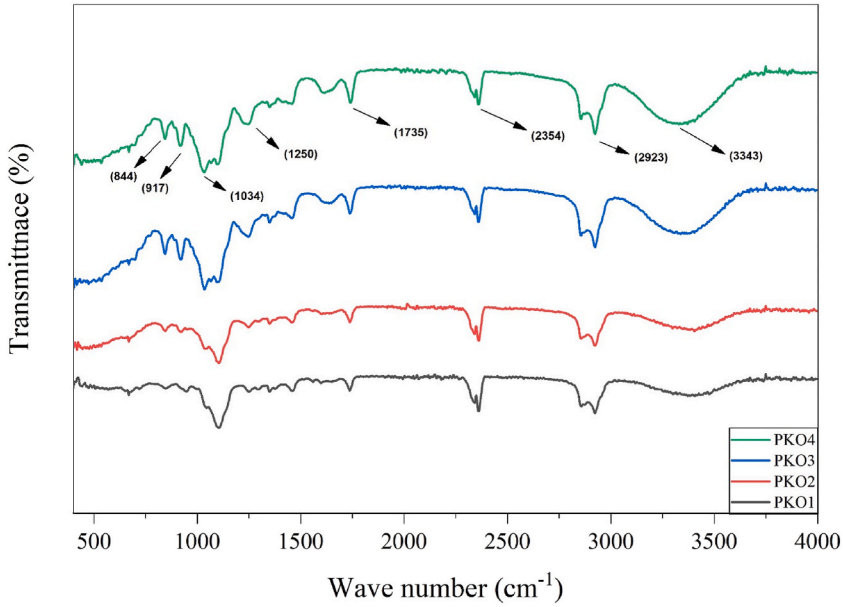


Fig. 4. FTIR analysis of the films. PKO1 (control), PKO2 (loaded with 0.1 % of PEO), PKO3 (loaded with 0.2 % of PEO), and PKO4 (loaded with 0.3 % of PEO).

3.9. TGA analysis of the films

TGA analysis in a temperature range of 25–600 °C was performed to assess the thermal stability of flexible packaging material that is usually sensitive to heat and to understand how the interaction between the polysaccharides and additives could affect stability. Fig. 5 demonstrates thermograms representing thermal events for all composite films and multiple steps of weight loss. The first stage (35–130 °C) may be attributed to water evaporation (free or bound) [34]. The second stage (160–260 °C) is usually attributed to the volatilization of glycerol [34]. The third stage at 290–450 °C is related to polysaccharide decomposition [22,54]. According to TGA assessments, adding PEO increased the film's thermal resistance. The better compatibility and stronger interactions between the various components of the oil-loaded films may be the cause of their increased thermal stability [55,56].

4. Conclusion

Incorporating peppermint essential oil (PEO) into pectin and κ-carrageenan composite films has shown substantial potential for enhancing key properties relevant to food packaging applications. These films exhibited improved antioxidant properties, as demonstrated by DPPH and ABTS assays, highlighting their capacity to extend the shelf life of packaged foods. The visual assessment and scanning electron microscopy analysis also emphasized the uniformity and structural integrity of PEO-loaded films, particularly at higher concentrations. Additionally, the thermal resistance of the films was improved with the addition of PEO as demonstrated by TGA. These results suggest that PEO effectively enhances the quality and functionality of edible films for food packaging.

Funding

The authors extend their appreciation to the Deputyship for Research & Innovation, Ministry of Education in Saudi Arabia for funding this research work through the project number (ISP23–81).

CRediT authorship contribution statement

Saurabh Bhatia: Writing – original draft, Investigation, Data curation, Conceptualization. **Aysha Salim Alhadhrami:** Writing – original draft, Resources, Methodology. **Yasir Abbas Shah:** Investigation, Formal analysis, Data curation. **Tuba Esatbeyoglu:** Writing – review & editing. **Esra Koca:** Writing – original draft, Software, Resources. **Levent Yurdaer Aydemir:** Writing – original draft, Resources, Project administration. **Ahmed Al-Harrasi:** Writing – original draft, Supervision, Resources. **Syam Mohan:** Writing – review & editing, Writing – original draft, Conceptualization. **Asim Najmi:** Writing – review & editing, Writing – original draft, Methodology, Investigation. **Asaad Khalid:** Writing – review & editing, Writing – original draft, Visualization, Supervision.

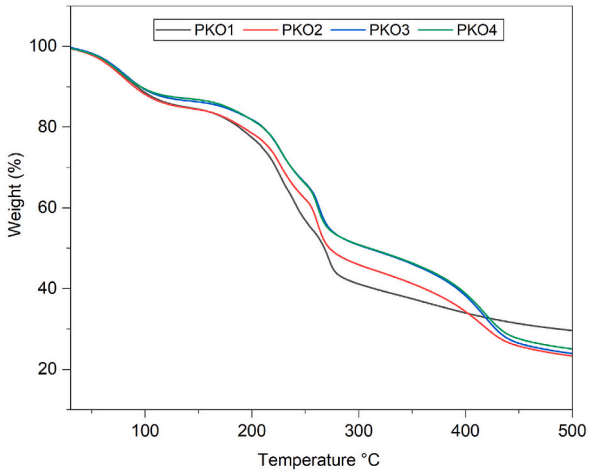


Fig. 5. TGA analysis of the films. PKO1 (control), PKO2 (loaded with 0.1 % of PEO), PKO3 (loaded with 0.2 % of PEO), and PKO4 (loaded with 0.3 % of PEO).

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: One of the corresponding authors in this manuscript is serving in editorial capacity for the Heliyon Journal. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

The Authors are thankful to the Natural and Medical Sciences Research Center, University of Nizwa, Oman, for providing research facilities to conduct the current study. The authors extend their appreciation to the Deputyship for Research & Innovation, Ministry of Education in Saudi Arabia for funding this research work through the project number (ISP23–81).

References

[1] S. Vallejos, et al., From classical to advanced use of polymers in food and beverage applications, Polymers 14 (22) (2022) 4954.
[2] D. Xu, T. Chen, Y. Liu, The physical properties, antioxidant and antimicrobial activity of chitosan–gelatin edible films incorporated with the extract from hop plant, Polym. Bull. 78 (2021) 3607–3624.
[3] T.M.P. Ngo, et al., Characteristics and antimicrobial properties of active edible films based on pectin and nanochitosan, Int. J. Mol. Sci. 21 (6) (2020) 2224.
[4] R. Priyadarshi, S.-M. Kim, J.-W. Rhim, Pectin/pullulan blend films for food packaging: effect of blending ratio, Food Chem. 347 (2021) 129022.
[5] F. Azizah, et al., Development of edible composite film from fish gelatin–pectin incorporated with lemongrass essential oil and its application in chicken meat, Polymers 15 (9) (2023) 2075.
[6] M. Zioga, et al., Characterization of pectin and carrageenan edible films in the presence of lemon balm infusion, Food Hydrocolloids 150 (2024) 109679.
[7] V.D. Alves, et al., Barrier properties of carrageenan/pectin biodegradable composite films, Procedia Food Science 1 (2011) 240–245.
[8] C. Lara-Espinoza, et al., Pectin and pectin-based composite materials: beyond food texture, Molecules 23 (4) (2018) 942.
[9] F. Yu, et al., κ-Carrageenan/Sodium alginate double-network hydrogel with enhanced mechanical properties, anti-swelling, and adsorption capacity, Chemosphere 237 (2019) 124417.
[10] E. Ayranci, S. Tunc, A method for the measurement of the oxygen permeability and the development of edible films to reduce the rate of oxidative reactions in fresh foods, Food Chem. 80 (3) (2003) 423–431.
[11] A.M. El-Mesallamy, et al., Antioxidant, antimicrobial activities and volatile constituents of clove flower buds oil, Journal of essential oil bearing plants 15 (6) (2012) 900–907.
[12] R. Ribeiro-Santos, et al., Use of essential oils in active food packaging: recent advances and future trends, Trends Food Sci. Technol. 61 (2017) 132–140.
[13] Z. Wu, et al., Chemical composition and antioxidant properties of essential oils from peppermint, native spearmint and scotch spearmint, Molecules 24 (15) (2019) 2825.
[14] N.R. Desam, et al., Chemical constituents, in vitro antibacterial and antifungal activity of Mentha× Piperita L.(peppermint) essential oils, J. King Saud Univ. Sci. 31 (4) (2019) 528–533.
[15] S. Bhatia, et al., Development, characterization, and assessment of antioxidant pectin–sodium alginate based edible films incorporated with cassia essential oil, Int. J. Food Sci. Technol. 58 (9) (2023) 4652–4665.
[16] S. Bhatia, Y.A. Shah, A. Al-Harrasi, S. Ullah, M.K. Anwer, E. Koca, L.Y. Aydemir, M.R. Khan, A novel film based on a cellulose/sodium alginate/gelatin composite activated with an ethanolic fraction of Boswellia sacra oleo gum resin, Food Sci. Nutrition 12 (2) (2024) 1056–1066.
[17] ASTM, Standard test methods for tensile properties of thin plastic sheeting, method D882-10. https://doi.org/10.1520/D0882-10, 2010.
[18] B. Gökkaya Erdem, S. Dıblan, S. Kaya, Development and structural assessment of whey protein isolate/sunflower seed oil biocomposite film, Food and Bioprod. Process. 118 (2019) 270–280, https://doi.org/10.1016/j.fbp.2019.09.015.
[19] J. Zhao, Y. Wang, C. Liu, Film transparency and opacity measurements, Food Anal. Methods 15 (10) (2022) 2840–2846.
[20] W. Brand-Williams, M.-E. Cuvelier, C. Berset, Use of a free radical method to evaluate antioxidant activity, LWT–Food Sci. Technol. 28 (1) (1995) 25–30.
[21] R. Re, et al., Antioxidant activity applying an improved ABTS radical cation decolorization assay, Free Radic. Biol. Med. 26 (9–10) (1999) 1231–1237.
[22] S. Bhatia, et al., Structural, mechanical, barrier and antioxidant properties of pectin and xanthan gum edible films loaded with grapefruit essential oil, Heliyon 10 (3) (2024) e25501.

[23] L.J. Pérez-Córdoba, et al., Physico-chemical, antimicrobial and antioxidant properties of gelatin-chitosan based films loaded with nanoemulsions encapsulating active compounds, Food Hydrocolloids 79 (2018) 544–559.
[24] S.F. Hosseini, et al., Development of bioactive fish gelatin/chitosan nanoparticles composite films with antimicrobial properties, Food Chem. 194 (2016) 1266–1274.
[25] L. Sánchez-González, et al., Effect of essential oils on properties of film forming emulsions and films based on hydroxypropylmethylcellulose and chitosan, J. Food Eng. 105 (2) (2011) 246–253.
[26] L.D. Daza, et al., Effect of essential oils from lemongrass and Tahiti lime residues on the physicochemical properties of chitosan-based biodegradable films, Foods 12 (9) (2023) 1824.
[27] S.-M. Hasheminya, et al., Development and characterization of biocomposite films made from kefiran, carboxymethyl cellulose and Satureja Khuzestanica essential oil, Food Chem. 289 (2019) 443–452.
[28] S.M. Gonçalves, et al., Functional and antimicrobial properties of cellulose acetate films incorporated with sweet fennel essential oil and plasticizers, Curr. Res. Food Sci. 3 (2020) 1–8.
[29] Y.A. Arfat, et al., Properties and antimicrobial activity of fish protein isolate/fish skin gelatin film containing basil leaf essential oil and zinc oxide nanoparticles, Food Hydrocolloids 41 (2014) 265–273.
[30] Z. Shen, D.P. Kamdem, Development and characterization of biodegradable chitosan films containing two essential oils, Int. J. Biol. Macromol. 74 (2015) 289–296.
[31] Y. Zhang, et al., Preparation and characterization of curdlan/polyvinyl alcohol/thyme essential oil blending film and its application to chilled meat preservation, Carbohydr. Polym. 247 (2020) 116670.
[32] A. Dashipour, et al., Antioxidant and antimicrobial carboxymethyl cellulose films containing Zataria multiflora essential oil, Int. J. Biol. Macromol. 72 (2015) 606–613.
[33] T. Nisar, et al., Characterization of citrus pectin films integrated with clove bud essential oil: physical, thermal, barrier, antioxidant and antibacterial properties, Int. J. Biol. Macromol. 106 (2018) 670–680.
[34] Y. Zhou, et al., Effects of cinnamon essential oil on the physical, mechanical, structural and thermal properties of cassava starch-based edible films, Int. J. Biol. Macromol. 184 (2021) 574–583.
[35] F.A. Di Giuseppe, et al., Physical properties of active biopolymer films based on chitosan, sodium caseinate, and rosemary essential oil, Food Packag. Shelf Life 32 (2022) 100817.
[36] M. Ahmad, et al., Physico-mechanical and antimicrobial properties of gelatin film from the skin of unicorn leatherjacket incorporated with essential oils, Food Hydrocolloids 28 (1) (2012) 189–199.
[37] J. Bonilla, et al., Effect of essential oils and homogenization conditions on properties of chitosan-based films, Food Hydrocolloids 26 (1) (2012) 9–16.
[38] S.F. Hosseini, et al., Bio-based composite edible films containing Origanum vulgare L. essential oil, Ind. Crop. Prod. 67 (2015) 403–413.
[39] L. Wang, et al., Preparation and characterization of active films based on chitosan incorporated tea polyphenols, Food Hydrocolloids 32 (1) (2013) 35–41.
[40] T. Liu, et al., Development and characterization of novel active chitosan films containing fennel and peppermint essential oils, Coatings 10 (10) (2020) 936.
[41] S. Benavides, R. Villalobos-Carvajal, J. Reyes, Physical, mechanical and antibacterial properties of alginate film: effect of the crosslinking degree and oregano essential oil concentration, J. Food Eng. 110 (2) (2012) 232–239.
[42] L. Sanchez-Gonzalez, et al., Characterization of edible films based on hydroxypropylmethylcellulose and tea tree essential oil, Food Hydrocolloids 23 (8) (2009) 2102–2109.
[43] T. Sun, et al., Degradation and antioxidant activity of κ-carrageenans, J. Appl. Polym. Sci. 117 (1) (2010) 194–199.
[44] B. Shan, et al., Antioxidant capacity of 26 spice extracts and characterization of their phenolic constituents, J. Agric. Food Chem. 53 (20) (2005) 7749–7759.
[45] S. Shojaee-Aliabadi, et al., Characterization of antioxidant-antimicrobial κ-carrageenan films containing Satureja hortensis essential oil, Int. J. Biol. Macromol. 52 (2013) 116–124.
[46] Y. Liu, et al., Preparation of pH-sensitive and antioxidant packaging films based on κ-carrageenan and mulberry polyphenolic extract, Int. J. Biol. Macromol. 134 (2019) 993–1001.
[47] S. Bhatia, et al., Novel applications of black pepper essential oil as an antioxidant agent in sodium caseinate and chitosan based active edible films, Int. J. Biol. Macromol. (2023) 128045.
[48] C.D. Grande Tovar, et al., Synthesis, characterization, and histological evaluation of chitosan-Ruta graveolens essential oil films, Molecules 25 (7) (2020) 1688.
[49] C. Valenzuela, L. Abugoch, C. Tapia, Quinoa protein–chitosan–sunflower oil edible film: mechanical, barrier and structural properties, LWT–Food Sci. Technol. 50 (2) (2013) 531–537.
[50] J.T. Martins, et al., Synergistic effects between κ-carrageenan and locust bean gum on physicochemical properties of edible films made thereof, Food Hydrocolloids 29 (2) (2012) 280–289.
[51] S. Bhatia, Y. Abbas Shah, A. Al-Harrasi, M. Jawad, E. Koca, L.Y. Aydemir, Enhancing tensile strength, thermal stability, and antioxidant characteristics of transparent kappa carrageenan films using grapefruit essential oil for food packaging applications, ACS omega 9 (8) (2024) 9003–9012.
[52] S.S. Moni, et al., Phytochemical and spectral analysis of the methanolic extracts of leaves of Murraya koenigii of Jazan, Saudi Arabia, Nat. Prod. Res. 35 (15) (2021) 2569–2573.
[53] A. Farhan, N.M. Hani, Characterization of edible packaging films based on semi-refined kappa-carrageenan plasticized with glycerol and sorbitol, Food Hydrocolloids 64 (2017) 48–58.
[54] F. Zhou, et al., Preparation and characterization of biodegradable κ-carrageenan based anti-bacterial film functionalized with Wells-Dawson polyoxometalate, Foods 11 (4) (2022) 586.
[55] T. Xu, et al., Cinnamon and clove essential oils to improve physical, thermal and antimicrobial properties of chitosan-gum Arabic polyelectrolyte complexed films, Carbohydr. Polym. 217 (2019) 116–125.
[56] R. Mo Syafiq, S.M. Sapuan, M.R. Zuhri, Effect of cinnamon essential oil on morphological, flammability and thermal properties of nanocellulose fibre–reinforced starch biopolymer composites, Nanotechnol. Rev. 9 (1) (2020) 1147–1159.