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ORIGINAL PAPER

Development of Biodegradable Films with Improved Antioxidant Properties Based on the Addition of Carrageenan Containing Olive Leaf Extract for Food Packaging Applications

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Abstract
This study aims at evaluating the bioactive compounds from olive leaf extract and to develop biodegradable carrageenan films with antioxidant properties by incorporating varying concentrations of olive leaf extract. The olive leaf extract obtained by MAE solvent-free had high antioxidant activity and has great potential for use as functional ingredient in food packaging. The results of thickness of biofilms were in the range of 0.097 to 0.162 mm and showed significant difference. Despite the addition of extract into biofilm resulted in a slight increase in the stretching capacity, reduction in its tensile strength and a higher water vapor permeability, the biodegradable films based on carrageenan containing olive leaf exhibited good barrier properties and mechanical properties. The total phenolic compounds and antioxidant activity of films significantly increased with an increase in olive leaf extract concentration, which means that the process to obtain the films does not provoke a degradation of these bio compounds. The incorporation of natural antioxidants appears to be a potential strategy to add additives into packaging material suitable for food products.

Keywords Biodegradable · Phenolic compounds · Antioxidant activity

Introduction

Olive leaves (*Olea europea* L.) are usually considered as one of the byproducts of olive grove farming and can be found in large amounts during the pruning of the trees (approximately 25% by weight) and during harvesting (more than 10% of the total weight of the olives) [1]. It has been reported that olive leaves are considered waste residue; however, they have a huge potential for value addition through recovery of phytochemicals which can be used in food and pharmaceutical products. Many studies have been published about the functional properties of olive leaf extracts due to the presence of phenolic compounds like oleuropein (OLE) and hydroxytyrosol (HYD) [2–5].

Aimed at the reduction of the use of chemical additives in the food industry, there has been a growing interest in the use of natural additives with antioxidant properties obtained from plant extracts as alternatives. However, in most cases, direct addition of natural compounds may adversely affect the palatability of food products. The incorporation of these natural antioxidants into films appears to be a good strategy to achieve gradual liberation of the additives into the food throughout its shelf life [6, 7]. The environmental impact of using synthetic packaging materials as vehicle for value addition has led researchers to develop biodegradable alternatives that can be used as substitutes for the current synthetic polymers [8]. Furthermore, the biopolymer based packaging films offers several advantages due to their good biodegradability, biocompatibility, environmental-friendliness, and even edibility [9]. Biopolymer films have been generally classified according to the source of the original

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polymer utilized. Carrageenans are natural and water soluble sulfated polysaccharides extracted from red seaweeds [10–12]. The differences in the chemical structure of carrageenans are essential for their physicochemical properties and the helical structure formation leading to varied applications [13]. Park [14] and Varela and Fiszman [15] reported that Kappa-carrageenan is able to produce a clear film with excellent mechanical and structural properties. It generally has high tensile strength and acts as a good barrier against oxygen.

In recent years, researchers have used olive leaf extract for producing films intended for food preservation based on polypropylene, Ecoflex-polylactic acid (PLA), polyethylene and gelatin [6, 16–18]. However, the literature does not show results of studies using carrageenan biodegradable films containing olive leaves extract to obtain an antioxidant food packaging. The present study aims at evaluating the properties of olive leaf extract and to develop biodegradable films with antioxidant properties by incorporating varying concentrations of olive leaf extract into carrageenan film.

Materials and Methods

Materials and Reagents

Kappa-carrageenan was used as biopolymeric matrix while glycerol (analytical grade) was used as a plasticizer. During the experimentation following chemicals were used: 2,2-diphenyl-1-picrylhydrazyl (DPPH), Folin Ciocalteu's phenol reagent, anhydrous sodium carbonate and gallic acid which were of analytical grade. Water, acetonitrile, acetic acid, oleuropein and hydroxytyrosol of HPLC grade were also used. All the chemicals were purchased from Sigma Aldrich (St. Louis, USA) unless specified otherwise.

Olive Leaf Extract Preparation

The olive leaves (*Olea Europaea* L.) cultivar Arbequina were collected from a private farm Estância Guarda Velha, located in Pinheiro Machado, Rio Grande do Sul, Brazil (31°30′04.0"S, 53°30′42.0"W), in late June 2017. The olive leaves were washed in running water, then dipped in commercial solution of 2% sodium hypochlorite and rinsed in sterilized distilled water. The leaves were then sorted by hand and diseased leaves were removed. The initial moisture content of the olive leaves was 51.65% (w.b.) they were dried for 24 h in a forced air oven operated at 40 °C. After drying, the leaves had a moisture content of 3.1% (w.b.) and they were kept in vacuum sealed opaque bags. Prior to analysis, the leaves were ground to a fine powder using a domestic blender (NutriBullet, USA). The fraction retained at 60-mesh sieve was used to ensure uniform particle size.

Microwave-assisted extraction (MAE) was performed at optimized conditions from a previous study. Extraction was carried out with 0.5 g of ground material in 25 mL of distilled water, used as solvent. The MAE of bioactive compounds from olive leaves was performed in a multi-mode (closed) Mini-WAVE microwave unit (SCP Science, Canada). The system consists of a touchscreen controller which is USB-connected with the module. The frequency and power of irradiation were 2.45 GHz and 1000 W; the duration of irradiation included: ramp time (time to reach the target process temperature) and hold time (elapsed time while irradiating sample at set temperature). The module houses the non-rotating digestion rack which holds six equidistant and radially-constructed 75 mL tubes. Tubes are made of quartz and they were used throughout the experimentation. Six infrared (IR) sensors, located on the side walls of the oven, provide real-time temperature monitoring of each sample temperature, with their average reported as the operating temperature. The magnetron is located below the floor to ensure even distribution of the MW energy across the digestion chamber. The unit had a forced air ventilation system for cooling. Optimized conditions for the MAE were 86 °C and 3 min with a ramp time set at 5 min [19]. After extraction process, the extract containing the solvent and leaves powder mixture was subjected to vacuum filtration using Whatman 4 filter paper (Sigma Aldrich, St. Louis, USA) and then characterized.

Olive Leaf Extract Characterization

Total Phenolics Quantification (TP)

A spectrophotometric method adapted from the work by Singleton and Rossi [20] was used to quantify total phenolics content with Folin-Ciocalteu reagent and gallic acid standard. For the determination, 0.5 mL of extract solution was mixed with 10 mL of distilled water and 1 mL Folin-Ciocalteu in a 50-mL centrifuge tube. After 5 min, 8 mL of 7.5% (w/v) aqueous solution of sodium carbonate was added. The mixture was vortexed (Corning LSE) and stored in the dark at room temperature for two hours. Then the absorbance of the mixture was measured at a wavelength of 765 nm with a spectrophotometer (Ultraspec1000, Amersham Pharmacia Biotech, USA). The total phenolics was quantified using a standard curve of absorbance obtained with concentrations of gallic acid ranging from 70 to 1800 mg L^{-1}. The measured concentration of total phenolics in the olive leaves was finally expressed as milligrams of gallic acid equivalent per gram of dry matter. All the data reported was the average of triplicate analyses.

Antioxidant Activity Quantification (AA)

Antioxidant activity was determined with the method developed by Brand-Williams et al. [21]. DPPH solution with 6×10^{-5} M concentration was prepared with DPPH standard and HPLC-grade methanol. One hundred microliters solvent solution considered as blank or extract solution was mixed with 3 mL DPPH solution and kept for 30 min in the dark at room temperature. The absorbance of blank as well as solutions with extracts were measured using a spectrophotometer (Ultraspec 2100 pro, Biochrom Ltd., Cambridge, UK) at 517 nm. The analysis was conducted in triplicate. The percentage of free radicals scavenged by DPPH radical was calculated using Eq. 1.

$$AA(\%) = \left(\frac{A_{control} - A_{sample}}{A_{control}} \right) \times 100 \tag{1}$$

where $A_{control}$ is the absorbance of the blank solution and A_{sample} is the absorbance of the extract solution.

Oleuropein (OLE) and Hydroxytyrosol (HYD) Quantification

HPLC analyses for identification and quantification of oleuropein and hydroxytyrosol were done with an Agilent 1100 series instrument (Santa Clara, USA), equipped with variable wavelength detector. The chromatographic method used for the analyses was the isocratic elution procedured. The separation was conducted at 25 °C using a reversed phase Discovery column (Supelco) RP C18 (5 µm, 25 cm × 4.6 mm), fitted with a Supelguard cartridge (Discovery), C18 (5 µm, 2 cm × 4 mm). The injection volume was fixed at 20 µL and the flow-rate was 1 mL/min. Mobile phase adopted for this study was water/acetonitrile/acetic acid (80/19/1 v/v/v) (Al-Rimawi, 2014). The extract obtained from MAE was filtered through a 0.45 mm syringe filter and directly injected to the HPLC. Chromatographic peaks of the samples were identified by comparing their retention time and UV spectrum with those of the reference standard. The detector was set at 280 nm and the sample run was conducted for 20 min. The concentrations of OLE and HYD in the extract were quantified using standard curves with concentrations of pure standards ranging from 20 to 1000 mg L^{-1}. The extraction yields of OLE and HYD were expressed in mg g^{-1} olive leaves (d.b.).

Biodegradable Film Formation

Carrageenan films were produced by the casting method, using the methodology proposed by Martiny [22]. The amount of biopolymer and plasticizer used were established through preliminary experiments and literature. The film forming solutions were obtained by dissolution of 0.5 g biopolymer powder in 30 mL of distilled water using a hot plate and magnetic stirrer (Cimarec2, Thermolyne, China) at 70 °C for 15 min. During continuous stirring, glycerol was added to the solution at a concentration of 60% w/w with respect to polymer. After cooling the solution, the lyophilized extract of olive leaf was added at three different concentrations of 50, 100 and 200 mg to the formulation. The lyophilized extract was obtained using a freeze dryer (Gamma 1–16 LSC, Christ, Osterode am Harz, Germany) for 48 h. Finally, the film forming solution was poured into polystyrene petri dishes (90 mm diameter) and biodegradable films were obtained by solvent evaporation using convective and vacuum dryers. After drying, film samples were peeled from Petri dishes and conditioned at room temperature for 48 h in a desiccator containing sulfuric acid solution with a relative humidity of 50% before testing. A biodegradable film of carrageenan without the addition of olive leaf extract was produced as control.

Biodegradable Film Characterization

Thickness

The biodegradable film thickness was measured using a digital micrometer (Marathon Co, CO030025, Canada) with 0.001 mm resolution. The mean thickness was calculated from ten measurements taken at different locations of biodegradable film samples, according to Ferreira [23].

Water vapor permeability

Water vapor permeability (WVP) of biodegradable films was determined gravimetrically using the ASTM [24] standard method. Samples of each biodegradable film in the form of discs were fixed in a cell permeation unit (diameter 30 mm) containing anhydrous calcium chloride. These cells were placed in desiccators with 50% relative humidity at room temperature. By increasing the mass of anhydrous calcium chloride in 7 days it was possible to determine the water vapor transferred through the biodegradable film according to the Eq. 2.

$$WVP = \frac{w_a}{t} \times \frac{l}{A \times \Delta P} \tag{2}$$

where, w_a is the amount of absorbed water (g), t is the time (s), l is the average biodegradable film thickness (m), A is the area of the exposed biodegradable film surface (m^2) and ΔP is the partial vapor pressure difference across the biodegradable film (Pa).

Mechanical Properties

Tensile strength (TS) and elongation percentage (E) at break point were measured uniaxially by stretching the specimen in one direction using a Universal Testing Machine (Instron 4502, USA) according to the ASTM [25] standard method. Samples were clamped and deformed under tensile loading using a 50 N load cell with initial grip separation of 25 mm and cross-head speed of 50 mm min^{-1}.

Color Measurement

Color measurement of films was performed using a Chroma-Meter spectrophotometer (CR-300, Konica Minolta, Japan). L^* (lightness), a^* (redness, greenness), and b^* (yellowness, blueness) color values were determined. The chromameter was calibrated before each series of measurements using a white ceramic plate. The mean of five measurements was recorded for each film from different points. The total color difference (ΔE) was determined as an estimate of color changes, according Eq. 3.

$$\Delta E^* = \sqrt{\left(L^* - L_s^*\right)^2 + \left(a^* - a_s^*\right)^2 + \left(b^* - b_s^*\right)^2} \quad (3)$$

where L^*, a^* and b^* are the color parameter values of the control and L_s^*, a_s^* and b_s^* are the color parameter values of the sample.

Total Phenolics (TP_{biof}) and Antioxidant Activity (AA_{biof}) of Biodegradable Films

The TP_{biof} and AA_{biof} of biodegradable films were determined by the same methods which were used for olive leaf extract. A 100 mg of each film sample was dissolved in 15 mL of distilled water by continuous stirring, then the solution was used to determine the compounds.

Morphology

The scanning electron micrographs (SEM) of the biodegradable films were obtained using a SEM (Hitachi, TM 3000, Japan) to analyze the microstructure of the film surface and the crosss-sections. The film samples were attached to a double-sided adhesive tape of carbon and mounted on the specimen holder. The images of the samples were captured with an accelerating beam voltage of 5 kV.

Fourier-Transform Infrared Spectroscopy

The Attenuated total reflectance Fourier transform infrared spectroscopy (FTIR-ATR) was used for the chemical characterization of biofilms and to observe the structural interaction of carrageenan-based films with added extract. A Perkin-Elmer spectrometer (UATR Two), in the range of 400 to 4000 cm^{-1}, was used with 32 scans per spectrum and with a resolution of 4 cm^{-1}. The samples were cut into small squares and then inserted into the sample portal of the FTIR-ATR apparatus.

Statistical Analysis

The experimental data were analyzed by Statistic software version 7.1. The mean comparisons were carried out by Tukey test to determine possible significant differences among the treatments at a 95% confidence level.

Results and Discussion

The results of total phenolic compounds (TP), antioxidant activity (AA), oleuropein content (OLE) and hydroxytyrosol content (HYD) from olive leaf extract obtained with MAE are shown in Table 1.

The results of antioxidant activity (AA) and total phenolic compounds (TP) indicated that olive leaves have a high potential for AA, corroborating with the literature [18, 26–28]. The AA revealed that the extract might prevent reactive radical species from reaching biomolecules such as lipoproteins, polyunsaturated fatty acids, amino acids, proteins and sugars in biological and food systems [29].

The MAE was efficient in extracting the bioactive compounds and this efficiency can be attributed to the cell wall disruption, which promotes faster release of the compounds into the solvent [30]. Rafiee et al. [31] reported the TP yield of 54.08 $mg.g^{-1}$ (d.b.) for olive leaf extract was obtained using water as solvent in MAE with similar extraction time (4 min).

The OLE content reported here was 14.468 $mg.g^{-1}$ (d.b.). Japon-Lujan et al. [32] achieved an OLE concentration of 20 $mg.g^{-1}$ (d.b.) in extracts obtained from MAE using 80:20 ethanol–water mixtures. Taamali et al. [33] carried out the MAE for 6 min and found the best recovery for oleuropein was microwave-assisted method compared to supercritical fluid extraction and pressurized liquid extraction techniques. However, the main disadvantage was the use of toxic organic solvent (methanol) in this study. Sahin et al. [27] reported a yield of oleuropein at 0.00006 $mg.g^{-1}$ (d.b.) and TP of

Table 1 Total phenolic compounds (TP), antioxidant activity (AA), oleuropein (OLE) and hydroxytyrosol (HYD) contents

TP (mg_{GAE} g^{-1} d.b.)	AA (%)	OLE (mg_{OLE} g^{-1} d.b.)	HYD ($mg_{HYD} g^{-1}$ d.b.)
104.22 ± 0.61	90.03 ± 0.04	14.468 ± 0.405	0.590 ± 0.001

0.00248 $mg_{GAE}.g^{-1}$ (d.b.) from MAE using water, these results were much lower than those reported in our study.

Moudache et al. [18] studied the use of different solvents to extract bioactive compounds from olive leaves to incorporate in to polyethylene films. They reported that the antioxidant activity of olive leaf extracts obtained using maceration with distilled water was 92.4%. However, OLE was not extracted with distilled water and then, they chose to perform the extraction study with 70% of aqueous ethanol.

Abaza et al. [34] mentioned that the radical scavenging of olive leaf extracts depends on the solvent polarity: % methanol 80 > % ethanol 70 > % acetone 80 > % H_2O. Nevertheless, due to toxicity, finding replacement of toxic solvents is highly desirable from the point of view of food application. One of the main needs in the development of extraction processes is to substitute inefficient and long extraction processes, usually requiring high volumes of toxic organic solvents, for nonconventional extraction procedures such as MAE, that require considerably less amounts of toxic solvents while providing higher extraction efficiencies and have a lower environmental impact. In this sense, the results achieved in this work with MAE using water as solvent showed as an excellent alternative for extraction method. They also lead to increase in the process efficiency thus reassuring the green technologies concept.

Table 2 shows the values obtained for properties of the carrageenan biodegradable films with added olive leaf extracts.

The carrageenan films produced with olive leaf extract is very promising as per the data shown in Table 2. Normally, a high tensile strength in the formulated film is common, however the flexibility of the film, indicated by the elongation is a very important parameter. This enables better applicability of this film in packaging [35].

Martins et al. [36] produced films with thickness of 0.052 mm, 16.18% elongation and 19.95 MPa for tensile strength. Several studies with carrageenan films have shown WVP in the range of 5.8.10 $^{-11}$ to 2.18.10^{-8} g. m^{-1}.s^{-1}.Pa^{-1}, tensile strength from 11.64 to 26.29 MPa, and elongation from 2.54 to 45% [8, 37–41]. The data reported in the literature for tensile strength and elongation of carrageenan films are smaller than those obtained in this work for the control sample. This was attributed to the differences in the preparation, composition and proportions of the film-forming solutions.

The film thicknesses were in the range of 0.097 to 0.162 mm and showed significant difference ($p > 0.05$) regarding films obtained with different concentrations of olive leaf extract, which could be due to an increase in solids because of the incorporation of the extract. The addition of extract to the biodegradable film resulted in a higher WVP, which not only depends on both molecular diffusion coefficient and water solubility of the film material [42], but mainly on the hydrophilicity of the film [43]. In general, polar compounds enhance the hydrophilic properties of the films, leading to an increase in WVP [44]. The literature reports that the polarities of phenolic compounds vary significantly; however, the MAE extraction has been shown to be more efficient for extracting polar compounds, such as oleuropein [45].

There was no significant difference ($p < 0.05$) between the elongation results, although films with extract showed slight increase in the stretching capacity. According to Marcos et al. [17], the addition of active substances to packaging films can modify the mechanical properties of the films. The inclusion of 100 and 200 mg of extract in carrageenan films induced a significant reduction in its tensile strength. Pastor et al. [46] reported similar results for chitosan and methylcellulose films added with resveratrol, which resulted in an increase in film thickness, elongation and redution in tensile strength. Moreover, the incorporation of phenolic compounds from olive by-products on the pectin-gelatin

Table 2 Properties of biodegradables films with addition of olive leaf extracts

	Control	Carr-50 mg	Carr-100 mg	Carr-200 mg
Thickness (mm)	0.097 ± 0.005^a	0.110 ± 0.004^b	0.128 ± 0.009^c	0.162 ± 0.003^d
WVP (g m^{-1} s^{-1} Pa^{-1})	$1.87.10^{-10} \pm 3.03.10^{-12a}$	$2.32.10^{-10} \pm 1.67.10^{-11ab}$	$2.50.10^{-10} \pm 3.27.10^{-12b}$	$3.19.10^{-10} \pm 1.10.10^{-12c}$
TS (MPa)	37.12 ± 0.16^a	35.46 ± 2.34^a	27.92 ± 2.94^b	20.86 ± 1.05^c
E (%)	78.88 ± 2.40^a	96.05 ± 6.76^a	81.71 ± 8.29^a	85.49 ± 2.79^a
L	92.63 ± 0.23^a	74.74 ± 0.7^b	70.42 ± 0.92^c	68.13 ± 0.65^d
a	-0.49 ± 0.09^a	4.76 ± 0.18^b	8.91 ± 1.04^c	9.27 ± 0.30^c
b	11.42 ± 0.87^a	40.38 ± 0.45^b	44.62 ± 1.99^c	47.53 ± 0.34^d
ΔE	–	34.32	40.91	44.59
TP_{biof} (mg_{GAE} g^{-1} d.b.)	3.02 ± 0.76^a	18.55 ± 0.06^b	44.08 ± 1.03^c	68.07 ± 1.75^d
AA_{biof} (%)	0^a	1.4 ± 0.7^a	9.27 ± 1.3^b	32.55 ± 0.5^c

Average ± std. deviation (n = 10 for thickness, n = 2 for WVP, n = 3 for mechanical properties, n = 5 for color, n = 2 for TP and AA)

Different letters in the same line indicate significant differences between samples (p < 0.05)

films caused a significant increase in the elongation at break point [47].

The effect of addition of extracts at different concentrations on color of biodegradable films are shown in Table 2. From this data, it was clear that the addition of extract resulted in higher redness and yellowness (increase of a^* and b^*) of the films, increasing the color difference between films with and without extract (control). There were significant differences in L^* and b^* values reported at different levels of extract concentrations. L^* has reduced and b^* increased with increasing concentrations. Furthermore, with regards to a* parameter the biodegradable films obtained with 100 and 200 mg of extract did not show any significant difference between them.

As expected, the total phenolic compounds and antioxidant activity of carrageenan films significantly increased with an increase in olive leaf extract concentration. The film containing 200 mg of extract had the highest TP_{biof} and $AA_{biof.}$. The phenolic compounds are responsible for the antioxidant power in quenching free radicals and in most cases, the antioxidant capacities of the films are proportional to the concentration of the active compounds in the film [48]. The same trend was observed in the antioxidant capacity and total phenolics reported by Albertos et al. [6] for gelatin films with olive leaf additives, where the maximum TP in biodegradable films was 26.69 mg_{GAE} g^{-1} d.b.

The appearance and SEM images of surface and cross-sections of carrageenan films incorporated with different concentrations of olive leaf extract as compared with control film are shown in Figs. 1 and 2.

The visual examination of the films showed that films showed a high opacity. The micrographs of surface and

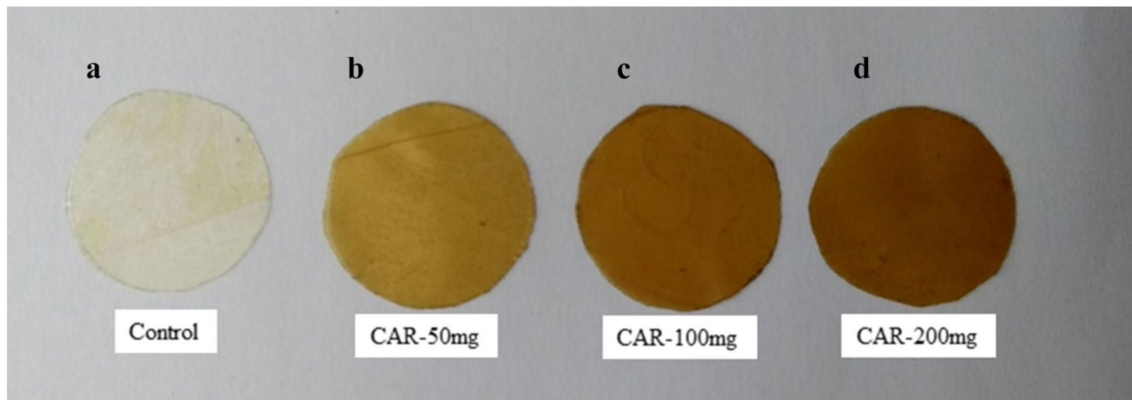

Fig. 1 Film appearance: **a** control, **b** Carr-50 mg, **c** Carr-100 mg, **d** Carr-200 mg

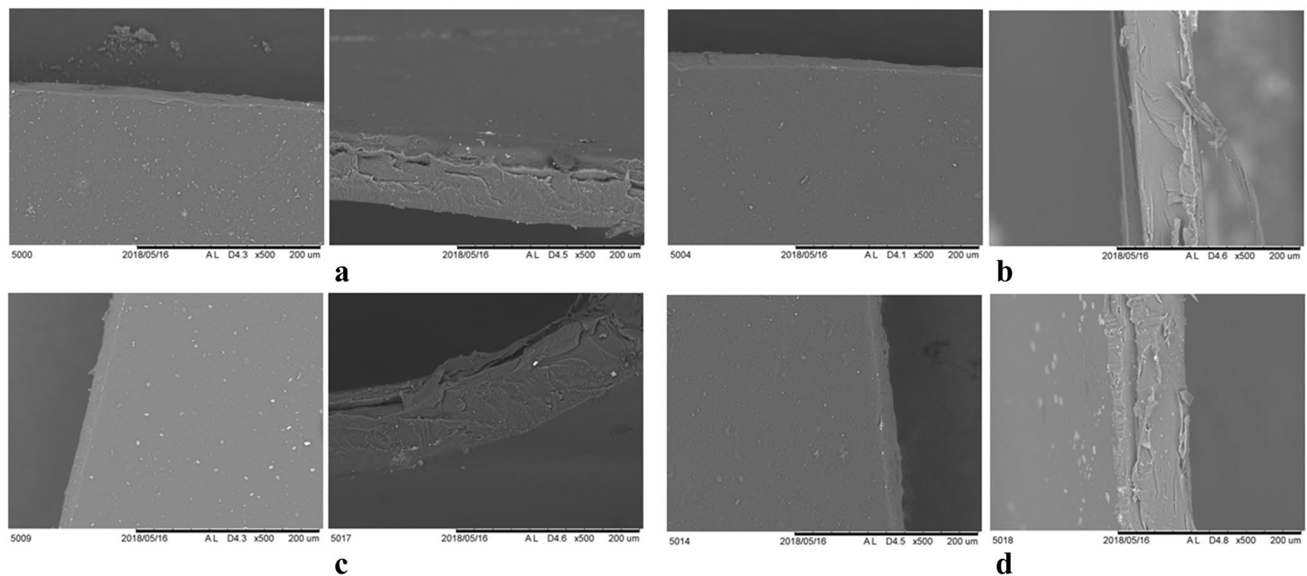

Fig. 2 SEM micrographs of surface and cross-sections of films: **a** control, **b** Carr-50 mg, **c** Carr-100 mg, **d** Carr-200 mg

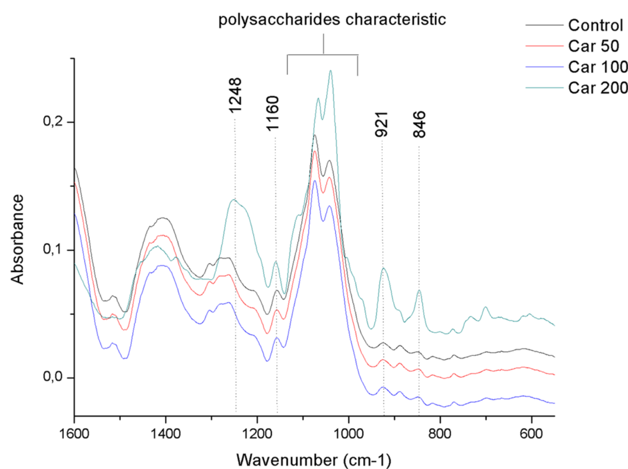

Fig. 3 FTIR-ATR spectra of carrageenan biodegradable films control and carrageenan biodegradable films with olive leaf extract

cross-sections of films exhibited a well-organized homogeneous structure, continuous, compact, nonporous morphology, and a smooth surface. The images also revealed that the incorporation of extract had a good dispersion of the bioactive compounds with no visual defects and cracks.

Figure 3 shows the FTIR-ATR spectra of carrageenan biodegradable films. The analyzed spectrum had strong absorption bands in the region of 1000–1100 cm^{-1}, attributed to the glycosidic bonds present in carrageenans. The spectrum shows bands of strong absorption in the region 921 cm^{-1} (C-O of 3,6-anhydrogalactose) and in the region 846 cm^{-1} (C–O–SO_4 ofgalactose-4-sulfate), typical of k-carrageenan [49, 50]. Intense bands between 1248 and 1160 cm^{-1} correspond mainly to C–O stretching vibration endocyclic and exocyclic carbohydrate [51]. With the analysis of the generated spectra there was a smooth displacement of the absorption bands of carrageenan films incorporated with different concentrations of olive leaf extract as compared with control film. More research is going on this approach to discover about the miscibility between the carrageenan biopolymer matrix and the olive leaf extract, which possibly lead to changes in the film properties such as barrier and mechanics, as showed previously.

Conclusion

This study demonstrates that olive leaf extract obtained by MAE solvent-free had high antioxidant activity and has great potential for use as functional ingredient in food packaging. The use of plant extracts, like olive leaf extract, as antioxidants is still a novel subject for designing antioxidant food packaging. Furthermore, it is also a sustainable way to provide an added value to byproducts from olive cultivation.

The biodegradable films based on carrageenan containing olive leaf extract developed in this study exhibited good barrier properties and mechanical properties. The film containing bioactive compounds showed higher thicknesses due to an increase in the solids incorporated from the extract. The addition of extract also resulted in a slight increase in the stretching capacity and a higher WVP, due to the presence of polar compounds like oleuropein. The total phenolic compounds and antioxidant activity of films significantly increased with an increase in olive leaf extract concentration, which means that the process to obtain the films does not provoke a degradation of these bio compounds.

Therefore, the results showed that a carrageenan films containing olive leaf extract can be a packaging material suitable for food products. In future, new research regarding the effect of extended contact between the biodegradable films and the food should be considered for the evaluation of their technological application.

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Compliance with Ethical Standards

Conflict of interest The authors have no conflicts of interest to report.

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