Development and Characterization of Edible Packaging Film, with Anise or Cinnamon Essential Oil addition, Plasticized with Glycerol or Sorbitol or Polyethylene Glycol, Using Lignin Extracted from Coffee Silverskin

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ABSTRACT

In this study, edible packaging films were developed by solvent casting method using lignin extracted from Costa Rican or Kenyan coffee silverskin and their barrier properties were investigated. Anise or cinnamon essential oil, glycerol or sorbitol or polyethylene glycol, and sodium alginate effects on the film properties were compared. The moisture content, swelling index and water vapor transmission rate analyses of the produced edible films were realized. Chemical structure analysis was done using Fourier Transform Infrared Spectroscopy device. Edible films plasticized with glycerol, have higher moisture content (23.21%), low swelling index (28.44%) and low water vapor transmission rate ($0.7991*10^{-8}$($g/cm^2.s$)), than the films plasticized with sorbitol and polyethylene glycol. As a result, it was observed that coffee processing waste coffee silverskin, plasticized in preference with glycerol and mixed with anise or cinnamon essential oil, can be successfully used as a cost-effective lignin source for edible packaging film production.

KEYWORDS: Coffee silverskin, lignin, edible film, packaging

1. INTRODUCTION

Edible films are used to preserve fruits, vegetables, meat, fish etc. They prevent gas and water vapor transfer, protecting the food from the air, lowering corruption and prolong the shelf life. A great part of the polymers used in food packaging are nonbiodegradable, petroleum-based materials. Nowadays lots of studies are carried out to produce new biodegradable, environment-friendly packaging films to reduce the use of environmentally hazardous plastics and replace them with low-cost biopolymers [1-3]. Coffee is a widely consumed drink in the world. World coffee production was reported 168.5 million bags and world coffee consumption was reported 175.6 million bags in year 2021/22 by International Coffee Organization [4]. The amount of the coffee processing wastes generated, such as coffee pulp, spent coffee grounds and coffee silverskin, has increased with this rise of coffee consumption in the world. Coffee silverskin is the major by-product of coffee roasting companies, and it contains averagely 24% cellulose, 17% hemicellulose, and 29% lignin [5-7]. Coffee by-products, rich in polysaccharides and lipids, having antioxidant and antimicrobial properties, can be used for the edible film production and could provide environmental and economic advantages [8-9]. Essential oils, being antioxidant, antimicrobial, antiviral and antifungal agents, are used to get minimum processed food products with extended shelf life. Anise, cinnamon, lemon, tea tree or bergamot essential oils were incorporated to films to develop barrier properties. Cinnamon essential oil (CEO) was incorporated to the films to improve their antibacterial, antioxidant and physical properties. Cinnamaldehyde, can form hydrogen bonds with the hydroxyl groups of lignin obtained from coffee silverskin, improving the network structure, hydrophobicity, and barrier properties of the edible films [10-12]. Anise has multiple properties such as antimicrobial, antioxidant and insecticidal activities. Essential oil extracted from this fruit is being widely used in food and medicine areas due to its strong bioactivity. Phenolic and/or hydrophobic components of anise essential oil (AEO) provide antimicrobial properties [13-14]. Sodium alginate, natural hydrophilic polysaccharide, is used in the biopolymer film production due to its good film-forming properties. Edible films produced using only sodium alginate have high hydrophilicity and poor heat stability. Various polymers and plasticizers can be combined with sodium alginate to overcome this problem [15-16]. Glycerol, sorbitol, propylene glycol, polyethylene glycol (PEG), and glycerin can be added to the edible film forming solution to improve plasticity and flexibility. Plasticizers decrease intermolecular resistance and moisture absorption, enhance chain mobility. Plasticizer type and concentration, affect the properties of the produced edible film [17-19]. It’s aimed in this study is to produce a new edible packaging film using lignin extracted from Costa Rican coffee silverskin (CRCS) and Kenyan coffee silverskin (KCS). CEO or AEO was incorporated to the film forming solution due to their hydrophobic nature. Glycerol or sorbitol or PEG was used as a plasticizer to determine the most suitable plasticizer for edible film production and sodium alginate was used to compose a polymeric matrix. Film forming solutions were produced by the solution casting method. The outcomes of this work will enable the coffee roasting companies major waste product, coffee silverskin to be recycled to generate biodegradable, environment-friendly and low-cost edible packaging film, contributing to the environmental protection.

2. MATERIALS AND METHODS

2.1. Materials

CRCS and KCS used in this study were obtained free of charge from Kahve Dünyası, CEO and AEO were bought from Arifoğlu (Istanbul). PEG and D-sorbitol were bought from Sigma-Aldrich (USA), glycerol and hexane were bought from Merck (Germany), ethanol was bought from Chem-Lab (Belgium), sodium alginate was bought from AFG Bioscience (USA).

2.2. Coffee lignin extraction from coffee silverskin

Fig. 1. Schematic diagram of the coffee lignin extraction from coffee silverskin

The CRCS and KCS were first grinded during 20 min. 10 g ground CRCS or KCS was dissolved in 200 ml distilled water, then the solution was mixed and heated at 95^0C for 30 min. After cooling, the solid fraction was separated by vacuum filtration. The CRCS or KCS residue and 100 ml of ethanol were mixed at 500 rpm for 60 for 2 hours at 70℃ and filtered again. Finally, the solid fraction was washed and dried in an oven at 60°C for 24 hours to obtain coffee lignin [20-21]. Schematic diagram of the coffee lignin extraction from CRCS or KCS was given in Fig.1.

2.3. Coffee lignin based edible film production.

Table 1. Edible packaging films compositions

Film	CRCSL (g)	KCSL (g)	AEO (g)	CEO (g)	Glycerol (g)	Sorbitol (g)	PEG (g)	Sodium Alginate (g)	Water (ml)
CRA$_1$	1	-	2	-	0.5	-	-	0.5	50
CRA$_2$	1	-	2	-	1	-	-	0.5	50
CRA$_3$	1	-	2	-	-	0.5	-	0.5	50
CRA$_4$	1	-	2	-	-	1	-	0.5	50
CRA$_5$	1	-	2	-	-	-	0.5	0.5	50
CRA$_6$	1	-	2	-	-	-	1	0.5	50
CRC$_1$	1	-	-	2	0.5	-	-	0.5	50
CRC$_2$	1	-	-	2	1	-	-	0.5	50
CRC$_3$	1	-	-	2	-	0.5	-	0.5	50
CRC$_4$	1	-	-	2	-	1	-	0.5	50
CRC$_5$	1	-	-	2	-	-	0.5	0.5	50
CRC$_6$	1	-	-	2	-	-	1	0.5	50
KA$_1$	-	1	2	-	0.5	-	-	0.5	50
KA$_2$	-	1	2	-	1	-	-	0.5	50
KA$_3$	-	1	2	-	-	0.5	-	0.5	50
KA$_4$	-	1	2	-	-	1	-	0.5	50
KA$_5$	-	1	2	-	-	-	0.5	0.5	50
KA$_6$	-	1	2	-	-	-	1	0.5	50
KC$_1$	-	1	-	2	0.5	-	-	0.5	50
KC$_2$	-	1	-	2	1	-	-	0.5	50
KC$_3$	-	1	-	2	-	0.5	-	0.5	50
KC$_4$	-	1	-	2	-	1	-	0.5	50
KC$_5$	-	1	-	2	-	-	0.5	0.5	50
KC$_6$	-	1	-	2	-	-	1	0.5	50

Edible film forming solutions based on Costa Rican coffee silverskin lignin (CRCSL) and Kenyan coffee silverskin lignin (KCSL) were obtained by adding CEO or AEO to increase hydrophobicity, glycerol or sorbitol or PEG as a plasticizer, and sodium alginate to form a polymeric matrix. Film forming solution were made by the solution casting method with respect to the combination given in Table 1. The film forming solution were mixed in a magnetic stirrer at 500 rpm for 30 minutes at 80°C. 2 g of hexane was added to the mixtures and the use of hexane and distilled water as solvent presented good dissolving. The solutions were homogenized at 90°C, for 60 minutes at 500 rpm using magnetic stirrer. To obtain completely homogeneous solutions, they were homogenized using ultrasonic homogenizer for 15 minutes and kept in an ultrasonic water bath at 40°C for 10 minutes. The film forming solution were poured into a petri dish and dried at 60°C for 24 hours. The films were removed and stored, at 50% RH and room temperature (22+0.5 °C) [3, 16, 22-23]. Schematic diagram of the coffee lignin based edible film production was given in Fig.2. Produced CRCSL and KCSL based edible films images were given respectively in Fig.3.

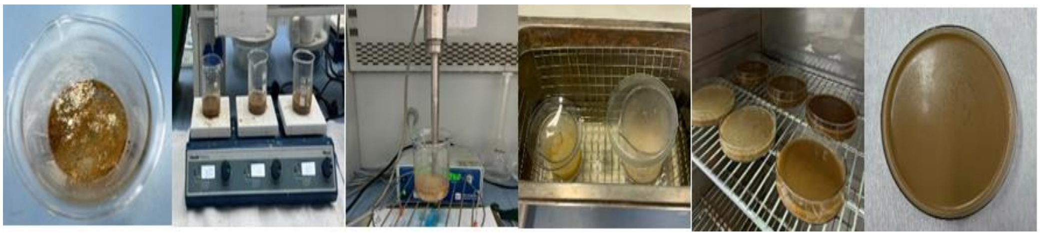

Fig. 2. Schematic diagram of the coffee lignin based edible film production.

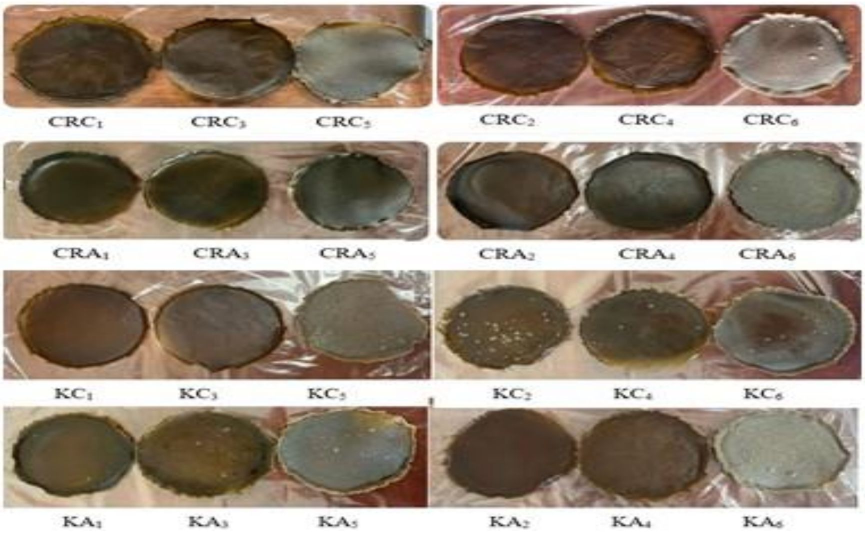

Fig. 3. Costa Rican and Kenyan Coffee silverskin lignin based edible films.

2.4. Characterization of the films

2.4.1. Moisture content

The moisture content was determined using the gravimetric method. The films were cut 3x3 cm and weighed. They were dried at 104 ± 2°C for 24 hours until the equilibrium weight was reached. The weight loss was defined, the moisture content was calculated as the percentage of water removed from the system using Eq. (1):

$$\text{Moisture (\%)} = \frac{(\mathbf{M_0 - M_1})}{\mathbf{M_0}} \times \mathbf{100} \qquad (1)$$

M_0 = initial weight, M_1 = dry weight of the films

2.4.2. Swelling index

The swelling index of the films was determined using the method described by Bhatia et al.[24]. The films were cut 3x3 cm, dried at 104 ± 2 °C for 24 h and weighed *(W_0)*. They were immersed in15 mL distilled water for 2 minutes at room temperature (22+0.5 °C). The swelled samples were wiped with filter paper and weighed *(W_1)*. The swelling index of the films was calculated using Eq. (2):

$$\text{Swelling index (\%)} = \frac{W_1 - W_0}{W_0} \times 100 \qquad (2)$$

2.4.3. Water vapor transmission rate (WVTR)

The *WVTR* of the films was calculated, with respect to the method described by Shafie et al.[25] and ASTM Standard Method E96-80 [26]. The films were dried in a hot air oven at 104 ± 2 ˚C for 24 h. The mouth of the test tubes, filled with 5 g of silica gel, was closed with the film sample. The prepared samples were weighed every 24 h for 5 days. The WVTR was calculated according to the formula below:

$$\text{WVTR} = \frac{\Delta W}{\Delta t} \div A \quad (3)$$

$\frac{\Delta W}{\Delta t}$ = the amount of water transferred (g) per unit time (s), and A = the exposed area (m^2).

2.4.4. Fourier transform infrared spectroscopy (FT-IR)

The films (10 × 10 mm) structures were determined by FT-IR with a spectral range between 4000 and 400 cm^{-1} with a resolution of 4 cm^{-1}.

2.4.5 Statistical analysis

Statistical analysis by the analysis of variance (ANOVA) was done by SPSS®16. All tests were carried out in three independent runs, and the obtained parameters were averaged and expressed as the mean standard error (±) where each value is considered as a significant at $p < 0.05$.

3. Results and discussion

3.1. Characterization of coffee lignin based edible films

3.1.1. Moisture content

Moisture content value represent total void volume occupied by water molecules in the microstructure network of the edible film and displays the possible effect of hydrocolloids interaction on the affinity of films to water [27-28]. Moisture content, which is related to the water resistance abilities of edible film, is a major feature for the shelf life of coated materials. It was understood from Table 2 that, the moisture content of edible films increased with increasing plasticizers (glycerol/sorbitol/PEG) amount which help retain water in polymeric matrix. Moisture contents of the edible films were affected also by the type of the plasticizer. Edible film plasticized with glycerol (1 gr), have higher moisture content than the films plasticized with sorbitol and PEG. Because glycerol has the smallest molecular weight and strongest hydrophilicity among them [7, 29]. The moisture content of edible films produced with AEO added CRCSL ranged from 5.47% to 17.72%, the moisture content of edible films produced with CEO added CRCSL ranged from 6.34% to 23.21%, the moisture content of edible films produced with AEO added KCSL ranged from 5.80% to 20.85%, the moisture content of edible films produced with CEO added KCSL ranged from 6.01% to 20.55%. Similar results were obtained by Paudel et al. [18] and Lim et al. [30].

3.1.2. Swelling index

The swelling index express the film matrix capability to absorb and keep water and indicates their biodegradation and maintenance of quality and storage of food products. The swelling capacity characterizes water resistance of edible films and affect their physical and barrier properties. This property is dependent to the hydrophilic groups present in the edible films. When the glycerol or sorbitol or PEG content in the edible film increase, the swelling index decrease, which might be due to the spaces occupied by plasticizer molecules in film network for absorbing water molecules [31-35]. The values of swelling index of edible films are given in Table 2. The swelling index of edible films produced with AEO added CRCSL ranged from

41.27% to 72.38%, the swelling index of edible films produced with CEO added CRCSL ranged from 45.64% to 92.87%, the swelling indexes of edible films produced with AEO added KCSL ranged from 28.44% to 78.16%, the swelling indexes of edible films produced with CEO added KCSL ranged from 44.62% and 116.28%. Edible films produced with AEO added KCSL and plasticized with glycerol (1 gr) have lowest swelling index.

Table 2 Physical properties of the coffee lignin based edible films.

Films	**Moisture Content (%)**	**Swelling Index (%)**	**WVTR (gram/cm^2.s)**
CRA$_1$	10.30 ± 1.35^a	58.48 ± 1.23^a	$2.6986\ x10^{-8} \pm 0.77^b$
CRA$_2$	17.72 ± 2.13^b	47.39 ± 1.78^b	$4.2706\ x10^{-8} \pm 1.68^a$
CRA$_3$	5.47 ± 0.14^b	$72,38 \pm 2.59^b$	$3.8907\ x10^{-8} \pm 1.37^b$
CRA$_4$	5.92 ± 0.11^a	63.71 ± 2.76^a	$1.2707\ x10^{-8} \pm 0.53^a$
CRA$_5$	6.06 ± 0.23^b	51.04 ± 0.98^a	$4.2707\ x10^{-8} \pm 0.99^b$
CRA$_6$	6.13 ± 0.35^a	41.27 ± 0.77^b	$2.0305\ x10^{-8} \pm 0.26^a$
CRC$_1$	11.83 ± 2.53^b	70.09 ± 2.96^a	$7.1527\ x10^{-8} \pm 1.04^a$
CRC$_2$	23.21 ± 1.42^a	53.19 ± 1.16^b	$6.1964\ x10^{-8} \pm 1.35^a$
CRC$_3$	6.34 ± 0.36^a	56.60 ± 1.35^a	$5.3318\ x10^{-8} \pm 1.88^b$
CRC$_4$	6.50 ± 0.45^b	45.64 ± 2.08^b	$6,3667\ x10^{-8} \pm 0.79^a$
CRC$_5$	7.80 ± 1.21^b	92.87 ± 1.54^b	$5.0560\ x10^{-8} \pm 1.34^a$
CRC$_6$	8.07 ± 0.84^a	69.13 ± 1.96^a	$1.6760\ x10^{-8} \pm 0.27^b$
KA$_1$	$10,77 \pm 1.08^a$	78.16 ± 2.24^b	$1.0480\ x10^{-8} \pm 0.12^b$
KA$_2$	20.85 ± 1.23^b	28.44 ± 0.82^a	$2.8165\ x10^{-8} \pm 0.51^a$
KA$_3$	5.80 ± 0.18^a	59.41 ± 1.41^b	$4.7684\ x10^{-8} \pm 1.54^a$
KA$_4$	5.86 ± 0.04^b	62.88 ± 2.53^a	$1.8209\ x10^{-8} \pm 0.11^b$
KA$_5$	6.64 ± 2.31^a	49.95 ± 0.93^a	$4.4017\ x10^{-8} \pm 2.19^a$
KA$_6$	7.42 ± 1.67^b	41.87 ± 0.87^b	$2.1746\ x10^{-8} \pm 0.07^b$
KC$_1$	13.27 ± 1.36^a	56.99 ± 1.81^a	$8.9344\ x10^{-8} \pm 2.11^b$
KC$_2$	20.55 ± 1.53^b	49.96 ± 1.67^b	$0.7991\ x10^{-8} \pm 0.08^a$
KC$_3$	6.01 ± 0.18^a	116.28 ± 2.75^b	$3.3667\ x10^{-8} \pm 0.27^a$
KC$_4$	6.13 ± 0.35^a	44.62 ± 1.28^a	$4.5851\ x10^{-8} \pm 2.44^b$
KC$_5$	7.86 ± 0.28^b	98.48 ± 2.49^b	$1.6506\ x10^{-8} \pm 0.16^b$
KC$_6$	8.57 ± 2.17^a	86.19 ± 3.07^a	$4.7554\ x10^{-8} \pm 1.45^a$

3.1.3. Water vapor transmission rate (WVTR)

WVTR is the measure of the steady state rate at which water vapor permeates through a film under specific conditions of temperature and relative humidity. Low

WVTR values avoid the moisture absorption of the film and avoid dehydration when they are kept in a low moisture condition. Hence, this characteristic extends the shelf-life of foods. The WVTR of the films generally depends on the polymeric matrix and can be changed using plasticizers. Using glycerol/sorbitol/PEG as a plasticizer prevent high WVTR values due to their hydrophilic nature [25, 36-38]. Edible films produced with CEO added KCSL, plasticized with glycerol (1 gr) has the lowest WVTR ($0.7991X10^{-8}$ $g/cm^2.s$) values because glycerol is more hydrophilic compared to sorbitol and PEG.

3.1.4. Fourier transform infrared spectroscopy (FT-IR)

FTIR spectra of some selected CRCSL and KCSL based edible films produced are shown in Figure 4. Glycerol and sorbitol plasticized edible films showed generally similar FTIR spectra to that of pure coffee silverskin lignin. The broad band at about 3280 cm^{-1} is related to the hydroxyl group of O-H stretching vibration. When PEG is used as a plasticizer, the O-H band shifted from 3280cm^{-1} to 3347 cm^{-1}, showing that hydrogen bonds between coffee silverskin lignin molecules became poor. The two peaks at around 2924 cm^{-1} and 2856 cm^{-1} were related to the C-H stretching vibration of coffee silverskin, CEO and AEO. The bands at around 1744 cm^{-1} were related to the vibration of the acetyl groups and ester linkages in hemicellulose and lignin. The bands at around 1604 cm^{-1} were related to the aromatic ring of lignin. The peaks observed at around 1400-1000 cm^{-1} region, marked the presence of cellulose, lignin, and hemicellulose, as revealed by the strong absorption bands including the aromatic ring and carbonyl vibration regions. The absorption bands appearing at 1320 cm^{-1}, 1236 cm^{-1} and 1097 cm^{-1} correspond to the stretching vibration peaks of C–O–C in AEO [29, 39-42]. FTIR spectra of edible films CEO or AEO added and plasticized with glycerol or sorbitol showed generally similar peaks to that of pure coffee silverskin lignin, which confirmed that there was no significant change in the chemical structure.

4. Conclusion

In this study, AEO or CEO added, and plasticized with glycerol or sorbitol or PEG edible packaging film was produced using lignin extracted from CRCS and KCS with casting method. Increasing plasticizers (glycerol/sorbitol/PEG) amount increased the moisture contents of the edible films, and glycerol is the most suitable plasticizer. KCSL based, AEO incorporated, and glycerol plasticized edible films have the lowest swelling index. KCSL based, CEO incorporated, and glycerol plasticized edible films have the lowest WVTR. When the FTIR spectrum was examined, it was seen that no new peaks were formed, indicating that there was no chemical interaction between the edible film components and that CRCS and KCS, essential oils, and plasticizers were successfully combined. This result proves that coffee silverskin, being biodegradable and cost-effective lignin source, can be used in edible packaging production. In conclusion, great amount of coffee roasting by product, coffee silverskin, will be transformed into a commercial product, at the same time environmental pollution will be prevented.

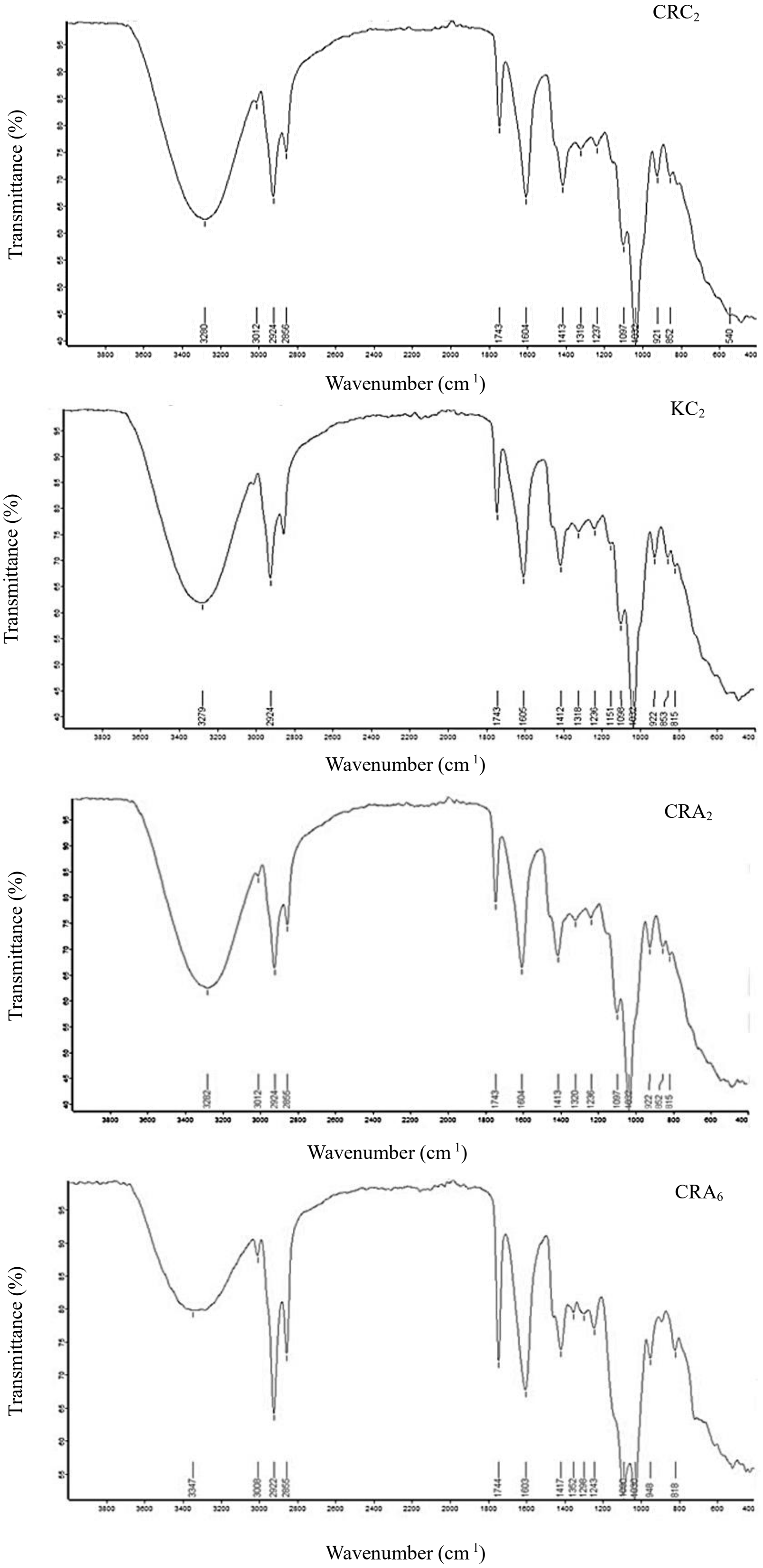

Fig. 4. FTIR spectra of CRC$_2$, KC$_2$, CRA$_2$ and CRA$_6$ edible packaging films

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