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Treatment of Kraft paper with citrus wastes for food packaging applications: Water and oxygen barrier properties improvement

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ABSTRACT

Hydrophobic materials extracted from citrus wastes, both peel of mandarin fruits and leaf of mandarin trees were used to treat food-grade Kraft paper. The chemical compounds of the extracts were identified by gas chromatography–mass spectroscopy and infrared spectroscopy, and their antioxidant activities were determined using a free radical scavenger agent (2,2-diphenyl-1-picryl-hydrazyl-hydrate, DPPH). Water vapor permeability, air transmission rate, peroxide value, and microstructure of treated and original papers were also determined. The experimental results showed that: (i) most components of the peel or peel/leaf extracts were terpenes; (ii) free volume existed among cellulose macromolecule chains of the original paper, occupied by a part of extract materials, and another part of the extracts was formed a thin layer on the paper surfaces; and (iii) air and water barrier properties and antioxidant activity of the treated papers were improved, indicating that the extracts were efficient materials for food packaging applications.

1. Introduction

There is a growing worldwide pressure from researchers, societies and environmentalists on governments to sign an agreement for reduction of sources of pollutions for the Earth. On this route, use of renewable resources instead of synthetic products for food coatings and packaging applications is a subdivision of the global demand. Hence, total or partial replacement of synthetic materials with natural products to minimize the negative environmental impact is a desirable goal for consumers, and the above-mentioned societies and organizations (Tserki, Matzinos, Zafeiropoulos, & Panayiotou, 2006). However, some of these derived materials are expensive and pose an economical problem for manufacturers. In order to reduce material costs, it is often desirable to combine less expensive natural products such as food and agricultural wastes.

Among a variety of natural materials available for food packaging, paper and paperboard materials play a significant role. They possess good mechanical and biodegradable properties and obtain from renewable resources (Conti, 1997). Due to their porous structure and high permeability to moisture and some gases, they are not suitable for packaging of food products with high water contents. Synthetic plastic materials with a hydrophobic nature, such as polyethylene have been used as coating layers for papers to improve their water barrier properties (Petersen et al., 1999). However, the addition of synthetic polymers as thin layers of the paper yield in reduction in their susceptibility to biodegradation (Butkinaree, Jinkarn, & Yoksan, 2008; Shawaphun & Manangan, 2010). Increasing environmental concerns caused by accumulation of plastic wastes, yielded in intensive attention for research and development on moisture barrier layers containing environmentally friendly materials (Despond, Espuche, Cartier, & Domard, 2005; Kjellgren, Gallstedt, Engstrom, & Jarnstrom, 2006). The barrier properties of papers coated with wheat gluten against gases and water vapor were improved (Gastaldi, Chalier, Guillemin, & Gontard, 2007; Gennadios, Weller, & Gooding, 1994). Bordenave, Grelier, Pichavant, and Coma (2007) reported that barrier properties of papers coated with chitosan were improved against moisture. However, the coated papers are not a good candidate for food applications, because of their hydrophilic nature. Parris, Vergano, Dickey, Cooke, and Craig (1998), demonstrated that zein or wax layer effectively increased water vapor barrier properties of

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Kraft papers. Addition of wax to lipid- hydroxyl propyl methyl cellulose (HPMC) composite films reduced their permeability to water vapor (Sothornvit, 2009).

Papers with mainly cellulose based materials have not reached their potential applications in food coatings and packaging, since cellulosic fibers are hygroscopic and moisture absorption result in swelling of the paper, and may lead to destroy easily. Water vapor barrier and mechanical properties are needed for materials, employing for food packaging. Biological based on hydrophobic materials, such as citrus wastes, provide a unique opportunity to incorporate them in the paper and paperboard, in order to enhance water barrier properties and to avoid mechanical properties reduction and finally to avoid their destruction. However, to the best of our knowledge, the citrus wastes have not been used to treat Kraft food-grade paper as a substrate. There is no report in the literature regarding the treatment of food- grade paper with a combination of citrus waste components to improve hydrophobic behavior.

The objective of this study was focused on the ultization of mandarin waste extracts to improve water and gas barriers of Kraft paper. Several steps as follows have been taken in this study: (i) various components of peel and leaf extracts were identified and characterized by GC–MS and FTIR; (ii) antioxidant activities of the extracts were determined; (iii) food-grade Kraft papers were treated with appropriate amounts of peel, leaf, or peel-leaf extracts; (iv) water vapor barrier permeability, air transmission rate and peroxide value (PV) were determined for the original and treated Kraft papers by the extracts; and (v) morphology of the original and treated paper surfaces were also determined.

2. Materials and methods

2.1. Materials

Satsuma mandarin fruits were harvested in Iran (Sari Fajr Gardening Company, Mazandaran, Iran). The leaf was picked from mandarin trees. The peel we removed from mandarin fruits. The fruit and leaf without any physical, mechanical and microbial damages/defects were chosen for this study. Kraft papers (Paper, Selection, USA), were treated using extract materials. Solvents (hexane and chloroform), magnesium chloride, sodium chloride, acetic acid, sodium thio-sulphate, and potassium iodide were purchased (Merck, Darmstadt, Germany). Sunflower oil without any additive (Ghoncheh Company, Sari, Mazandaran, Iran) was used to determine PV of treaded papers at different storage time. The oil was stored to protect from oxidative deterioration at −18 °C, except for the short intervals of measurements. A synthetic antioxidant, butyrated hydroxyl anisol (BHA) provided from Sigma-Aldrich (St. Louis, MO, USA).

2.2. Extraction procedure

Peel and leaf were dried at 30 °C for 72 h in an oven (Vfesuo, Memmert, Germany), equipped with a fan. The dried peel and leaf were then converted into powder separately by a mechanical grinder before the extraction procedures. The powders were stored (within its preparation and extraction) at −20 °C, in order to protect them from any deterioration. The extraction was performed using a mixture of hexane/chloroform (1:1, v/v) at 25 °C for 3 h on a shaker (OL30-ME, OVAN, Spain). The ratio between peel (or leaf) and solvents was (1:4; w/v). The suspended particles remaining in the solutions were removed by filtration and followed by centrifugation (4000 rpm, 20 min.) (Hermle, Labor Technik GMBH, Z 200 A, Germany). The clear solution was then concentrated using a rotary evaporator (IKA, RV10, Germany), under vacuum. The temperature and pressure used to concentrate the extracts by a rotary evaporator were 22 °C and 100 ± 10 mmHg. The period of evaporation procedure was 18 ± 2 min. Under this condition 80% of the solvents were separated by the rota-evaporator, and the solution was concentrated by 80% removal of the solvent. The concentrated solution containing the extracts was used to treat the Kraft paper.

2.3. Gas chromatography–mass spectrometry, GC/MS

The extracts were analyzed by a GC/MS (Agilent 7890A, USA) equipped with a HP-5 ms capillary column (30 m × 0.25 mm, film thickness 0.25 μm). The oven temperature was initially maintained at 50 °C for 3 min, and then, the temperature was raised by a rate of 3 °C/min and finally kept at 185 °C for 1 min. The injector and detector temperatures were set at 250 °C and 280 °C, respectively. Helium was used as a carrier gas with a flow rate of 1.4 mL min^{-1}.

2.4. Fourier transform infrared analysis, FTIR

The chemical structure of the extracts was examined using a FTIR spectrometer (Bruker, Tensor 27, Germany). A few drops of an extract were placed on a thin plate (13 × 7 mm, thickness 3 mm). The spectra were recorded in the range of 400–4000 cm^{-1} at room temperature. The signals were collected with 32 scans and a resolution of 4 cm^{-1}.

2.5. Measurement of antioxidant activity of extracts by DPPH

2,2-Diphenyl-1-picryl-hydrazyl-hydrate (DPPH) was used to evaluate the level of scavenging activity (antioxidant activity) of the extracts by spectrophotometry (Huang, Ou, & Prior, 2005). Measurement of antioxidant activity of the extracts was performed based on the reported method (Oliveira et al., 2008). The procedure

Table 1
Components identified in peel- and peel- leaf mandarin extracts.

	Peel-leaf			Peel			
Peak	R_t (min)	Area	Composition	R_t (min)	Area	Composition	Description for each component
1	7.71	0.99	α- Pinene	7.47	0.38	α- Pinene	A cyclic terpene (C_{10} H_{16})
2	10.04	1.04	β- Myrcene	10.45	1.13	β- Myrcene	A linear monoterpene (C_{10} H_{16})
3	11.50	90.72	Limonene	11.43	87.12	Limonene	A cyclic terpene (C_{10} H_{16})
4	12.42	4.83	γ- Terpinene	12.42	4.7	γ- Terpinene	A cyclic terpene (C_{10} H_{16})
5	23.37	0.36	β- Elemen	22.36	0.35	β- Elemen	A sesquiterpene (C_{15} H_{24})
6	25.42	0.84	Phenol	25.42	0.43	Phenol	An aromatic alcohol (C_6 H_5OH)
7	35.2	0.98	Palmitic acid	35.2	0.73	Palmitic acid	A fatty acid (C_{15} $H_{31}COOH$)
8	24.64	0.24	β-Sabinene[a]	18.67	5.16	Linalyl acetate[b]	

[a] A bicyclic monoterpene.
[b] An esterified oxygenated terpene.