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Cellulose acetate and adsorbents supported on cellulose fiber extracted from waxy corn husks for improving shelf life of frying oil

Patchimaporn Udomkun[a,*], Bhundit Innawong[b], Nitiporn Jumrusjumroendee[b]

[a] International Institute of Tropical Agriculture (IITA), Bujumbura, Burundi
[b] Silpakorn University, Faculty of Engineering and Industrial Technology, Department of Food Technology, Thailand

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ABSTRACT

The efficacy of cellulose paper consolidated with cellulose acetate and mixed adsorbents (bentonite: activated clay: celite = 37.5: 50: 12.5 g with 1 g citric acid/100 g mixed adsorbents) on the physico-chemical properties of oil was evaluated during deep-fat frying of chicken nuggets for 7 days. Cellulose fiber and cellulose acetate were produced from waxy corn husks. Using adsorbents supported on cellulose fiber with or without cellulose acetate resulted in lower poly-unsaturated fatty acids (PUFAs), *trans*-fatty acid, free fatty acid (FFA), peroxide value (PV), and total polar materials (TPMs) compared with results from commercial filter paper (control). The L*, a*, and b* changes of treated oils were better than in control. Limiting the polarity in cellulose acetate might influence TPMs reduction but was more effective in overall quality improvement. The PUFAs, total *trans*-fatty acid, FFA, PV, and TPMs decreased by 1.8, 7.4, 37.0, 133.3, and 20.5%, respectively when compared with control. Therefore, the use of cellulose paper and cellulose acetate obtained from corn husks was found to significantly lengthen the life cycle of frying oil.

1. Introduction

Deep-frying food in hot oil (150–190 °C is a method commonly used in preparing meals, snacks, and fast food from frozen foodstuffs. Generally, high temperatures create the desirable and unique flavor, color, and texture of fried foods. However, frying leads to several oxidative and thermal reactions resulting in the alteration of the physico-chemical, nutritional, and organoleptic properties of the oil (Santos, Molina-Garcia, Cunha, & Casal, 2018; Che Man & Jaswir, 2000) and some of these byproducts such as free fatty acid (FFA), alcohols, cyclic compounds, dimers, and polymers (Tabee, Jagerstad, & Dutta, 2009) have been identified as crucial factors contributing to several health risks (Seppanen & Sarri-Csallany, 2002; Romero, Bastida, & Sanchez-Muniz, 2006, although other commonly identified compounds might not be factors determining biological toxicity (Totani, Burenjargal, Yawata, & Ojiri, 2008). Some crucial health problems after exposure to deteriorated frying oil have been reported such as metabolic alterations (Chen et al., 2008; Chiang et al., 2011), atherosclerosis (Kummerow, 2013; Xian et al., 2012), hypertension (Jaarin, Mustafa, & Leong, 2011), coronary heart disease (Kummerow, 2013), and cancer (Srivastava et al., 2010; Chopra & Schrenk, 2011). Thus, controlling the quality of frying oil is important for health and economic concerns.

In fried food industries, where the same food is usually fried daily in large amounts for many hours, the frying oil is replaced periodically, treated, or discarded to maintain quality. Nonetheless, some amounts of used oils remain in the fryer and are adsorbed by every single fried product. To reduce the detrimental effects and to prolong the usable life of frying oil, several scientific studies have addressed the safety of the used oils (Somnuk, Innawong, & Tirawattannawanich, 2013) and claimed that regular cleaning and maintenance of equipment, good quality frying fat, and proper frying conditions are required (Stevenson, Vaisey-Jenser, & Eskin, 1984).

Filtration has been also used to recover deteriorated frying oil after use, thereby extending its lifespan. Passive filtration, wherein the used oil filtrates pass through a filter paper or cloth, is used to slow oil breakdown by limiting the surface area (insoluble particles) available for attachment, thereby reducing oil deterioration (Akoh & Reynolds, 2001). However, although coarse particles can be separated in this manner, very fine particles, chemical products, and/or water remain in the oil. There is intense interest in active filtration through the adsorptive property of single or several natural and synthetic adsorbents (Bhattacharya, Sajilata, Tiwari, & Singhal, 2008; Lin, Akoh, & Reynolds, 2001). When these adsorbents are in contact with oil, some of the polar degradation products, such as peroxides, aldehydes, ketones, hydroperoxides, polymers, and oxidized monomers that occur during deep frying, are held on the adsorbents by physical and/or chemical forces.

* Corresponding author.
E-mail address: P.Udomkun@cgiar.org (P. Udomkun).

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The main parameters that affect adsorption efficiency are contact time, temperature, and the amount of adsorbent per treatment (Bheemreddy, Chinnan, Pannu, & Reynolds, 2002; Miyagi & Nakajima, 2003; Sonkaew & Chaisena, 2012). The most popular adsorbents for the regeneration of frying oils are activated carbon, magnesium oxide, diatomaceous earth, bleaching earth, calcium silicate, and various forms of silica.

Many reports are available on the effect of adsorbents in controlling oil quality. For example, Mancini-Filho, Smith, Creveling, and Al-Shaikh (1986) showed that the dielectric constant changes (DCC) obviously improve with the utilization of bleaching clay and charcoal. McNeill, Kakuda, and Kamel (1986) also examined the feasibility of various mixtures between activated carbon and silica for improving the quality of used canola oil. Oil treated with mixed adsorbents was reported to show more effective reduction in acid values (AV), saturated and unsaturated carbonyl contents, peroxide values (PV), total polar components (TPC), and photometric color than the control (without treatment). These results might be due to the greater capability and higher adsorption power of mixed adsorbents to react with the chemical compounds in the used oils. In addition, Lin et al. (2001) reported reductions of final values in used oil by a mixture of synthetic chemical absorbents (3% Hubersorb600, 2% Frypowder, and 3% Magnesol) after daily treatment for 4 days compared to the untreated control.

The need for environmentally friendly materials and processes has motivated much research in the use of raw materials from agricultural waste because this represents an abundant, inexpensive, and readily available source. Cellulose is one of the useful fibers from agricultural waste and is widely used in many industries owing to the presence of functional groups such as hydroxyl, carboxyl, methoxy, phenols, etc. (Hassanein & Koumanova, 2010). Apart from cellulose fiber, cellulose acetate is an esterified cellulose derivative widely known for its numerous applications in membrane separation processes, production of matrices for controlled drug release (membranes or particles), protection of optical films, cigarette filters, among other applications (da Silva et al., 2017). Cellulose acetate is easy to process and biocompatible, with the ability to change its hydrophilic/hydrophobic characteristics through acetylation/deacetylation (Zhou et al., 2016). Hence this polymer presents attractive properties for its use in the incorporation of active compounds such as adsorbents.

The preliminary study showed that the quality of cellulose fiber and cellulose acetate obtained from waxy corn husks was better than that from sweet corn husks, rice straw, and bagasse owing to higher holocellulose and alpha-cellulose contents. This indicated its potential for extracting cellulose in the process of frying oil filtration. Therefore, this study aimed at improving the overall quality of frying oil by using cellulose filter paper together with cellulose acetate from waxy corn husks and mixed adsorbents. The physico-chemical characteristics were studied in detail to evaluate the potential of value-added products from agricultural waste for improving the quality of used oil.

2. Materials and methods

2.1. Sample preparation

Waxy corn husks from Nakhon Pathom province, Thailand, were boiled in water at 100 °C for 30 min and then hot air dried at 60 °C for 24 h to moisture content of 7.5 ± 0.5 g water/100 g. The dried sample was cut into pieces of 3–5 cm prior to the steam explosion process.

2.2. Steam explosion process

The waxy corn husk sample was loaded into a 2.5 L stainless steel reactor (model 316, Nitto Koatsu Co. Ltd., Tsukuba, Japan) and treated with saturated steam at 215 °C and a pressure of 21×10^6 Pa for 5 min. The steam exploded sample was separated by filtration, thoroughly washed with water, centrifuged to eliminate fluid and then hot air dried at 70 °C for 24 h to moisture content of 6.5 ± 0.5 g water/100 g.

2.3. Cellulose acetate preparation

To extract cellulose, 25 g of sample after steam explosion was first soaked in 500 mL of 5 g/L NaOH solution then placed in a water bath at 80–90 °C for 1 h; subsequently, it was cleaned with water and hot air dried at 55 °C for 24 h. The dried sample was ground to obtain cellulose powder and sieved with 250 μm mesh size.

Cellulose acetate was produced by modifying the method of Biswas, Saha, Lawton, Shagren, and Willett (2005) that involved weighing 2 g of the cellulose powder into a 100-mL round bottomed flask with 0.5 mL acetic acid, 5 g acetic anhydride, 30 mL methylene chloride, and 0.04 mL of 18.4 mol/L concentrated sulfuric acid. The mixture was shaken at 80 °C for 4 h using a water bath. The sample was cooled at room temperature then filtered through Whatman No. 1 paper. The extracted was mixed with 60 mL chloroform, stirred at room temperature for 30 min, and filtered under vacuum conditions. The collected solution was entirely evaporated to dryness in a rotary evaporator (model V-500, Büchi, Postfach, Switzerland). After that, 20-mL ethyl alcohol was added to dissolve cellulose acetate before vacuum evaporation. The collected cellulose acetate on the filter paper was hot air dried at 80 °C for 24 h.

2.4. Preparation of filter paper

In this study, four different adsorbents were used: celite (Celite® coarse 545, Flukachemical, Buchs, Switzerland); activated clay (Power Dry Co., Ltd., Bangkok, Thailand); bentonite (Sigma Aldrich Chemica GmbH, Hamburg, Germany); and citric acid. Celite is a white, odorless powder composed of silicon dioxide (SiO_2). Activated clay is gray granules, odorless, insoluble in water, and composed of calcium alumino silicate. Bentonite is an odorless, light-brown powder composed of silicon dioxide (SiO_2) and aluminum dioxide (Al_2O_3). The adsorbent combinations consisted of bentonite, activated clay, and celite at the ratio 37.5: 50: 12.5 g with 1 g citric acid/100 g mixed adsorbents. The selection of the types of adsorbent and their appropriate ratio was based on the cost and efficiency of adsorbents in the preliminary study.

To prepare the cellulose filter paper for three different treatments, cellulose extracted from waxy corn husks was mixed with distilled water in the ratio of 1:35 (g/mL) using a blender (model DDF 341, Moulinex, Paris, France). The control was Whatman No. 4 filter paper without the addition of adsorbents and cellulose acetate. Treatment I was cellulose paper with the addition of 35 g mixed adsorbents/100 g cellulose paper. Treatment II was cellulose paper with the addition of 30 g mixed adsorbents and 5 g cellulose acetate/100 g cellulose paper. The filter paper with a mixture of cellulose suspension with and without adsorbents and cellulose acetate was spread on a sieve, dried at room temperature for 24 h, removed from the sieve and prepared for testing. The scanning electron micrographs of cellulose paper with and without adsorbents are shown in Fig. 1.

2.5. Frying experiments

Chicken nuggets were purchased from CPF Food Products Co. Ltd., Lopburi, Thailand and palm oil was obtained from Morakot Industry Co. Ltd., Bangkok, Thailand. The frying process used a commercial dual-unit electric batch fryer (model IV, Protech Co. Ltd., Bangkok, Thailand) with a capacity of 5 L of frying oil. Before frying, fresh oil was preheated at 175 °C for 10 min to simulate normal frying conditions. The oil was maintained at ± 5 °C of the set temperature (175 °C) using a programmable temperature controller. No fresh oil was added during frying. However, at the beginning of the next frying date, a small amount of fresh oil (about 200 mL/day) might be added in the fryer to keep the initial oil level constant.

Each batch of 100 g chicken nuggets was randomly fried at a setting temperature of 175 °C in 2 L of heated oil. For each cycle, the samples were placed in a stainless steel basket to keep them submerged and