Natural Cellulosic Material Characteristics: A Possibility to Develop Antimicrobial Active Fiber-based Packaging

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Coconut husk, rubberwood sawdust, and palm leaf base are cellulosic agricultural wastes that have potential to be processed to fiber as absorbing material. This study investigated characteristics (morphological, physiological properties, chemical composition, absorption capacity, and water absorption isotherms) of coconut, rubberwood, and palm fiber. Also, the study aimed to develop an antimicrobial sachet packaging to resist against foodborne pathogens (*Listeria monocytogenes*, *Staphylococcus aureus*, *Salmonella* spp., and *Escherichia coli*) by adding lime oil (LO) emulsion or *Litsea cubeba* (LC) oil at 50 to 700 µL into the material (1 g) before, and then dried and placed in the 1-L seal box. Results showed that among the three, coconut performed the best in terms of releasing the essential oil (EO) emulsion against bacteria. Coconut could adsorb and release volatile LO or LC at the lowest concentrations (LO, 500 µL/L; LC, 300 µL/L) to inhibit bacteria compared with the other fibers (700 µL/L) at 35 °C. Results indicated that coconut has a low water absorption rate, which influenced the faster adsorption of EO emulsion in the beginning of the process; therefore, using low concentrations of EO in coconut for bacterial inhibition is possible. Coconut contains 34.5% lignin, 68.7% holocellulose, 37.6% cellulose, and 31.2% hemicellulose. Coconut is suitable as an alternative to the biocomposite material in developing a new antimicrobial packaging design.

Keywords: Natural cellulosic material; Water absorption; Essential oil emulsion absorption; Morphological properties

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INTRODUCTION

In recent years, many research attempts have been made to produce new polymers including sustainable polymers using renewable natural resources (Zhu *et al*. 2016). Additionally, the use of natural polymers, which are normally biodegradable, is an important design parameter for opening up new market opportunities to develop value-added products from natural polymer feedstocks. Such biodegradable materials are preferably combined with renewable raw materials, such as cellulose, proteins, and polysaccharides extracted from agricultural, marine, microbial sources, or animal sources,

because those materials can degrade in the environment into simple substances and become biomass products (Malathi *et al*. 2014).

In Southeast Asia, southern Thailand stands out in the production of various types of agricultural crops, including coconut, rubberwood, and palm waste. The farming sectors generally produce a large amount of agricultural waste, so it is important to develop a method for disposing of these wastes to add value. For instance, value could be added by improving properties of these fibres *via* adding antimicrobial agent. Such an approach can involve application of the material as an antimicrobial absorbent, promoting the use of natural products, and assuring good environmental management to support a circular economy. In the literature, good properties of natural cellulosic materials are well established. Their intrinsic features, such as structural aspects and tensile and chemical properties, are key points to identify properties and applications. These properties have received increasing attention in investigations on natural materials. However, contrary to the research on jute, coconut, sisal, flax, and bamboo fibers (Li *et al*. 2000; Ali 2011; Khalil *et al*. 2012; Yan *et al*. 2014; Pickering *et al.* 2016), research on coconut, rubberwood, and palm is rare. Available knowledge, exploration, and utilization of rubberwood, palm, and coconut remain limited, including research on water adsorption and isotherm properties for developing a biodegradable food packaging. Therefore, studying the material characteristics of such natural fibers is important to evaluate their effective exploitation in the food industry, particularly in the food-packaging sector.

Antimicrobial packaging is a system that kills or inhibits the growth of microorganisms, extends the shelf life of perishables, and enhances the safety of packaged products. Such packaging is one of the most promising innovations of active packaging technologies. It can be constructed using antimicrobial packaging materials or antimicrobial agents or both in the packaging space. Therefore, various antimicrobial compounds can be combined with different types of packaging materials such as films, paper, and bioplastics (El-Sakhawy *et al*. 2018; Sofi *et al*. 2018). However, in recent years, because consumers' demand for natural food ingredients has increased due to food safety and availability, these natural compounds are beginning to replace the classical chemical agents for food preservation and are recognized to be safer and claimed to relieve safety concerns. Essential oils (EOs) are well-known antimicrobial agents and can be used to control food spoilage, foodborne pathogens, and toxin-producing microorganisms in foods (Marino *et al*. 2001; Burt 2004). Many researchers have tested EOs, including lime oil (LO) and *Litsea cubeba* (LC), adding them to packaging materials to obtain antimicrobial packaging (Muriel-Galet *et al*. 2013; Ahmed *et al*. 2018; Arfat *et al*. 2018; Suhem *et al*. 2019). Furthermore, LO and LC have been used to enhance food flavor (Valenzuela *et al*. 2016).

In this study, natural cellulosic materials of coconut, rubberwood, and palm fiber were characterized by means of water and EO emulsion absorption capacity, water adsorption isotherms, and morphological studies. Additionally, the water and LO emulsion adsorption capacity was also investigated and compared in finding the most suitable absorbent to develop biodegradable antimicrobial packaging.

EXPERIMENTAL

Materials

Natural cellulosic material preparation

Coconut (*Cocos nucifera*) fiber from a local coconut husk, palm leaf base (*Elaeis guineensis* Jacq.) from a local palm tree, and rubberwood *(Hevea Brasiliensis*) sawdust from Nakorn Sri Parawood Co., Ltd., Nakhon-Si-Thammarat province, Thailand were used in this study. These materials were cut into small pieces (~20 wide×20 long ×10 mm thickness) and a small powder (0.05 ± 0.01 m) were produced by a sifter machine, then separated by having passed through a stainless sieve mesh number 20 (Endecotts Ltd., London, UK). For fibre extraction, these powders were heated with 1.5 N NaoH solution at 70 °C for 5 h, then were washed with water and air oven at 100 °C for 2 h and stored in a desiccator.

Chemicals used in the study

The LO and LC oil were obtained from Thai China Flavors & Fragrances Co., Ltd., (Bangkok, Thailand) and kept refrigerated (4 °C) until use.

The EO emulsions (50 to 700 µL L^{-1}) were prepared with LO or LC oil and polyethylene glycol sorbitan monooleate (Tween 80; 2%, w w^{-1}; Toriko Co., Ltd., Bangkok, Thailand). Then, the mixture was homogenized at 20,000 rpm min^{-1} for 10 min.

Bacterial strains and culture conditions

Listeria monocytogenes, *Staphylococcus aureus*, *Salmonella* spp., and *Escherichia coli* strain samples were obtained from the Department of Allied Health Sciences (Walailak University, Thai Buri, Thailand). The strains were maintained as freeze-dried stocks at -80 °C and cultured twice in Tryptic Soya Broth (Oxoid, Chesire, UK) for 24 h at 35 °C with shaker incubator before the experiments. The bacterial population in all of the inoculated media was standardized to 10^4 cell mL^{-1} by dilution with peptone water, and the viability of all strains was assessed using quantitative colony counts at 10^4 CFU mL^{-1}.

Methods

Morphological characterization of natural cellulosic material

Images of particle fibers at mesoscale level were acquired on a Zeiss Axio Zoom.V16 stereo microscope equipped with a Zeiss Axiocam 503 mono CCD camera, using the 1x 0.25 NA objective, and the bright field optics (Zeiss, Oberkochen, Germany). The stereo microscope offers an adequate magnification and depth of field for studies at the mesoscale.

The fiber ultrastructure was examined by scanning electron microscopy (SEM) (Zeiss, Oberkohen, Germany). Samples selected by stereo microscopy as representative were mounted on circular specimen holders (Agar Scientific, Plain stubs 12 × 12 mm, (Agar Scientific, Essex, UK) with double carbon tape (Carbon Tabs 9 mm, Agar Scientific, Essex, UK) and gold coated with a sputter coater (SEMPREP2, Nanotech, England, UK). Observations were carried out using a Zeiss LEO 1430 SEM at 5 kV (Zeiss, Oberkochen, Germany).

Physiological properties

The moisture content of the cellulose material was determined according to the Association of Official Analytical Chemists (AOAC) standard (1999). Then, it was calculated and expressed as a percentage of moisture content.

Water activity was measured using an electronic hygrometer (CX-2-Decagon Devices, Aqua Lab, Pullman, WA, USA), based on the determination of the dew point and previously calibrated with standard solutions of LiCl and NaCl of known activity (prepared by High-Purity Standards for Decagon Devices).

Chemical composition

The chemical composition was analyzed according to the Technical Association of the Pulp and Paper Industry (TAPPI) T222 om-02 (2006) (acid-insoluble lignin in wood and pulp) and the TAPPI T203 (1999) (α-, β-, and γ- cellulose in pulp) standard methods and the method described by Wise *et al.* (1946) (holocellulose in pulp).

The ash content of the material was investigated using a standard method of American Society for Testing and Materials (ASTM) D2866-94 (2004). Then, it was calculated and expressed as a percentage of ash content.

For each experiment, the triplicate samples were determined ($n = 3$).

Fourier transform infrared spectroscopy (FTIR)

The structural characteristics and chemical bonding of cellulose material were determined by a PerkinElmer FTIR SpectrumTM 100 series spectrometer (PerkinElmer, Waltham, MA, USA) with the single-reflection sampling plate of a 1.8-mm round germanium surface. Spectra were recorded at room temperature within the range of 650 to 4000 cm^{-1} at 4 cm^{-1} resolution. Spectrum 6.0 software (PerkinElmer, Waltham, MA, USA) was used for data acquisition and analysis.

Brunauer–Emmett–Teller (BET) and Barrett-Joyner-Halenda (BJH) analysis

The BET theory was applied to determine the specific surface area and the percentage of porosity of materials. The total pore volume was determined by converting the amount of N_2 absorbed at a relative pressure of 0.994 to the volume of liquid nitrogen. Pore size diameter was considered based on the Barrett-Joyner-Halenda (BJH) theory.

Water and emulsion absorption capacity

For water and emulsion absorption capacity determination as described follows Najafi *et al.* (2007). For this, 1 g of the sample was mixed with 10 mL distilled water or emulsion and kept at room temperature (30 °C). For each measurement, samples were removed from the water or emulsion. The samples were filtrated by Whatman filter paper No. 1, which was connected to the vacuum pump. Then, the surface water or emulsion was wiped off using blotting paper. Weight of the sample was measured at different times during the prolonged immersion. The measurement was terminated after the equilibrium water absorption of the sample was reached. The values of the water absorption in percentage were calculated using the following equation,

$$W(\%) = [(W_t - W_0) \times 100]/W_0 \quad (1)$$

where W_t is the weight (g) of sample at a given immersion time and W_0 is the oven dried weight (g). Triplicate determinations were performed for each experiment ($n = 3$).

Water absorption isotherms

The samples (in triplicate) were dried at a proper temperature to a constant weight and stored in air-tight plastic boxes. Then, the salt solutions, which included CH_3CO_2K, $CaCl_2$, $C_2H_3LiO_2$, $(NH_4)_2SO_4$, $BaCl_2$, and K_2SO_4 of known relative humidity (% RH): 23, 34, 71, 81, 90, and 97% RH (Greenspan 1977), respectively, were weighted and mixed with distilled water in a closed container until a saturated point was reached. Approximately 1 g of each material (coconut, palm, and rubberwood) was weighed into a plastic cup and placed in the plastic container containing saturated salt solutions. The containers were kept in an incubator at 25 °C. The weighing was done every week until apparent equilibrium was reached. Anhydrous calcium chloride was used to control for checking the dryness of samples. The weight of the sample in a container, after equilibration time, was used to calculate the initial dry matter content of a mixture (Lewicki 1997). The values of the sample moisture content were calculated using Eq. 2,

$$M = [W_2 - W_1] + \frac{\%H_2O}{100} \times W_1 / [W_1 \times \frac{100 - \%H_2O}{100}] \quad (2)$$

where W_1 is the initial weight (g) of the sample, W_2 is the final weight (g) of the sample, and $\%H_2O$ is the initial moisture content of the sample. Triplicate determinations were performed for each experiment ($n = 3$).

The effect of EO emulsion contained in natural cellulosic material against foodborne pathogens

For the material loading, natural cellulosic fiber (1 g) was weighed and soaked in 10 mL of LO or LC oil emulsion at different concentrations (50 to 700 µL L^{-1}) for 2 h. The mixture was treated using the homogenized at 20,000 rpm min^{-1} for 10 min to capture the oil. The material was then filtered using filter paper and dried with a dehydrator (Septree Multi-functional Food Dehydrator; Foshan Dalle Technology, Guangdong, China) at 35 °C for 4 h. After that, the treated fiber was packed into the tea filter sachets and placed on the lid of a 1-L sterile plastic box. A total of 100 µL of each pathogen (*L. monocytogenes*, *S. aureus*, *Salmonella* spp., and *E. coli*) were dropped onto the nutrient agar (NA). Then, the agar plate was put inside the sterile plastic box. All of the treatments were incubated at 35 °C ± 2 °C for 1 month, and the lowest concentration with no bacterial colony growth on the NA plate was reported to minimum inhibitory concentrations (MICs). Control was done without an EO in fiber.

Statistical analysis

All results were expressed as the mean ± standard deviation. One-way analysis of variance (ANOVA) and Duncan's *post hoc* test, with $p < 0.05$ as statically significant, were applied in the statistical analysis achieved using Statistica software (StatSoft Inc., version 11, Tulsa, OK, USA).

RESULTS AND DISCUSSION

Natural Cellulosic Material Characteristics

Morphological characteristics

Investigation of morphological properties of particles of the three fibers of this study was carried out by stereo microscopy (Fig. 1). Particles were shown to be extremely heterogeneous in size and shape among the different fibers as well as within each fiber type. Coconut showed a particle length ranging from 1.40 to 3.15 mm with a generally elongated shape, thus suggesting a low surface/volume ratio. Particles could be divided into fine (~250 to 300 µm), medium (~301 to 450 µm), and thick (> 451 µm) calculated by linear density as already observed by da Costa *et al*. (2013). Rubberwood showed both powdery particles and more elongated fibers with a particle size ranging from 1.05 to 2.10 mm. Palm showed two different particle types, which are the sponge and long fiber structures. The rubberwood and coconut long fibers showed to have a rough surface, less evident in long fibers of palm.

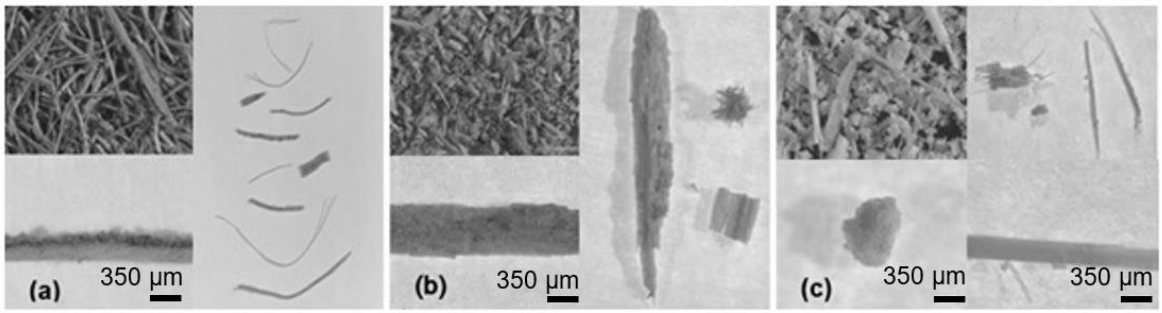

Fig. 1. Images of coconut (a), rubberwood (b), and palm (c) fibers obtained by stereo microscopy

Fibers were observed at higher magnification by SEM (Fig. 2). Palm showed the most different structure compared with coconut and rubberwood. Its external surface was characterized by a scale-like organization, as previously observed by Zhang *et al*. (2015), that determines an irregular surface. The phloem tissue is evident inside the fiber, and it consists of elongated hollow fiber cells (FCs) that exhibit a honeycomb-like structure. Figure 2b, that is an enlarged frame of the honeycomb organization, shows that FCs comprise the lumen (L) and the cellular wall, this was last known to be organized in the primary and secondary wall (Zhang *et al*. 2015). The FCs were parallel, unidirectional, and connected each other through the middle lamella (ML, Fig. 2b) that consists of lignin and hemicellulose. Such ultrastructure is reported to provide palm with potential use in adsorption technology. Figure 2c shows the surface of coconut with some globular protuberances placed at regular intervals on the surface. These protuberances have been reported to be clusters of calcium oxalate crystals (CO, Fig. 2c) and also wax deposits (Lomelí-Ramírez *et al*. 2014). In contrast, rubberwood showed a woven structure with some empty spaces as well.

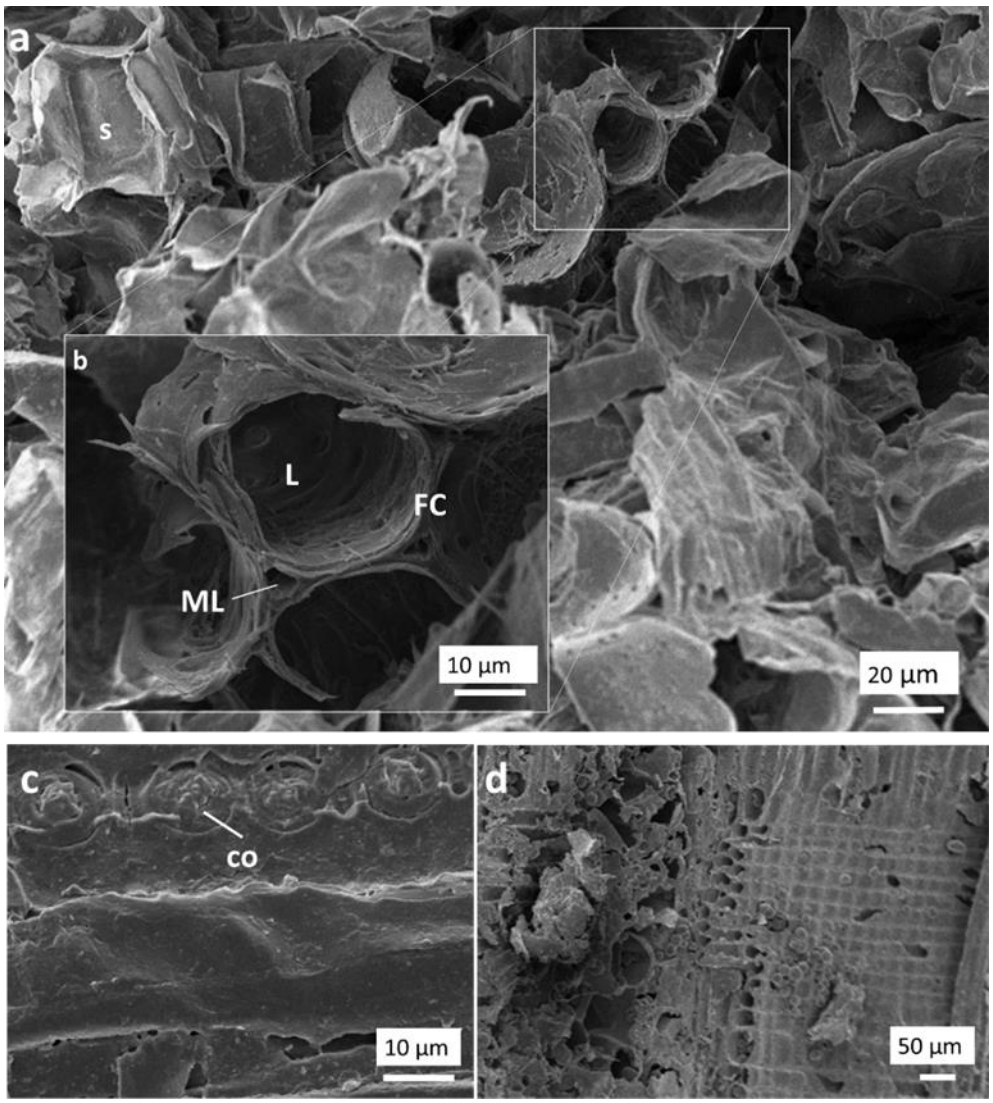

Fig. 2. SEM images of palm (a), coconut (b), and rubberwood (c) fiber. A higher magnification of the area framed in (a) is shown in (b) where the fiber cells (FC) are clearly visible together with the middle lamella (ML) and the lumen (L). Scales (S) are present on the surface of palm. Globular protuberances on the surface of coconut are clusters of calcium oxalate crystals (CO).

Physiological Properties

The physiological properties of natural cellulosic materials are shown in Table 1. The average diameters of coconut, palm, and rubberwood were ~171.72, 243.72, and 133.39 µm, respectively. Palm had the highest percentage of moisture content (6.30%), followed by rubberwood (4.17%), and coconut (2.70%). The moisture content is an important aspect that should be considered when determining if the natural material could be an appreciated filler for polymer composites. It has been reported that the stability, dimensions, porosity formation, and tensile strength of biocomposites can be weakened if the biocomposites have high-moisture content (Alvarez *et al*. 2004; Réquilé *et al*. 2019). Accordingly, the lowest moisture content is required. However, there were no significant differences in water activity of natural cellulosic materials ($p>0.05$) due to the properties of natural fiber which are normally hydrophilic in nature presence of the same hydroxyl groups (Rozyanty *et al.* 2021). Coconut (0.67-0.1 g cm^{-3}) had the lowest density followed

by palm and rubberwood, respectively. In comparison, low density but high porosity has been considered as an essential oil absorbent. However, other properties such as surface area, pore area could also cause to essential oil absorption. Therefore, one of the most crucial factors that need to be considered when selecting new materials as fillers in polymer composites is their weight. This characteristic, probably more than others, affects the fulfillment of the final product.

Table 1. Physiological Properties of Cellulose Material

Fibers	Diameter (µm)	Moisture Content (%)	Water Activity (a_w)	Density (g cm^{-3})
Coconut	171.72 ± 1.35^b	4.17 ± 0.25^b	0.55 ± 0.10^a	0.67 to 1.0
Rubberwood	133.39 ± 1.50^c	2.70 ± 0.10^c	0.55 ± 0.01^a	1.50
Palm	243.72 ± 1.87^a	6.30 ± 0.24^a	0.54 ± 0.00^a	0.71
Note: a through c: Mean in each roll with different superscript letters are significantly different ($p < 0.05$)				

Chemical Composition

The chemical composition of cellulosic material is shown in Table 2. Rubberwood had the highest percentage of cellulose (41.31%), followed by coconut (37.55%), and palm (31.60%). Moreover, coconut had the highest percentage in both hemicellulose (31.18%) and lignin (34.15%). Rubberwood and palm hemicellulose content was at 30% and 22.18%, respectively. Palm had the lowest percentage of lignin (8 %), whereas rubberwood had 22.87% of lignin. In contrast, rubberwood showed the highest percentage of holocellulose (71.31%), followed by coconut (68.73%), and palm (53.78%). For the ash percentage, palm had the highest percentage at 5.63%, followed by coconut (1.60%), and rubberwood (1.50%).

Table 2. Chemical Composition of Cellulose Material

Fibers	Cellulose (%)	Hemicellulose (%)	Holocellulose (%)	Lignin (%)	Ash (%)
Coconut	37.55 ± 0.15^b	31.18 ± 0.33^a	68.73 ± 0.79^b	34.51 ± 1.82^a	1.60 ± 0.32^b
Rubberwood	41.31 ± 0.24^a	30.00 ± 0.47^b	71.31 ± 1.25^a	22.87 ± 2.27^b	1.50 ± 0.41^b
Palm	31.60 ± 0.27^c	22.18 ± 0.51^c	53.78 ± 0.64^c	8.66 ± 1.35^c	5.63 ± 0.17^a
Note: a through c: Mean in each roll with different superscript letters are significantly different ($p < 0.05$)					

The main chemical components of cellulose materials are cellulose, hemicellulose, holocellulose, lignin, and ash. Normally, the natural polymers from wood contain hydroxyl groups, which could be formed between hydrogen and water bonds, causing a high rate of water uptake in cellulosic materials. Hemicellulose is the most hydrophilic and unstable of the wood polymers and has a great influence on water absorption (Zhang *et al*. 2015), which is also shown in the results of this study in which coconut having the highest hemicellulose has the highest water adsorption capacity than any other materials (see water adsorption capacity property).

FTIR Spectroscopy Analysis

The spectra of the functional groups in the FTIR results are shown in Fig. 3. The FTIR spectra showed that all cellulosic materials in this test contain xylan in hemicellulose because the band appeared at 1762 to 1719 cm^{-1}, which was attributed to the C=O stretching of the acetyl and uronic ester groups of hemicellulose (Carballo-Meilán *et al*. 2016; Cichosz *et al*. 2019). Moreover, the peaks in the 1279 to 1274 cm^{-1} region were investigated with the C–H bending of cellulose. The bands observed at 1132 to 950, 1245 to 1220, and 1687 to 1385 cm^{-1} correspond to lignin, lignin and xylan, and tannin, respectively. The intense peaks at 3500 to 3200 cm^{-1} indicated O–H groups in all materials because of the appearance of hydroxyl groups in cellulose, hemicellulose, and lignin related to the different water adsorption results found in this study. Finally, the peaks at 1800 to 1600 cm^{-1} implied carbonyl groups (C=O) in lignin and hemicellulose (da Silva *et al*. 2016). Therefore, all FTIR spectrum bands acted from natural materials, which can be related to the chemical composition analysis of the natural cellulosic material reported here.

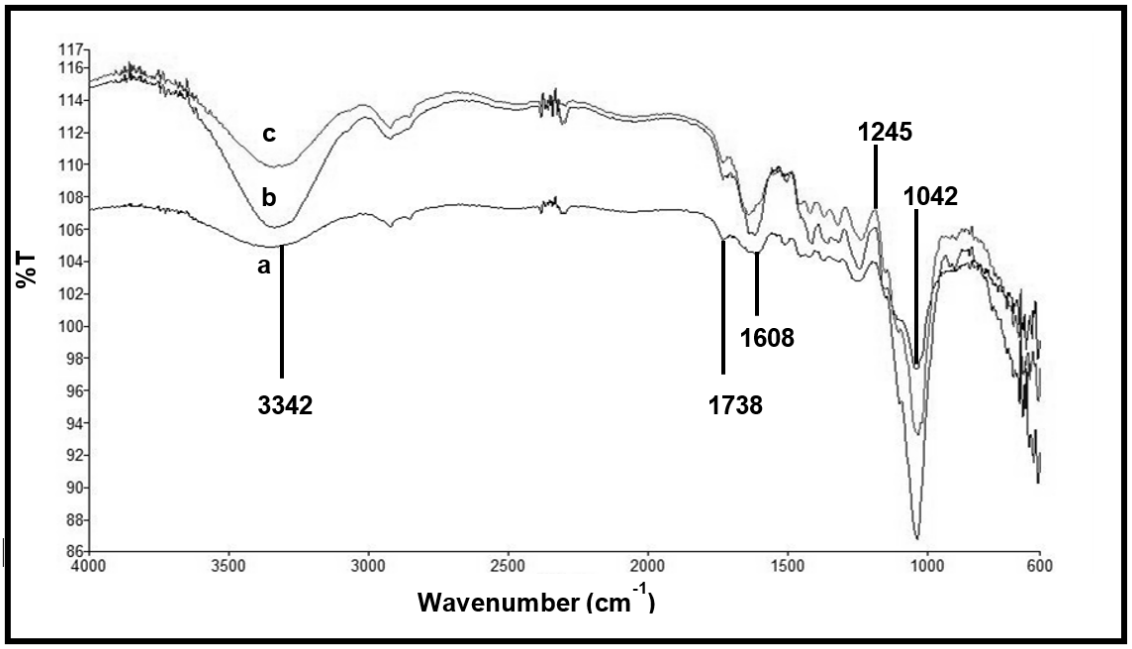

Fig. 3. FTIR spectra of the (a) coconut, (b) rubberwood, and (c) palm

Brunauer–Emmett–Teller Analysis

The average pore size and specific surface area of the material is demonstrated in Table 3. Coconut had the highest percentage of porosity (3.3%) compared with that of palm (2.9%) and rubberwood (1.9%). Palm had the highest BET specific surface area (1.96 m^2 g^{-1}), followed by coconut (1.92 m^2 g^{-1}), and rubberwood (1.30 m^2 g^{-1}). The BET specific surface area shows the complex function of material fineness, fiber fibrosis degree, and fiber length. However, BET specific surface area of fiber is related to the inner surface area of the pore of the fiber cell wall (Chen *et al.* 2012). Palm had the highest BET surface area in this study and had the highest emulsion absorption (see emulsion absorption) due to a large surface area, which plays a key role in the fast elimination of emulsion that are in agreement with the report of Haghbin and Shahrak (2021).

Table 3. Percentage of Porosity and Surface Area of Natural Cellulosic Material

Fibers	Porosity (%)	Specific Surface Area (m^2 g^{-1})
Coconut	3.3 ± 0.1[a]	1.92 ± 0.02[a]
Rubberwood	1.9 ± 0.3[c]	1.30 ± 0.09[b]
Palm	2.9 ± 0.1[b]	1.96 ± 0.13[a]
Note: a through c: Mean in each roll with different superscript letters are significantly different ($p < 0.05$)		

Barrett-Joyner-Halenda Analysis

The BJH analysis can also be employed to determine the pore area and specific pore volume using adsorption and desorption techniques. This technique characterizes pore size distribution independent of the external area due to the particle size of the sample. As shown in Fig. 4, the pore size distributions and pore volume were similar for all the samples at 8.0 to 14.0 nm regions. As is known, a pore size less than 2.0 nm is a micropore, whereas between 2.0 to 50.0 nm it is a mesopore (Kimura *et al*. 2016). The peaks that were seen at 4.0 to 7.0 nm, showed palm has a higher mesopore volume in this region in comparison to rubberwood. The highest volume of pores present for palm and coconut (cm^3 g^{-1}) are in the 2.0 to 4.0 nm region, and hence finer mesopores are the dominating pores for these materials. This result can explain why palm and coconut are the best absorbent compared to rubberwood.

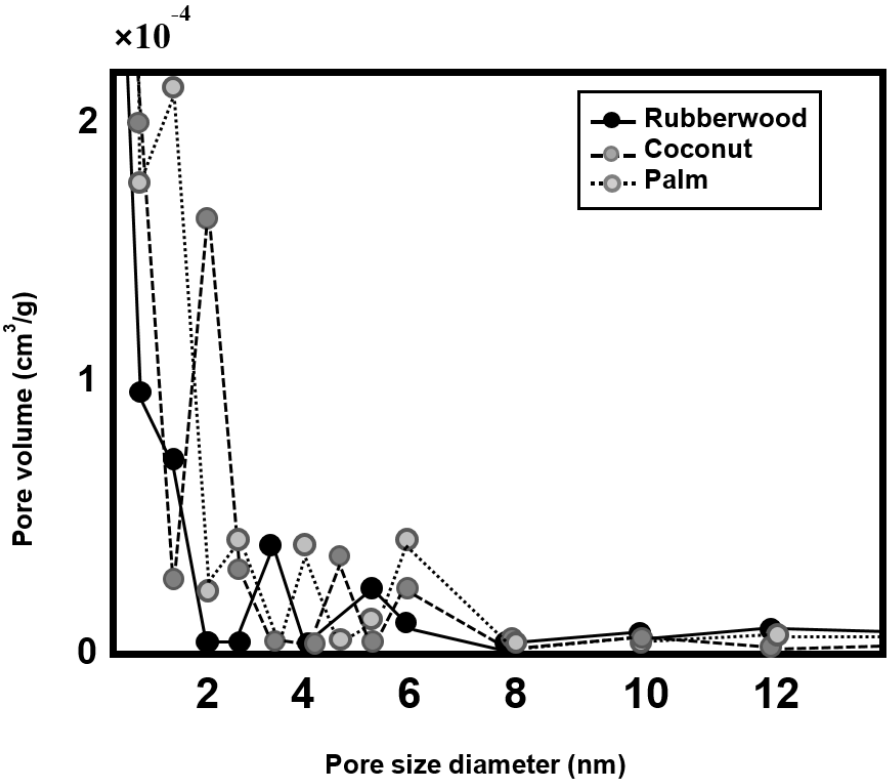

Fig. 4. The BJH pore size distribution of natural cellulosic materials

Water and EO Emulsion Absorption Capacity

The water and emulsion absorption capacity of the material is shown in Fig. 5. It was found that coconut has the highest percentage of water absorption, with an absorption capacity of 70%, followed by palm (50%), and rubberwood (30%).

As reported in previous research, it was found that each natural cellulose material has different chemical compositions and affects mechanical properties and their potential ability as food-packaging materials (Espert *et al*. 2004; Al-Haidary *et al*. 2011; Rowell 2012).

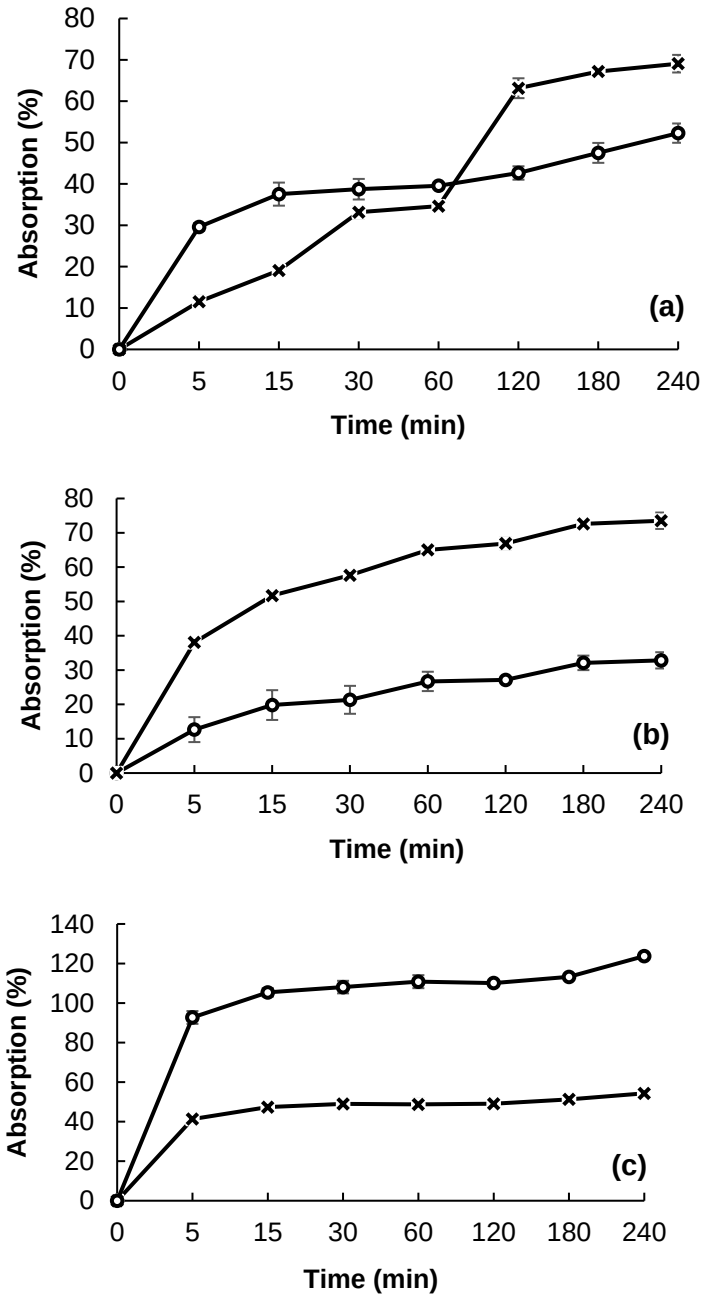

Fig. 5. Percentage of water (-x-) and emulsion (-o-) absorption for (a) coconut, (b) rubberwood, and (c) palm

For emulsion, palm is the best absorbent material, being able to absorb LO emulsion at 60% to 70%, followed by coconut (50% to 60%), and rubberwood (30% to 40%). In addition, bulk density after LO absorption of coconut, rubberwood, palm were 0.37±0.01, 0.23±0.01, and 0.76±0.01 g cm^{-3}, respectively. Palm had highest in bulk density due to a pore size with potential use in EO adsorption (Fig. 2). Zhang *et al*. (2015) reported the porous characteristics of palm compared with coconut. Palm has a lower surface area than coconut because the majority of pores in palm are mesopores (diameter: 2 to 50 nm), whereas coconut has a micropore volume of 66%.

Water Adsorption Isotherms

The materials were determined at 25 °C using a gravimetric method. Several saturated salt solutions were selected to obtain different water activities in the range from 0.23 to 0.97. The results are shown in Fig. 6. The highest water activity (0.97) showed that palm absorbs water to equilibrium moisture content better than rubberwood and coconut. This means that coconut and rubberwood have good qualifications to be the biocomposite material in food packaging because they do not absorb water as fast as palm does. Using BET analysis, palm had the highest porosity and specific surface area; therefore, palm had the highest water adsorption isotherms in this study. Additionally, the OH-group (see FTIR section) in all materials could be related to the water absorption in this study. In addition, the OH-group is also related to OH from cellulose, hemicelluloses, and lignin (see chemical composition section). The nature of water sorption to different chemical composition of materials has always been a complex matter due to hydrogen-bond formation to all sorbing sites, which can be considered for adsorption of water to the different material Chaouki *et al.* (2021).

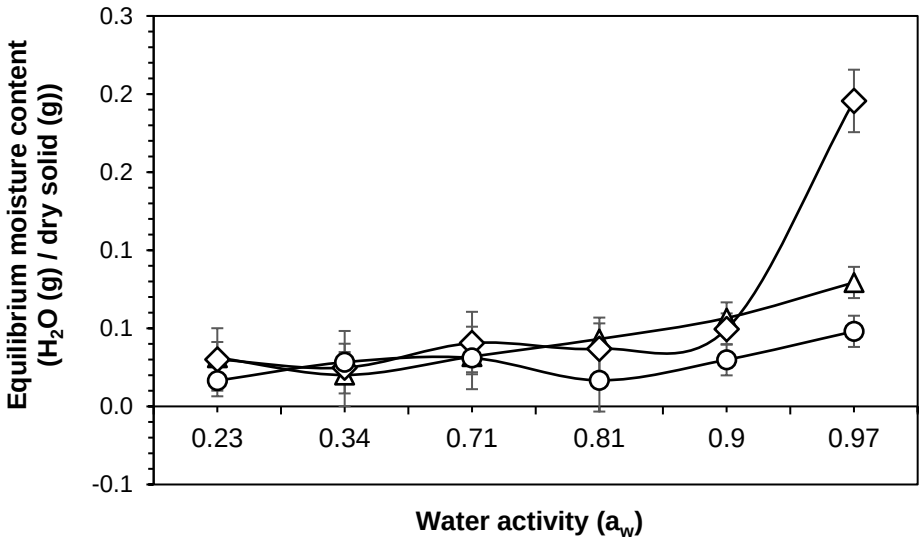

Fig. 6. Water adsorption isotherms of coconut (-○-), rubberwood (-Δ-), and palm (-◊-) at 25 °C

The Effect of Essential Oil Emulsion Containing in Natural Cellulosic Material Against Foodborne Pathogens

The materials absorbed with 50 to 700 µL LO and LC oil emulsion were tested with foodborne pathogens, as shown in Table 4. Coconut showed the best adsorption capacity to inhibit all pathogens, since it absorbed LO and LC oil emulsion at 500 and 300 µL,

respectively. Rubberwood and palm also inhibited the pathogens, but a higher amount of both emulsions at 700 µL was needed. Overall, coconut had medium properties in terms of diameter, moisture, and emulsion adsorption capacity, but low BET properties. The authors also found that 5 min in the process, coconut adsorbs water in a slow rate but adsorbs emulsion in a high rate. Therefore, the oil phased in emulsion should uptake into coconut more than water, then volatile as antibacterial function could be released into the headspace to inhibit microorganism). This property could also make the material in the coconut adsorb EO and release it as a volatile component and antibacterial agent into the air space of the package. Therefore, lower concentrations of LO and LC adsorbed in coconut are needed to inhibit bacteria.

For the purpose of food safety and packaging, this result is in agreement with other reports that EO could be applied in biomaterial-based food packaging (Matan *et al*. 2011; Suhem *et al*. 2019; Songsamoe and Matan 2020). Phothisuwan *et al*. (2020) also demonstrated that paper egg tray containing 10 to 80 µg g^{-1} clove oil could inhibit *E. coli*, *Salmonella enterica* serovar Typhimurium, and *S. aureus* on chicken eggs for 30 days. This study showed the ability of coconut, rubberwood, and palm to adsorb and release EO and inhibit the bacteria kept inside the fiber sachets in the closed box. This may help the industry in developing a biomaterial-based food packaging as a new alternative material from agricultural waste.

Table 4. Minimum Inhibition Concentration of Lime Oil (LO) and *Litsea cubeba* Oil (LC) Emulsion Compared to Control at 35 °C for 1 Month

Foodborne Pathogens	Fibers					
	Coconut		Rubberwood		Palm	
	Essential Oil Emulsion (µL)					
	LO	LC	LO	LC	LO	LC
Gram Positive						
L. monocytogenes	400	100	600	400	600	400
S. aureus	400	200	600	500	700	500
Gram Negative						
E. coli	500	300	700	500	700	500
Salmonella spp.	500	300	700	500	700	500

CONCLUSIONS

1. Coconut fiber contains 31.2% hemicellulose, which is hydrophilic and influences the fiber water absorption; 34.5% lignin, which provides the rigidity of the fiber; and 37.6% cellulose, which provides the strength of the fiber. The relatively high porosity of 3.30% and specific surface area of the coconut (1.92 m^2 g^{-1}) play a role in the fast elimination of water and emulsion absorption. In addition, the coconut fiber exhibits a good release to control bacteria growth on a NA plate using only the lowest amount of LO or LC at 300 to 500 µL.
2. Palm fiber showed high water and essential oil absorption capacity similar to that of coconut fiber. The fiber has a high percentage of porosity of 2.9% and specific surface area of 1.96 m^2 g^{-1}. However, palm fiber has a high percentage of ash of 5.63%

consisting of various minerals, which can absorb water and support the fungal growth at a high water activity (097). The palm fiber is, therefore, not suitable for fresh food applications.

3. Coconut fiber is more suitable as an absorbent than palm fiber in the development of high value-added biodegradable antimicrobial food packaging from agricultural waste.

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