Journal of Environmental Science and Sustainable Development

Volume 6
Issue 1 *July*

Article 7

7-31-2023

THE EFFECT OF DIFFERENT NATURAL WAXES TO HYDROPHOBIC PROPERTIES OF STARCH-BASED BIODEGRADABLE FOAMS

Ikbal Alexander
School of Environmental Science, Universitas Indonesia Jakarta 10430, Indonesia, mochamad.ikbal@ui.ac.id

Ahyahudin Sodri
School of Environmental Science, Universitas Indonesia Jakarta 10430, Indonesia, ahyahudin.sodri@ui.ac.id

Kosuke Mizuno
School of Environmental Science, Universitas Indonesia Jakarta 10430, Indonesia, kosuke.mizuno@ui.ac.id

Recommended Citation

Alexander, Ikbal; Sodri, Ahyahudin; and Mizuno, Kosuke (2023). THE EFFECT OF DIFFERENT NATURAL WAXES TO HYDROPHOBIC PROPERTIES OF STARCH-BASED BIODEGRADABLE FOAMS. *Journal of Environmental Science and Sustainable Development*, 6(1), 86-101.
Available at: https://doi.org/10.7454/jessd.v6i1.1199

Journal of Environmental Science and Sustainable Development
Volume 6, Issue 1, Page 86-101
ISSN: 2655-6847
Homepage: http://scholarhub.ui.ac.id/jessd

THE EFFECT OF DIFFERENT NATURAL WAXES TO HYDROPHOBIC PROPERTIES OF STARCH-BASED BIODEGRADABLE FOAMS

Ikbal Alexander[1]*, Ahyahudin Sodri[1], Kosuke Mizuno[1]
[1]School of Environmental Science, Universitas Indonesia Jakarta 10430, Indonesia

*Corresponding author: mochamad.ikbal@ui.ac.id

(Received: 23 April 2023; Accepted: 25 July 2023; Published: 31 July 2023)

Abstract

Biodegradable foam has been vastly developed to replace polystyrene foam. However, its water absorption capacity has become a significant obstacle to being used in food packaging. Therefore, this study aims to assess the effect of incorporating natural waxes as a coating material in producing biodegradable foams. The four natural waxes are soy wax, candelilla wax, beeswax, and carnauba wax. The biodegradable foams were fabricated using a thermal pressing machine from cassava starch and rice straw as natural fibre sources. The Meyer-Rod coating method was adopted to produce high contact angle and high water resistance starch-based biodegradable foams. Water absorption analysis was performed according to the Cobb60, and water solubility analysis was conducted based on International Standardization for Organization 10634:2018 procedure. The result shows that the surface modification of starch-based biodegradable foams with natural waxes significantly improved water absorbency and solubility. Moreover, it is demonstrated that carnauba wax had the highest decrease of Cobb60 index of 1.5 g/m2 and the lowest water solubility of 2% after carrying water for 90 min. This study concludes that using natural waxes as a coating material for starch-based biodegradable foam could replace conventional polystyrene foam for the food packaging industry.

Keywords: Biodegradable foam; Hydrophobic; Natural waxes; Sustainable packaging.

1. Introduction

Biopolymer-based material packaging has been dramatically investigated because it has the potential to substitute synthetic plastic foams like extruded polystyrene (Poletto et al., 2011; Raimondi et al., 2021). This thermoplastic foam is a non-decomposable material due to polystyrene's chemical structure. This synthetic polymer material can release hazardous chemicals such as styrene and benzene into the environment harming wildlife and water sources and causing environmental deterioration (Araújo et al., 2008). This environmental catastrophe has created significant concerns among the public, researchers and related industrial actors (Salam et al., 2010). Therefore, there is growing interest in developing eco-friendly and decomposable materials such as biodegradable foams to reduce plastic dependency in the food packaging industry.

Biodegradable foam is an alternative to polystyrene packaging. One of the most common ingredients that can be utilised to create biodegradable foams is polylactic acid (PLA), which is a biodegradable polymer manufactured from lactic acid (Bruscato et al., 2019). Commonly, PLA can be created from the fermentation of sugar and polysaccharides. However, PLA is expensive and hard to produce with a sophisticated synthesis (Falua et al., 2022). Natural polymers such as starch, chitosan, and fibre are one alternative for substituting fossil-based synthetic polymers. Those natural polymers are widely available, sustainable, biodegradable, and affordable.

Starch has been used to manufacture biodegradable foams because of its biodegradability and physicochemical properties. Starch can expand and easily modify physically and chemically (Ago et al., 2016). Starch-based biodegradable material has proven the versatility to create sustainable goods with various purposes, such as food packaging, home decoration, construction, furniture, and a wide range of products (Jiang et al., 2020). Another beneficial factor of the starch-based biodegradable foam is the possibility of utilising agricultural waste and lignocellulosic biomass such as rice husk and corn cob (Tapia-Blácido et al., 2022). Agricultural waste can be valorised rather than discarded, which leads to a circular economy (Spada et al., 2020). Starch-based biodegradable foam can be manufactured at an affordable price due to cheaper raw materials and offers environmental and economic benefits (Donati et al., 2022). However, starch-based biodegradable foam reportedly has poor hydrophilic performances, high brittleness, low elasticity, and strong water absorption capacity, preventing its use as food packaging (Chaireh et al., 2020; Meng et al., 2019). This research investigates new materials that can improve the hydrophobic performances of starch-based biodegradable foams.

Many researchers and entrepreneurs have addressed the limitations of starch-based biodegradable foams. Moreover, various studies have tried to enhance the hydrophobic properties of biodegradable foams by incorporating materials such as alkyl ketene dimer (AKD), polylactic acid (PLA), and chitosan (Indarti et al., 2023; Kaisangsri et al., 2012; Zhang et al., 2022). However, those materials are expensive and tend to only partially adhere to the surface of starch foams (Srisuwan & Baimark, 2021). The study of Reis et al. (2018) described that natural waxes, such as beeswax and carnauba wax, can be used as coating agents. The addition of natural wax coating significantly increases water barrier properties, and the coated product portrays a more homogeneous and solid exterior. Surface modification of starch-based biodegradable foam can be executed using natural waxes, which are locally available, inexpensive, sustainable, hydrophobic, and water resistant (Hafila et al., 2022). Additionally, natural waxes are a preservative agent for some fruits and vegetables in nature. Natural waxes are more desirable than synthetic waxes because they are produced from renewable sources such as plants and insects. Natural waxes have been used in the food industry, medicine, and skincare for decades (Wang et al., 2019). However, studies on using natural waxes to improve the hydrophobic performances of biodegradable foams are still relatively limited.

Some previous work has shown that biodegradable foams are being adopted in the packaging application material (Donati et al., 2022). Generally, biodegradable foams can be fabricated using conservative pressing methods such as injection and extrusion machines in the downstream plastic industry. The concept known as thermal pressing for biodegradables includes gelatinisation of starch, moisture evaporation, foam development, and foam drying up to a final water rate of around 2-4% (Bucio et al., 2021). This paper aims to investigate the performance of the effect of surface modification of cassava starch and rice husk-based biodegradable foams that were produced with a thermal pressing machine using four different natural waxes on their hydrophobic characteristics. Those natural waxes are beeswax, candelilla, carnauba, and soy wax. To the best of our knowledge, the present study will fill the gap of enhancing the hydrophobic properties of biodegradable foams by comparing the performance of four different natural waxes on appearance and structure, water absorbency, and water solubility.

2. Methods

2.1. Main material

Cassava starch, rice husk, polyvinyl alcohol, magnesium stearate, and distilled water were obtained from local Tangerang, Banten, Indonesia sources. The extraction of the cellulose fibres from the rice husk was based on the method developed by Pratiwi et al. (2016). The rice husk was collected from a local rice field. The rice husk was washed and cleaned with running tap water to remove impurities and dried in the sun for around five days to reduce moisture content. Hence, the dried rice husk was milled and pulverised repeatedly by a hammer mill machine into powder.

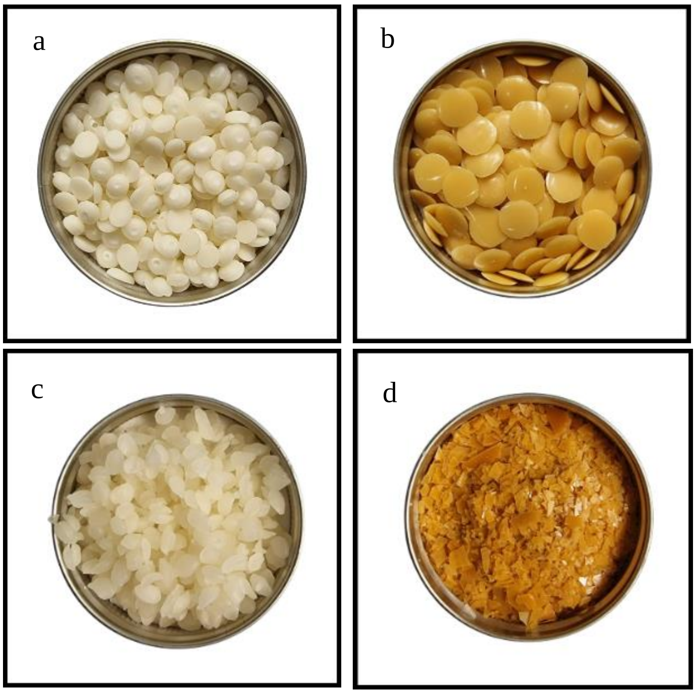

Figure. 1 (a) soy wax; (b) candelilla wax; (c) beeswax; (d) carnauba wax
(Source: Author's documentations, 2022)

Figure 1 displays the four natural waxes, which are soy wax (Figure 1a), candelilla wax (Figure 1b), beeswax (Figure 1c), and carnauba wax (Figure 1d), that were also procured locally in Tangerang, Banten, Indonesia. Soy wax (SW) and beeswax (BW) were both whitish. Meanwhile, candelilla wax (CW) and carnauba wax (PW) were yellowish and orangish, respectively. All the natural waxes came in the form of granules and tiny flakes. Table 1 shows the five biodegradable foam dough compositions. The compositions were developed based on the work of Wahyuningsih et al. (2020), who explored the production of biodegradable foam modified with oil palm empty fruit bunches pulp. Five different compositions differ in hydrophobic material. Sample code no wax (NW) has no hydrophobic material, sample code SW has 5 ml of soy wax, sample code CW has 5 ml of candelilla wax, sample code BW has 5 ml of beeswax, and sample code PW has 5 ml of carnauba wax.

DOI: https://doi.org/10.7454/jessd.v6i1.1199

Table 1. Composition of biodegradable foam dough

Sample Code	Main material					Hydrophobic material (ml)
	Cassava starch (gr)	Rice husk (gr)	PVA (gr)	Mg. stearate (gr)	Water (ml)	
NW	450	60	60	30	400	No wax
SW	450	60	60	30	400	5 ml of soy wax
CW	450	60	60	30	400	5 ml of candelilla wax
BW	450	60	60	30	400	5 ml of beeswax;
PW	450	60	60	30	400	5 ml of carnauba wax

(Source: Author's composition. 2022)

2.2. The production of starch-based foams

Strach-based biodegradable foam production was conducted using a method Iriani et al. (2015) developed with an experimental surface modification. All ingredients, such as cassava starch, powdered rice husk, polyvinyl alcohol, magnesium stearate, and distilled water, were thoroughly mixed and stirred with a mixing machine at room temperature. The batter was mixed for 10 minutes until all the ingredients were well dispersed. The 40 grams of the homogenous dough was filled in a mold and pressed using a thermal pressing machine. The square mould temperature was set at around 185°C. The pressing process was conducted for 3 minutes at a pressure of 1000 atm. After pressing, the starch foams were left overnight at ambient temperature before being coated with natural waxes.

The thermal pressing machine was that was owned by the Indonesian Agency of Agricultural Research and Development (IAARD). IAARD explained that the thermal pressing machine is an automatic heat press machine engineered to imprint the design of food packaging foam. The size of the mould can be adjusted to the final product. Each moulding is placed between platens with a certain temperature level. Water evaporation in the composites begins soon after the mould's closing, and the water vapour's venting occurs around the edge of the mould. After several minutes, the foams are removed from the press and cooled for coating.

2.3. Surface coating method

In order to hold water for food packaging purposes, the Meyer-Rod method was adopted based on the study of Nicu et al. (2013) to apply a thin layer of melted natural waxes on the top side of the starch foams. The natural waxes were weighed and placed in a breaker to be heated in a water bath at around 70°C. After the natural waxes were melted, 5 ml of melted natural waxes were filled in the surface of the foams respectively and promptly dispersed with a glass stirring rod for uniform distribution along the size of the foams.

Since the natural waxes solidified as quickly as they came into contact with the surface of the starch foams, the wax coating was not distributed uniformly. A method by Dao et al. (2018) was applied by using a hair dryer to uniformly coat the surface of the starch foam. The hair dryer was controlled at 70°C and blew high-temperature air, which melted the waxes again, making it spread evenly throughout the starch foams. After the waxing, the coated starch foams were dried for 3 hours at ambient temperature. Furthermore, the residual wax on the surface of the starch foams was scrapped off, and a clean cloth was used to clean carefully to make the surface wax layer uniformly dispersed. The amount of the natural waxes was controlled by weighing the coated starch foam pre and post-waxing with a digital balance.

2.4. Hydrophobicity analysis

The first analysis conducted was a water absorbency test. The coated starch foams were examined on a water absorption test based on the Cobb method SNI ISO 535:2016. This test is adopted to measure starch foam resistance to moisture by analysing sample mass before and after the starch foams are immersed in water. The water absorption capacity was executed using a circular flat sample measuring 7 mm in diameter. The Cobb test defined the volume of water absorbed into the sample's surface in 60 seconds, quoted in g/m2. The test was repeated two times and conducted at Cellulose Industrial Research and Development, Bandung, Indonesian Ministry of Industry. The calculation of the water absorbency was expressed in grams per square meter to the first decimal place for each test piece from Equation 1 below. The first analysis conducted was a water absorbency test.

$$\text{Water absorbency} = \frac{m_2 - m_1}{S} \qquad (1)$$

Where m_1 is the sample mass before immersion in water in grams, m_2 is the sample mass after immersion in water in grams, and S is the nominal cross-sectional area of the cylindrical tank in m^2. Another hydrophobicity analysis, the water solubility test, was carried out to support the result of the water absorbency test. The water solubility test will be implemented according to International Organization for Standardization (ISO) 10634:2018 procedure. The samples were appropriately exposed to 100 ml of water for 30 minutes, 60 minutes, and 90 minutes. The samples were weighed before and after water exposure, and the whole process was repeated two times. Equation 2 to measure the water absorption analysis is outlined below.

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

W_0 is the original sample weight in grams, and W_1 is the final weight in grams.

3. Results and Discussions

3.1. Characterisation of natural waxes

Natural waxes are a mixture of medium chain-length hydrophobic material with various properties, such as low melt viscosity, solid at room temperature, liquid at a higher temperature, insoluble in water, and melting points from around 40 to 140ºC (Habashy et al., 2020). Natural waxes are generally used as food coatings and additives, protecting fresh products, especially for the food packaging industry. Natural waxes are non-polar and dissolved in an organic solvent. Furthermore, natural waxes self-assemble quickly at ambient temperature and can develop crystalline compounds. Natural waxes are the most effective materials for decreasing water permeability due to their excellent hydrophobicity level and high content in esters of long-chain fatty alcohols, acids, and alkanes (Orhan & Eroglu, 2022).

Based on the origin, natural waxes are generally classified into mineral, animal, and plant waxes (Janesch et al., 2020). Plant and animal waxes are further categorised as renewable, while mineral waxes are nonrenewable from coal or lignite (Torun et al., 2019). The most common natural waxes for the food packaging industry available from local sources are soy wax, candelilla wax, beeswax, and carnauba wax.

Soy wax is made of soybean oil derived from soy flakes and is then hydro generated to form soy wax (Surendran et al., 2022). The main element of soy wax is triacylglyceride, consisting of stearic acid, which presents excellent liquid-repelling properties against cold and hot water. Hence, soy wax is one of the most affordable plant waxes due to the abundance of soybean production and the low manufacturing cost of soybean oil. Apart from making candles, other applications of soy wax include developing food packaging products because of their hydrophobicity, which can enhance the shelf life of food and vegetables (Forsman et al., 2020).

On the other hand, candelilla wax is a vegetable wax derived from the leaves of the candelilla shrub, Euphorbia antisyphilitica (Aguirre-Joya et al., 2019). Chemically, candelilla wax is a complicated mixture of fatty components and hydrophobic organic material that protects the stems from hot and cold temperatures and microbial and insect activity (Aranda-Ledesma et al., 2022). Due to its insolubility, candelilla wax has a promising potential for application in enhancing food packaging and compositions.

At the same time, beeswax is one of the most common animal waxes with a broad spectrum of uses from food packaging and the pharmaceutical industries due to its rich hydrophobic protective qualities. This bee product is glass-clear natural waxes generated by honey bees of *Apis cerana mellifera* and *Apis mellifera*, which are the most farmed by humans and offer more accessible access to this natural wax. Chemically, beeswax consists primarily of esters of fatty acids, hydrocarbons, and various long-chain alcohols (Pérez-Vergara et al., 2020). According to Diyana et al. (2021), beeswax has a low melting temperature of around 62-64°C and can significantly reduce water vapor permeability (WVP).

Contrastingly, carnauba wax is a plant-based natural wax derived from carnauba leaves. It is extensively utilised in the food industry due to its physicochemical properties, with a predominance of fatty esters (de Freitas et al., 2019). Carnauba wax mainly consists of a complex mixture of esters and alcohols, predominating aliphatic esters and diesters of cinnamic acid. Carnauba wax offers microencapsulation of flavours in developing edible packaging and is outstanding hydrophobic. Furthermore, carnauba wax is the hardest wax and has low solubility with the highest melting point of 82-86°C compared with other natural waxes (Devi et al., 2022).

One common technique to develop hydrophobic surfaces is a wax coating. Coating provides a significant advantage of being immensely versatile regarding scalability and materials. Almost all kinds of natural waxes can be applied using the coating technique. During a wax coating, natural waxes are impregnated on the starch foam surface, and it goes through the fibre structure. Accordingly, a silky substrate with hydrophobic qualities can be obtained, enhancing the substrate's moisture barrier qualities. On the other hand, it is also reported that using natural waxes for coating might have adverse effects such as frailty, heterogeneity, and cracks or pinholes (Gupta et al., 2021).

3.2. The effect of natural waxes on appearance and structure

The visual property of the coated starch foams is a critical quality as it directly affects consumer acceptability. A method by Buxoo & Jeetah (2020) was implemented by analysing the coated starch foams' colour, grease, texture, crack, and stable base. Analysis of the appearance and structure of coated starch foams was reviewed by visualisation. After the coating process, all samples were dried for two days before being documented and qualitatively measured. The appearance and structure of the coated foams are essential in their application as packaging material in the food industry. The results for the structure and appearance analysis of coated starch foams are provided in Table 2.

From Figure 1, it can be concluded that each natural wax has its colour and pigmentation. The colour is due to the type of trees or flowers the wax was extracted from and how much the wax was cleaned and filtered (Chen et al., 2021).

Table 2. Appearance and structure evaluation

Sample Code	Hydrophobic Material	Colour	Grease/dirt/dust	Texture	Cracks	Stability of base
SW	Soy wax	Normal	Absent	Smooth	No cracks	Stable
CW	Candelilla wax	Yellowish	Absent	Rough	Cracked up	Stable
BW	Beeswax	Orangish	Absent	Smooth	No cracks	Stable
PW	Carnauba wax	Whitish	Absent	Rough	Cracked up	Stable

(Source: Author's analysis, 2022)

Figure 2 outlines the effect of incorporating natural waxes as a coating agent on the appearance of the foams. The starch foam coated with soy wax appeared normal and uniform, with no discolouration or stains (Figure 2a). Adding soy wax showed no change in colour and effectively maintained the original colour of the product. However, other natural waxes changed the colour of the starch foams.

According to Figure 2b, applying a beeswax coating gave a smooth whitish colour to the surface of the starch foam. This result contradicts previous research conducted by Nasrin et al. (2020), who explained that applying beeswax coating changed a product's surface colour, turning it yellowish. The difference in colour change might happen due to the cleaning and extraction process of the beeswax.

Candelilla wax gave yellowish colour, and carnauba wax gave orangish colour, respectively, as portrayed in Figure 2a and Figure 2b. A similar circumstance was investigated by Zhang et al. (2018), who investigated that using natural waxes such as carnauba wax changed the colour of surface products. The authors of that report stated that carnauba wax gave greater yellowness to the product than other natural waxes. The yellowness is due to the original colour of carnauba leaves which are yellow and grey. Generally, the natural wax addition increased opacity and decreased the clarity of the coated foams.

This study also discovered no grease, dust, or dirt on any coated foams, as shown in Table 2. This finding happened because there were no contaminations, including dirt, dust, and sand. All the natural waxes were procured and proceeded in hygiene and sterilised conditions. The edges of the coated foams were trimmed to be neat, smooth, and clean. In terms of the stability of the base, the study revealed that all wax coating layers showed stability properties. A similar finding was also declared by Woch et al. (2022), who discovered that applying natural waxes for coating improved the physical stability of the product. Furthermore, the authors also mentioned that the superhydrophobic surface could be kept for at least six months under normal conditions.

Regarding the texture, it was analysed that the starch foams coated with soy wax and beeswax had smooth substrates with no cracks, as described in Figure 2. This result happened due to their viscoelastic, as they were more flexible and deformed after the liquid-solid transition. Both beeswax and soy wax contain a small amount of unsaturated hydrocarbons with low melting points, such as oleic acid, which is responsible for their flexibility (Omar-Aziz et al., 2021). However, starch foams coated with candelilla wax and carnauba wax had rough substrates and significant cracks on the surface, as portrayed in Figure 2. This result is because both waxes are more fragile and tend to break apart during solidifying. From this study, it was revealed that there were slight differences in the cracks. Candelilla wax caused prominent fractures concentrated around the edge of the starch foams.

In comparison, carnauba wax created microcracks throughout the surface. This result showed that carnauba wax might appear in a brittle material that did not resist deformation after shrinkage. The greater viscoelasticity of wax makes it more flexible and less susceptible to cracking after the liquid-solid transition (Romani et al., 2020).

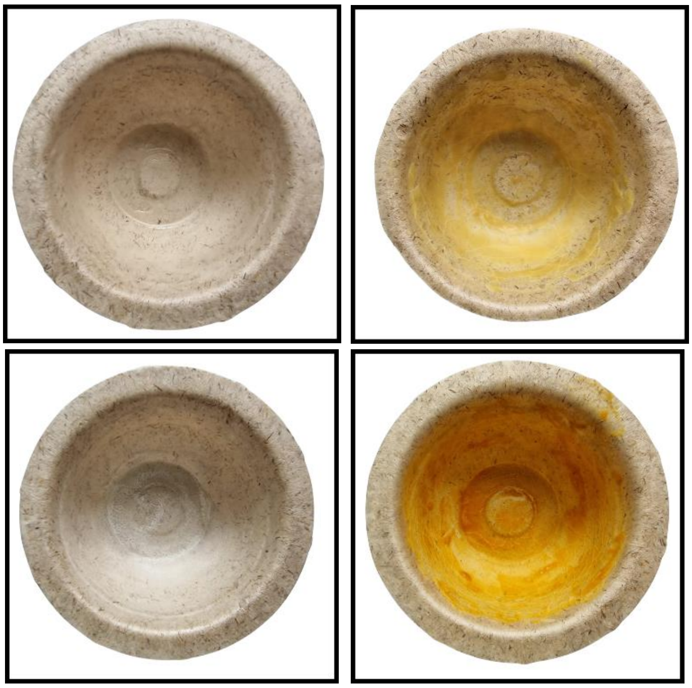

Figure. 2 (a) starch foams that were coated with soy wax; (b) starch foams that were coated with candelilla wax; (c) starch foams that were coated with beeswax; (d) starch foams that were coated with carnauba wax (Source: Author's documentations, 2022)

Moreover, there was no significant problem concerning the initial visual appearance and texture of the starch foams coated with soy wax and beeswax, which is consistent with the findings from Yoo et al. (2019). Contradictorily, candelilla and carnauba wax caused major disadvantages in colour, texture, and cracks. This result was in line with Atta et al. (2022), who stated that natural waxes such as candelilla and carnauba wax gave yellowish colour due to their natural pigments. Furthermore, the authors reported that the starch foams coated with these vegetable waxes were coloured with an increase in the concentration of the waxes, which had a significant effect on the total colour difference. However, this colour change was not apparent in the case of beeswax. Starch foams that were coated with beeswax exhibited no discolouration.

3.3. The effect of natural waxes on water absorbency capacity

Water absorption defines the amounts of permeating molecules retained or dissolved in the foams at equilibrium conditions. Starch-based foams were recorded to be only suitable for packaging dried food products since the hydrophilic qualities of foams cause them to break down quickly in the presence of water. The quality is a significant obstacle to applying these biodegradable foams in the packaging of water-rich food such as fruit and vegetables. Starch foams with low water absorbency rates are desirable to prevent water transfer between perishable goods and their surrounding environment. Many articles outline the performance of starch foams (Chaireh et al., 2020; Wardani & Hendrawati, 2021).

Natural waxes have a dense structure that prevents water absorption because of their strong hydrophobicity from intense levels of long-chain fatty alcohols and alkanes. Table 3 shows that the water absorption capacity (Cobb60 index) decreases significantly for all the coated starch foams. The biodegradable foam without coating had the worst water absorbency of 44,6 g/m^2. This result happened because the hydroxyl materials on the starch foam contain many water-binding sites that allow water particles to go through the starch foam structure. The starch foams with coating had significantly lower levels of water absorbency because the natural waxes prevented water penetration by filling surface pores. The starch foam coated with carnauba wax showed the smallest water absorbency of 1,5 g/m^{2,} as described in Table 3. Devi et al. (2022) also identified the same result, who wrote that incorporation of carnauba wax enhanced mechanical qualities and showed remarkable water, gas, and light barrier properties more than other natural waxes.

Table 3. Water absorbency of starch foams with varied natural waxes

Sample Code	Hydrophobic Material	Water Absorbency in g/m^2 (Cobb$_{60}$)
NW	No wax	44,6
SW	Beeswax	2,8
CW	Candelilla wax	4,9
BW	Soy wax	10,0
PW	Carnauba wax	1,5

Meanwhile, starch foam coated with beeswax had a water absorbency level of 2,8 g/m^2, followed by candelilla wax of 4,9 g/m^2, which had a slightly higher absorbency rate than beeswax. This finding aligns with the study of Bucio et al. (2021), who analysed the performance of beeswax and candelilla wax on their water absorption level. Moreover, the author declared that beeswax coating had a lower absorbency rate than candelilla wax coating due to the significantly greater concentration of fatty acids, fatty alcohols, and esters in the beeswax coating. On the other hand, soy wax had the lowest water absorbency of 10 g/m^{2,} as found in Table 3. This finding was similar to research carried out by (Shen et al., 2020), who stated that applying soy wax as a coating had slightly lower hydrophobic properties than beeswax and candelilla wax. However, soy wax has several advantages, of being more affordable and readily available.

A previous study reported that adding natural waxes, such as beeswax, decreased the water absorbency level of starch foams (Chaireh et al., 2020). This finding was attributed to the hydrophobic qualities of the natural waxes that preserved the starch foams from touching base with water. In brief, carnauba wax reduced the water absorbency rate more than beeswax, soy wax, and candelilla wax. Hence, carnauba wax can enhance food stability better than other natural waxes.

3.4. The effect of natural waxes on water solubility properties

Starch-based biodegradable foams with low water solubility retard perishable goods from humidity and moisture loss. However, such foams commonly have strong water solubility due to their hydrophobicity. Starch foams with low water solubility are desirable to prevent the functional and active components from decomposing. Many studies have reported that adding natural waxes in starch foams helps maintain foam integrity when exposed to water. Water solubility analysis defined the starch foam resistance to moisture by analysing sample mass changes before and after being filled with 100ml of water.

Table 4 shows that the longer the exposure time, the level of water solubility increases. It is clearly seen from the table that all coated foams had lower water absorption rates compared with the uncoated foam. Research from Celik et al. (2021) also highlighted that using natural waxes such as carnauba wax in starch-based foams led to low water solubility. As can be seen in Table 4, it outlined that the uncoated foam absorbed significantly more water than the coated foams and had the highest water solubility since the hydroxyl materials in the starch chain could react readily with water particles. The uncoated foam was heavier due to moisture permeation into the biodegradable foam, gaining 34.7% more weight in 30 minutes, 46% more in 60 minutes, and 57% more in 90 minutes, as portrayed in Table 4.

Table 4. Water solubility analysis of starch foams

Sample Code	Hydrophobic material	Duration of water exposure		
		30 min	60 min	90 min
NW	No wax	34.9%	46%	57%
SW	Beeswax	1.5%	2.2%	2.2%
CW	Candelilla wax	3%	3.3%	3.7%
BW	Soy wax	4%	4.2%	4.5%
PW	Carnauba wax	1.4%	1.7%	2%

On the other hand, the coated foams were still well-shaped even after 90 minutes of carrying water. The natural waxes remained as solidified lumps on the starch foam surface. This result is because the starch foams were already covered with melted waxes that blocked all pores of the starch foams. The weight increase for all coated foams was less than 5%, with carnauba wax coating showing the best result of a 1.4% increase after 30 minutes, 1.7% after 60 minutes, and 2% after 90 minutes holding up water, as outlined in Table 4. According to de Castro e Silva et al. (2020), carnauba wax mainly consists of aliphatic esters and diesters of cinnamic acid, making it almost lower in solubility. They also analysed that carnauba wax formed a higher resistance surface than other natural waxes. Another previous study by Woch et al. (2022) also declared that incorporating carnauba wax in starch foams significantly decreased the coated foams' water solubility and water absorbency.

Other natural waxes also showed remarkable improvement in repelling water. As shown in Table 4, beeswax had a 1.5% increase after 30 minutes, 2.2% after 60 minutes, and 2.6% after 90 minutes of water exposure. This discovery was in line with the study of York et al. (2019), who found that beeswax had a slightly higher solubility than carnauba wax due to its different chemical structures; hydrocarbon and a carboxylic acid group.

Subsequently, candelilla wax had a 3% increase after 30, 3.3% after 60 minutes, and 3.7% after 90 minutes of carrying water. At the same time, soy wax had the highest water solubility, with an increase of 4% after 30 minutes, 4.2% after 60 minutes, and 4.5% after 90 minutes coming into contact with water, as portrayed in Table 4. These significant improvements between uncoated and coated foams exhibited that natural waxes decreased the water solubility of the starch foams and retained the structural integrity of the starch foams. Miranda et al. (2022) outlined a similar finding and discovered that the dimensional stability of coated foams was upgraded as a function of their hydrophobicity.

Using natural waxes presents several benefits. Natural waxes are sustainable and regarded as food-grade material since they are used as additives in the food and pharmaceutical industries (Soleimanian et al., 2020). They are also rich in lipids, which can increase water repellency in a coating due to their strong hydrophobicity (Galus et al., 2020). The different hydrophobic characteristics of natural waxes are owed to their different chemical structure and dissolution-precipitation quality in a solvent. This result concluded that carnauba wax was the most effective in reducing water solubility compared with other natural waxes, which correlates with the water absorbency rate as described in Table 3, which was lowest in carnauba wax coating foam. This was probably because the carnauba wax increased the hydrophobic properties to a significant extent than other natural waxes. Thus, adding carnauba wax into starch foams will be expected to improve the stability of perishable goods more than other natural waxes.

4. Conclusion

This study demonstrated the effects of natural wax coating on water absorption capacity, appearance and texture. The modification on the surface of starch-based biodegradable foams with four different natural waxes, soy wax, candelilla wax, beeswax, and carnauba wax, has improved water absorbency and water solubility rates, which is a crucial element in storing and preserving perishable goods. The production of starch-based biodegradable foams coated with 5 ml of melted carnauba wax had the lowest water absorbency and water solubility. Moreover, the carnauba wax coating significantly enhanced the hydrophobicity of the starch-based foam composite to offer a product with low water solubility.

Based on the appearance and structure test, beeswax and soy wax had outstanding appearance and structure tests with no grease, dust, or dirt on the coated foams. Both beeswax and soy wax had smooth substrates without any cracks. On the other hand, there were significant issues concerning the visual appearance and texture of the starch foams coated with candelilla wax and carnauba wax. Both natural waxes caused significant cracks and changes in colour. The obtained experimental data can provide prospects for using natural waxes as natural coatings in starch-based biodegradable foams for sustainable protection and packaging of food and post-harvest vegetables and fruits to replace conventional polystyrene foam packaging.

The limitations encountered in this study are the mechanical properties of coated starch foam, such as mechanical strength, tensile strength, and biodegradability. Moreover, future researchers are encouraged to use other similar data resources to analyse the mechanical properties of coated starch foams and conduct other research related to starch-based biodegradable foams to enhance their service performances for further comparisons.

Acknowledgement

The authors would like to thank significantly to the School of Environmental Science, University of Indonesia, for all the guidance and feedback. We sincerely thank the Indonesian Agency of Agricultural Research and Development (IAARD), Ministry of Agriculture of the Republic of Indonesia, for providing all the necessary equipment and tools to fabricate biodegradable foams. We are also thankful to all the Cellulose Industrial Research and Development, Indonesian Ministry of Industry staff, for helping and analysing the water solubility test. We also thank Herdis Herdiansyah of the JESSD journal for his continuous support.

Author Contribution

Conceptualisation, I.A., A.S. and K.M.; Methodology, I.A., A.S. and K.M.; Validation, I.A.; Formal Analysis, I.A.; Investigation, I.A.; Resources, I.A.; Data Curation, I.A.; Writing – Original Draft Preparation, I.A.; Writing – Review & Editing, I.A., A.S., K.M.; Visualization, I.A.; Supervision, A.S. and K.M.; Project Administration, I.A.

References

Ago, M., Ferrer, A., & Rojas, O. J. (2016). Starch-based biofoams reinforced with lignocellulose nanofibrils from residual palm empty fruit bunches: Water sorption and mechanical strength. *ACS Sustainable Chemistry & Engineering*, *4*(10), 5546-5552. https://doi.org/10.1021/acssuschemeng.6b01279

Aguirre-Joya, J. A., Cerqueira, M. A., Ventura-Sobrevilla, J., Aguilar-Gonzalez, M. A., Carbó-Argibay, E., Castro, L. P., & Aguilar, C. N. (2019). Candelilla wax-based coatings and films: Functional and physicochemical characterisation. *Food and Bioprocess Technology*, *12*, 1787-1797. https://doi.org/10.1007/s11947-019-02339-2

Aranda-Ledesma, N. E., Bautista-Hernández, I., Rojas, R., Aguilar-Zárate, P., del Pilar Medina-Herrera, N., Castro-López, C., & Martínez-Ávila, G. C. G. (2022). Candelilla wax: Prospective suitable applications within the food field. *LWT*, *159*, 113170. https://doi.org/10.1016/j.lwt.2022.113170

Araújo, J. R., Waldman, W. R., & De Paoli, M. A. (2008). Thermal properties of high density polyethylene composites with natural fibres: Coupling agent effect. *Polymer Degradation and Stability*, *93*(10), 1770-1775. https://doi.org/10.1016/j.polymdegradstab.2008.07.021

Atta, O. M., Manan, S., Shahzad, A., Ul-Islam, M., Ullah, M. W., & Yang, G. (2022). Biobased materials for active food packaging: a review. *Food Hydrocolloids*, *125*, 107419. https://doi.org/10.1016/j.foodhyd.2021.107419

Bruscato, C., Malvessi, E., Brandalise, R. N., & Camassola, M. (2019). High performance of macrofungi in the production of mycelium-based biofoams using sawdust—Sustainable technology for waste reduction. *Journal of Cleaner Production*, *234*, 225-232. https://doi.org/10.1016/j.jclepro.2019.06.150

Bucio, A., Moreno-Tovar, R., Bucio, L., Espinosa-Dávila, J., & Anguebes-Franceschi, F. (2021). Characterisation of beeswax, candelilla wax and paraffin wax for coating cheeses. *Coatings*, *11*(3), 261. https://doi.org/10.3390/coatings11030261

Buxoo, S., & Jeetah, P. (2020). Feasibility of producing biodegradable disposable paper cup from pineapple peels, orange peels and mauritian hemp leaves with beeswax coating. *SN Applied Sciences*, *2*, 1-15. https://doi.org/10.1007/s42452-020-3164-7

Celik, N., Kiremitler, N. B., Ruzi, M., & Onses, M. S. (2021). Waxing the soot: Practical fabrication of all-organic superhydrophobic coatings from candle soot and carnauba wax. *Progress in Organic Coatings*, *153*, 106169. https://doi.org/10.1016/j.porgcoat.2021.106169

Chaireh, S., Ngasatool, P., & Kaewtatip, K. (2020). Novel composite foam made from starch and water hyacinth with beeswax coating for food packaging applications. *International Journal of Biological Macromolecules*, *165*, 1382-1391. https://doi.org/10.1016/j.ijbiomac.2020.10.007

Chen, Y., Liu, W., Xu, C., Liu, Y., He, J., Tian, D., Long, L., Yang, G., Zhang, X., & Zhang, Y. (2021). Synthesis of a novel superamphiphobic coating with a hierarchical three-dimensional structure inspired by bird's nest. *Applied Clay Science*, *204*, 106031. https://doi.org/10.1016/j.clay.2021.106031

Dao, T. G., Nguyen, T. T., Song, X., Wang, X., & Song, K. (2018). The effect of pretreatment method on the decorative effect of the wax furniture. *IOP Conference Series: Materials Science and Engineering*, *452*(2), 22009. https://doi.org/10.1088/1757-899X/452/2/022009 de Castro e Silva, P., de Oliveira, A. C. S., Pereira, L. A. S., Valquíria, M., Carvalho, G. R., Miranda, K. W. E., Marconcini, J. M., & Oliveira, J. E. (2020). Development of bionanocomposites of pectin and nanoemulsions of carnauba wax and neem oil pectin/carnauba wax/neem oil composites. *Polymer Composites*, *41*(3), 858-870. https://doi.org/10.1002/pc.25416

de Freitas, C. A. S., de Sousa, P. H. M., Soares, D. J., da Silva, J. Y. G., Benjamin, S. R., & Guedes, M. I. F. (2019). Carnauba wax uses in food–A review. *Food Chemistry*, *291*, 38–48. https://doi.org/10.1016/j.foodchem.2019.03.133

Devi, L. S., Kalita, S., Mukherjee, A., & Kumar, S. (2022). Carnauba wax-based composite films and coatings: recent advancement in prolonging post-harvest shelf-life of fruits and vegetables. *Trends in Food Science & Technology*, 129 (November 2022), 296-305 https://doi.org/10.1016/j.tifs.2022.09.019

Diyana, Z. N., Jumaidin, R., Selamat, M. Z., & Suan, M. S. M. (2021). Thermoplastic starch/beeswax blend: characterisation on thermal mechanical and moisture absorption properties. *International Journal of Biological Macromolecules*, *190*, 224-232. https://doi.org/10.1016/j.ijbiomac.2021.08.201

Donati, N., Spada, J. C., & Tessaro, I. C. (2022). Recycling rice husk ash as a filler on biodegradable cassava starch-based foams. *Polymer Bulletin*, 1-18. https://doi.org/10.1007/s00289-022-04557-9

Falua, K. J., Pokharel, A., Babaei-Ghazvini, A., Ai, Y., & Acharya, B. (2022). Valorisation of starch to biobased materials: a review. *Polymers*, *14*(11), 2215. https://doi.org/10.3390/polym14112215

Forsman, N., Johansson, L.-S., Koivula, H., Tuure, M., Kääriäinen, P., & Österberg, M. (2020). Open coating with natural wax particles enables scalable, non-toxic hydrophobation of cellulose-based textiles. *Carbohydrate Polymers*, *227*, 115363. https://doi.org/10.1016/j.carbpol.2019.115363

Galus, S., Gaouditz, M., Kowalska, H., & Debeaufort, F. (2020). Effects of candelilla and carnauba wax incorporation on the functional properties of edible sodium caseinate films. *International Journal of Molecular Sciences*, *21*(24), 9349. https://doi.org/10.3390/ijms21249349

Gupta, S., Ivvala, J., & Grewal, H. S. (2021). Development of natural wax based durable superhydrophobic coatings. *Industrial Crops and Products*, *171*, 113871. https://doi.org/10.1016/j.indcrop.2021.113871

Habashy, R., Khoder, M., Zhang, S., Pereira, B., Bohus, M., Wang, J. T.-W., Isreb, A., & Alhnan, M. A. (2020). An innovative wax-based enteric coating for pharmaceutical and nutraceutical oral products. *International Journal of Pharmaceutics*, *591*, 119935. https://doi.org/10.1016/j.ijpharm.2020.119935

Hafila, K. Z., Jumaidin, R., Ilyas, R. A., Selamat, M. Z., & Yusof, F. A. M. (2022). Effect of palm wax on the mechanical, thermal, and moisture absorption properties of thermoplastic cassava starch composites. *International Journal of Biological Macromolecules*, *194*, 851–860. https://doi.org/10.1016/j.ijbiomac.2021.11.139

Indarti, E., Muliani, S., & Yunita, D. (2023). Characteristics of biofoam cups made from sugarcane bagasse with rhizopus oligosporus as binding agent. *Advances in Polymer Technology*, *2023*. https://doi.org/10.1155/2023/8257317

Iriani, E. S., Irawadi, T. T., Sunarti, T. C., Richana, N., & Yuliasih, I. (2015). Effect of corn hominy and polyvinyl alcohol on mechanical properties of cassava starch-baked foam. *Polymer-Plastics Technology and Engineering*, *54*(3), 282-289. https://doi.org/10.1080/03602559.2014.977423

Janesch, J., Arminger, B., Gindl-Altmutter, W., & Hansmann, C. (2020). Superhydrophobic coatings on wood made of plant oil and natural wax. *Progress in Organic Coatings*, *148*, 105891. https://doi.org/10.1016/j.porgcoat.2020.105891

Jiang, T., Duan, Q., Zhu, J., Liu, H., & Yu, L. (2020). Starch-based biodegradable materials: Challenges and opportunities. *Advanced Industrial and Engineering Polymer Research*, *3*(1), 8-18. https://doi.org/10.1016/j.aiepr.2019.11.003

Kaisangsri, N., Kerdchoechuen, O., & Laohakunjit, N. (2012). Biodegradable foam tray from cassava starch blended with natural fiber and chitosan. *Industrial Crops and Products*, *37*(1), 542–546. https://doi.org/10.1016/j.indcrop.2011.07.034

Meng, L., Liu, H., Yu, L., Duan, Q., Chen, L., Liu, F., Shao, Z., Shi, K., & Lin, X. (2019). How water acting as both blowing agent and plasticiser affect on starch-based foam. *Industrial Crops and Products*, *134*, 43-49. https://doi.org/10.1016/j.indcrop.2019.03.056

Miranda, M., Sun, X., Marín, A., Dos Santos, L. C., Plotto, A., Bai, J., Assis, O. B. G., Ferreira, M. D., & Baldwin, E. (2022). Nano-and micro-sized carnauba wax emulsions-based coatings incorporated with ginger essential oil and hydroxypropyl methylcellulose on papaya: Preservation of quality and delay of post-harvest fruit decay. *Food Chemistry: X*, *13*, 100249. https://doi.org/10.1016/j.fochx.2022.100249

Nasrin, T. A. A., Rahman, M. A., Arfin, M. S., Islam, M. N., & Ullah, M. A. (2020). Effect of novel coconut oil and beeswax edible coating on post-harvest quality of lemon at ambient storage. *Journal of Agriculture and Food Research*, *2*, 100019. https://doi.org/10.1016/j.jafr.2019.100019

Nicu, R., Lupei, M., Balan, T., & Bobu, E. (2013). Alkyl-chitosan as paper coating material to improve water barrier properties. *Cellulose Chemistry and Technology*, *47*(7–8), 623-630.

Omar-Aziz, M., Khodaiyan, F., Yarmand, M. S., Mousavi, M., Gharaghani, M., Kennedy, J. F., & Hosseini, S. S. (2021). Combined effects of octenylsuccination and beeswax on pullulan films: Water-resistant and mechanical properties. *Carbohydrate Polymers*, *255*, 117471. https://doi.org/10.1016/j.carbpol.2020.117471

Orhan, N. O., & Eroglu, Z. (2022). Structural characterisation and oxidative stability of black cumin oil oleogels prepared with natural waxes. *Journal of Food Processing and Preservation*, e17211. https://doi.org/10.1111/jfpp.17211

Pérez-Vergara, L. D., Cifuentes, M. T., Franco, A. P., Pérez-Cervera, C. E., & Andrade-Pizarro, R. D. (2020). Development and characterisation of edible films based on native cassava starch, beeswax, and propolis. *NFS Journal*, *21*, 39-49. https://doi.org/10.1016/j.nfs.2020.09.002

Poletto, M., Dettenborn, J., Zeni, M., & Zattera, A. J. (2011). Characterisation of composites based on expanded polystyrene wastes and wood flour. *Waste Management*, *31*(4), 779-784. https://doi.org/10.1016/j.wasman.2010.10.027

Pratiwi, R., Rahayu, D., & Barliana, M. I. (2016). Pemanfaatan selulosa dari limbah jerami padi (Oryza sativa) sebagai bahan bioplastik. *Indonesian Journal of Pharmaceutical Science and Technology*, *3*(3), 83-91. https://doi.org/10.15416/ijpst.v3i3.9406

Raimondi, G., Maucieri, C., Toffanin, A., Renella, G., & Borin, M. (2021). Smart fertilisers: What should we mean and where should we go? *Italian Journal of Agronomy*, *16*(2). https://doi.org/10.4081/ija.2021.1794

Reis, M. O., Olivato, J. B., Bilck, A. P., Zanela, J., Grossmann, M. V. E., & Yamashita, F. (2018). Biodegradable trays of thermoplastic starch/poly (lactic acid) coated with beeswax. *Industrial Crops and Products*, *112*, 481-487. https://doi.org/10.1016/j.indcrop.2017.12.045

Romani, V. P., Olsen, B., Collares, M. P., Oliveira, J. R. M., Prentice, C., & Martins, V. G. (2020). Cold plasma and carnauba wax as strategies to produce improved bi-layer films for sustainable food packaging. *Food Hydrocolloids*, *108*, 106087. https://doi.org/10.1016/j.foodhyd.2020.106087

Salam, A., Pawlak, J. J., Venditti, R. A., & El-tahlawy, K. (2010). Synthesis and characterisation of starch citrate−chitosan foam with superior water and saline absorbance properties. *Biomacromolecules*, *11*(6), 1453-1459. https://doi.org/10.1021/bm1000235

Shen, T., Fan, S., Li, Y., Xu, G., & Fan, W. (2020). Preparation of edible non-wettable coating with soybean wax for repelling liquid foods with little residue. *Materials*, *13*(15), 3308. https://doi.org/10.3390/ma13153308

Soleimanian, Y., Goli, S. A. H., Shirvani, A., Elmizadeh, A., & Marangoni, A. G. (2020). Wax‐based delivery systems: Preparation, characterisation, and food applications. *Comprehensive Reviews in Food Science and Food Safety*, *19*(6), 2994-3030. https://doi.org/10.1111/1541-4337.12614

Spada, J. C., Jasper, A., & Tessaro, I. C. (2020). Biodegradable cassava starch based foams using rice husk waste as macro filler. *Waste and Biomass Valorization*, *11*, 4315-4325. https://doi.org/10.1007/s12649-019-00776-w

Srisuwan, Y., & Baimark, Y. (2021). Improvement of water resistance of thermoplastic starch foams by dip-coating with biodegradable polylactide-b-polyethylene glycol-b-polylactide copolymer and its blend with poly (D-lactide). *Progress in Organic Coatings*, *151*, 106074. https://doi.org/10.1016/j.porgcoat.2020.106074

Surendran, A. N., Ajjarapu, K. P. K., Arumugham, A. A., Kate, K., & Satyavolu, J. (2022). Characterisation of industry grade soybean wax for potential applications in natural fiber reinforced composite (NFRC) filaments. *Industrial Crops and Products*, *186*, 115163. https://doi.org/10.1016/j.indcrop.2022.115163

Tapia-Blácido, D. R., Aguilar, G. J., de Andrade, M. T., Rodrigues-Júnior, M. F., & Guareschi-Martins, F. C. (2022). Trends and challenges of starch-based foams for use as food packaging and food container. *Trends in Food Science & Technology*, *119*, 257-271. https://doi.org/10.1016/j.tifs.2021.12.005

Torun, I., Ruzi, M., Er, F., & Onses, M. S. (2019). Superhydrophobic coatings made from biocompatible polydimethylsiloxane and natural wax. *Progress in Organic Coatings*, *136*, 105279. https://doi.org/10.1016/j.porgcoat.2019.105279

Wahyuningsih, K., Iriani, E. S., & Yuanita, E. (2020). Characterisation of migration rate and biodegradability of cassava starch‐based biofoam modified with alkyl ketene dimer. *Macromolecular Symposia*, *391*(1), 1900131. https://doi.org/10.1002/masy.201900131

Wang, F., Lei, S., Ou, J., Li, C., & Li, W. (2019). Novel all-natural material for oil/water separation. *Industrial & Engineering Chemistry Research*, *58*(5), 1924-1931. https://doi.org/10.1021/acs.iecr.8b05535

Wardani, L. K., & Hendrawati, N. (2021). Kajian literatur karakteristik biodegradable polymer berbahan baku pati dengan penambahan filler dan beeswax. *distilat: jurnal teknologi separasi*, *7*(2), 333–340. https://doi.org/10.33795/distilat.v7i2.246

Woch, J., Małachowska, E., Korasiak, K., Lipkiewicz, A., Dubowik, M., Chrobak, J., Iłowska, J., & Przybysz, P. (2022). barrier coatings containing natural and paraffin waxes. *Molecules 27*(3) https://doi.org/10.3390/molecules27030930

Yoo, J., Chang, S. J., Wi, S., & Kim, S. (2019). Spent coffee grounds as supporting materials to produce bio-composite PCM with natural waxes. *Chemosphere*, *235*, 626-635. https://doi.org/10.1016/j.chemosphere.2019.06.195

York, D. W., Collins, S., & Rantape, M. (2019). Measuring the permeability of thin solid layers of natural waxes. *Journal of Colloid and Interface Science*, *551*, 270-282. https://doi.org/10.1016/j.jcis.2019.03.104

Zhang, Yi, Simpson, B. K., & Dumont, M.-J. (2018). Effect of beeswax and carnauba wax addition on properties of gelatin films: a comparative study. *Food Bioscience*, *26*, 88-95. https://doi.org/10.1016/j.fbio.2018.09.011

Zhang, Y., Liao, J., Li, J., Guo, S., Mo, L., Liu, Z., & Xiong, Q. (2022). Synthesis of a robust, water-stable, and biodegradable pulp foam by poly-lactic acid coating towards a zero-plastic earth. *Environmental Pollution*, *306*, 119450. https://doi.org/10.1016/j.envpol.2022.119450