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Processing and Characterization of Bioplastic Film Fabricated from a hybrid of Cocoa Pod Husk and Kenaf for the Application in Food Industries

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Abstract: The increase in synthetic plastic packaging usage has led to severe environmental problems due to their non-biodegradability. Replacement of synthetic with eco-friendly packaging films could reduce the ecological crisis and increase the potential of bio-material wastes to be utilized. Thus, this study focuses on developing a cost-effective film to be potentially exploited as a novel food packaging material using a combination of agricultural wastes, cocoa pod husks and kenaf fibre. Five different ratios of cocoa pod husk to kenaf fibre (100:0, 75:25, 50:50, 25:75, and 0:100) were used in the bioplastic preparation. The evaluated bioplastic reinforcement properties, including drying time to be bioplastic, water content, water absorption and water vapour permeability, were evaluated. This study found that the bioplastic with 50% cocoa pod husks integrated with 50% kenaf fibres exhibited the lowest water absorption and water vapour permeability. The moisture content of the 50% ratio of both materials presented an acceptable result (6.92%), while it took five days as the time taken for the bioplastic to be dried. The tested properties proved that the bioplastic from the ratio of 50% of cocoa pod husks and 50% of kenaf fibres is suitable as the novel of food packaging materials. The improvement of bioplastic made of cocoa pod husks incorporated with kenaf fibres could be exploited as a source for bio-plastic production in a food container and food wrapping.

INTRODUCTION

The plastic manufacturer reported that the usage of the total plastics over the year have been increasing. The enormous use of petroleum-based plastic compounds emphasized is an essential need for a sustainable alternative of renewable resources (Jain and Tiwari, 2015). There are various disadvantages present for petroleum-based plastic. This plastic type has been stated as a product that does not readily break down and is non-biodegradable as it is stable and hydrophobic. Apart from that, the main disadvantage of this petroleum-derived plastic is it does not quickly decompose and biodegrade, which lead to the adverse effect of littering on the planet (Vaverková et al., 2012). Hence, an alternative method must be identified to save our environment, such as producing bioplastic to sustain greener earth (Shah et al., 2020).

Bioplastic is widely known as biodegradable plastics that can be decomposed within a shorter time (Shen et al., 2020). This bioplastic is generally made up of renewable resources such as starch, cellulose, and fibre. Besides, bioplastic has mainly used the crops as the vital ingredients rather than crude oil production like petroleum as its primary resources due to its degradability processes (Song et al., 2009). Kenaf fibre (automotive industries) and the cocoa pod husks (chocolate industries) are known as wastes that can be innovated and invented into a new material such as bioplastic that is "green" to nature.

Cocoa, Theobroma cacao L. is vitally crucial in the agricultural and economical crop that donating an enormous value to the economics of Malaysia (Adi-Dako et al., 2016). It is the primary source of cocoa beans in the chocolate production industry. Cocoa fruit consists of cocoa beans and cocoa pod husks (CPH), where this CPH becomes typically a waste of about 52- 76% of the cocoa fruit after extracted the cocoa bean from the fruit. After harvesting the cocoa beans, the CPH is generally abandoned and left as a waste to become rotten on a farm. It is reported that the massive amount of CPH waste is still underutilized and unexploited.

Kenaf originates from the Hibiscus cannabinus, a fast-growing plant of the hibiscus plant, and can be found in Malaysia as it is a tropical plant. This plant is mallow, a family of Malvaceae and composts of fibre from the bast fibre group. Kenaf fibres show excellent tensile strength and dense network fibre (Wambua et al., 2003). Kenaf is a polymer composite that is flexible and nature friendly. Besides, kenaf fibre is a secure and robust plant. The fibrous stalk possesses remarkable mechanical properties that replace glass fibres in polymer composites as reinforcing elements. The natural fibres source from kenaf can be initiated as glass replacement in fibre reinforced plastics. The mechanical properties of this natural fibre are lower compared to glass fibres (Wambua et al., 2003). Malaysian automotive industries use kenaf fibre to produce composite materials competitive with synthetic fibre (Mohanty et al., 2000).

Meanwhile, it is proven that kenaf natural fibre is lighter, about 50% of estimation than the glass fibres, and thus it is cheaper than the glass. Kenaf fibre has been added with the non-biodegradable polymers to produce bioplastic, which is biodegradability, lower density, higher strength and toughness, do not harmful, low cost and reduction of the use of the resources that cannot be easily renewed (Lee et al., 2009). This natural fibre is an innovative idea to replace and reinforce natural ingredients in bioplastics, especially for food product packaging, in prolonging their shelf life. The purpose of this kenaf fibre will be able to reduce the expenditure of plastic industries production. The kenaf addition will significantly impact and affect the mechanical properties of the type of initiative replacement of packaging.

Nowadays, plenty of research has been invented the idea of the potential of the combination of cellulose and fibre in bioplastic reinforcement. Specifically, the combination of natural cellulose and natural fibre in bioplastic production presents a lack of attention even though the product is beneficial and could be expended. Promising results from our previous study (Azmin et al., 2020) in formulating bioplastic films using a combination of cellulose (cocoa pod husks) and fibre (sugarcane bagasse) to be applied as food packaging contribute to this study. Thus, this research is conducted to analyze the physicochemical properties of the bioplastic reinforced with CPH and kenaf, the wastes from the chocolate and automotive industries, respectively. Cellulose from CPH and fibres from kenaf will be extracted and combined in five different ratios in bioplastic formulations before analyzing the physicochemical properties of these five bioplastics. This study found that the combination of CPH cellulose and kenaf fibre in bioplastic formation could be commercialized and safe for food packaging. Thus, the reduction of wastes from the chocolate and automotive industries can be made.

MATERIALS AND METHODS

Sample Collection and Pre-treatment

Ground and bleached kenaf sample was obtained from Kenaf and Tobacco State located at Kota Bharu Kelantan, Malaysia. In this study, the collected sample of kenaf fibre was ready to be used in bioplastic formulation. CPH sample was collected from the chocolate factory at Tanah Merah Kelantan, Malaysia. 2 kg of CPH sample was washed using running tap water to remove impurities before being cut into small sizes. CPH then was dried under sun drying until a constant weight was obtained. The dried CPH was milled into a fine powder form using a blender.

Cellulose Extraction from Cocoa Pod Husk

40 g of the CPH powder was weighed and diluted with 10% of the Sodium Hydroxide, NaOH solution in a beaker for 3 hours at 100°C. The CPH was washed in a muslin cloth several times with distilled water until pH 7 of the filtrate was obtained. These processes were conducted to separate the lignin components in the CPH, which mix with the cellulose in raw CPH. The CPH then was bleached with Hydrogen peroxide at a temperature of 60-70°C for 60 minutes to remove any impurities before rewashing it with the distilled water. Finally, the yellowish cellulose from the CPH was dried for 24 hours. Figure 1 presents the cellulose extraction procedure starting from a fine powder of CPH until yellowish cellulose was obtained.

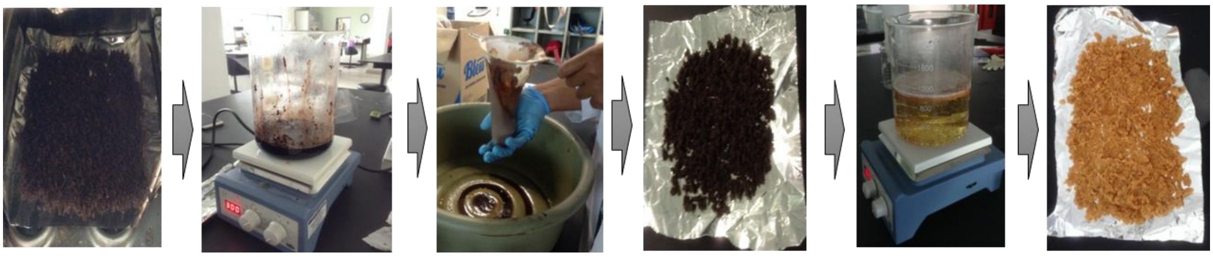


Figure 1. Cellulose extraction procedure from CPH

Bioplastic Development

In this study, five formulations (ratio of CPH to kenaf were 100:0, 75:25, 50:50 and 25:75) of bioplastics were developed where the bioplastic made from 100% CPH was set to be the control plastic. Control bioplastic was prepared using the following procedure. The mixture of 3 g of the starch, 1 g of bleached CPH powder, 1 ml of glycerine, 80 ml of distilled water and 0.5 g of sorbitol was stirred using the glass rod and heated on a hot plate at 90°C. The mixture was continuously stirred to ensure that there was no clump formed. The mixture was removed from the heat after the formation of yellowish liquid thickened occurred. The mixture was poured into a petri dish and left to dry in an oven at 50°C for 24 hours. Then, the undried bioplastic was leftover under sunshade for drying purposes. The other four bioplastics formulations were followed the controlled bioplastic procedure with 1 g of the total weight of the ratio of CPH to kenaf.

Bioplastic Characterisation and Measurement

The characterization and measurement of developed bioplastic were conducted for five parameters: sensory evaluation, drying time, water absorption, moisture content, and water vapour permeability (WVP). The sensory evaluation was tested for all five developed bioplastics where the bioplastic aroma, colour, and texture were observed.

After the bioplastic was developed, it was left under sunshade for drying purposes. Each plastic took a different time to dry. The time for bioplastics drying was recorded. The drying time measurement was done for triplicate to ensure the accuracy of the result.

The water absorption test of bioplastic was conducted by soaking the plastic into the water to measure the amount of water absorbed by the formed bioplastic. In this study, each developed plastic was soaked in 15 ml of water at room temperature. The bioplastic appearance was strictly observed where the time of bioplastic starting to break down was recorded. At this critical time, the bioplastic was taken out from the submerged water and immediately wiped off the access water on the plastic surface before weighing the bioplastic mass. The same procedure was triplicate done to ensure the accuracy of the measurement. The trial-and-error test was conducted before the actual experiment to estimate the duration for the bioplastic to be degraded. The percentage of weight increase was calculated for each bioplastic using equation 1.

$$A_w(\%) = \frac{W_f - W_i}{W_f}\ x\ 100\% \qquad \textit{Equation 1}$$

where A_w is water absorption, W_f is bioplastic final weight (wet weight) and W_i is the initial weight (dry weight).

The moisture content was tested by placing the developed plastic into an oven for 2 minutes at 105 °C. The initial and final weights of each bioplastic were recorded. The final weight was obtained by weighing the plastic using an electronic balance until the constant weight was gained. The test was repeated consecutively. The percentage of moisture content was calculated using equation 2.

$$M_c(\%) = \frac{m_f - m_i}{m_f}\ x\ 100\% \qquad \textit{Equation 2}$$

where M_c is moisture content, m_f is bioplastic final weight (constant weight after drying in an oven) and m_i is the initial weight (weight before drying in an oven).

An experimental study of water vapour permeability (WVP) for bioplastic was conducted following the gravimetric method at 25 ℃ as stated in ASTM E-96-00. The measurement started with sealing the plastic samples using elastic-plastic in Erlenmeyer flasks containing silica. The flasks were placed in a desiccator at 75% relative humidity (RH) at 25 ℃, as Luchese et al. (2015) described. WVP was estimated by determining the sample weight gained after 48 h, according to Equation 3.

$$WVP = \frac{w}{t} . \frac{1}{A} . \frac{e}{P_S(RH_1 - RH_2)} \qquad \textit{Equation 3}$$

where $\frac{w}{t}$ is weight gained (g/d), A is biofilm area (m^2), e is the average thickness of biofilm (µm), P_S is saturation pressure of water vapour at tested temperature (mmHg), RH_1 is relative humidity inside the Erlenmeyer flask, and RH_2 is relative humidity inside the desiccator. This mathematical equation assumes that the film solubility and diffusivity are constant where Henry's law and Fick's first law are applied in computing plastic film WVP (Bertuzzi et al., 2007).

RESULTS AND DISCUSSSION

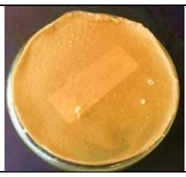
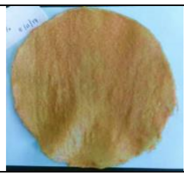
In this research, bioplastic with 100% CPH acted as a control. It was used to compare the bioplastic properties enhancement after incorporating CPH with kenaf fibre in bioplastic formulation.

Sensory Evaluation of Bioplastic

Table 1 shows the sensory evaluation of the developed bioplastics. Bioplastic made with 100% CPH showed a sweet smell due to the CPH contains 33.0 ± 0.6 g of sugar in 1 kg CPH as elaborated by Eghosa et al. (2010). This bioplastic also produced light brown and has a soft texture. Next, the two different ratios of bioplastic from 75% and 50% of CPH also shows the same result of the sensory evaluation, except the 50% CPH offers less sweet odour and more hard texture in the sensory evaluation.

Besides that, bioplastic with 25% of CPH shows no odour as the kenaf fibres help reduce the aroma of sweetness from the CPH component. Besides, the colour of this bioplastic is darker than the bioplastic made with 100%, 75% and 50% of CPH. Thus, it can be concluded that the higher the amount of kenaf, the darker the colour of bioplastic will be produced. This bioplastic texture is harder than the other bioplastic with a lower quantity of kenaf fibres. The bioplastic made from 100% kenaf fibres shows no odour, dark brown, and the hardest texture compared to the other four plastics.

Table 1. Sensory evaluation of bioplastic

Physical Appearance					
Ratio of CPH to Kenaf	100:0	75:25	50:50	25:75	0:100
Colour	Light yellowish brown	Yellowish brown	Dark yellowish brown	Reddish brown	Dark reddish brown
Texture	Soft, smooth, chewy	Soft, rough	Medium, rough	Slightly hard, rough	Hard, rough, brittle
Smell	Sweet Smell	Sweet Smell	Less Sweet smell	Odourless	Odourless

Drying Time of Bioplastic

The drying time of bioplastic is the time taken for the bioplastic to be completely dried. Plastic made from 100% CPH took seven days to dry, the longest drying time than the other bioplastics. The Time taken for the bioplastic made from 75% and 50% of CPH was the same (five days). The bioplastic produced from 25% of CPH took six days, while 100% of kenaf bioplastic only took two days to dry, as shown in Figure 2. The graph indicates that the amount of CPH to kenaf in bioplastic formation affects bioplastic drying time. For example, plastic from 100% CPH took the longest drying time while plastic from 0% CPH took the shortest drying time. However, plastic with 25% CPH presented a longer drying time than plastic with 75% and 50% CPH. This extended drying time might be due to an error which the weather and temperature surrounding the experimental workplace.

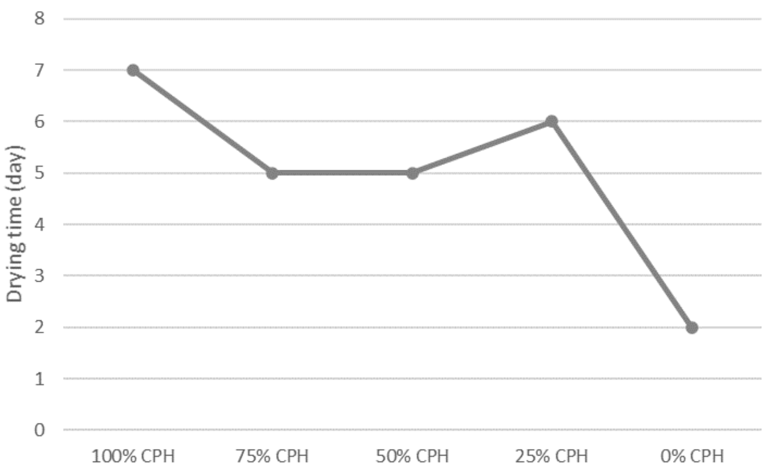


Figure 2. Drying time for the developed bioplastic films

Water Absorption

The percentage of water absorption for all developed bioplastics was measured and calculated using Equation 1. Water absorption for 100% CPH bioplastic shows 25.51%, the highest value exhibited in Figure 3. This result indicates that the CPH can easily absorb water around them. The addition of kenaf fibre in plastic formulation presents the reduction of water absorption in plastic. This phenomenon can be seen for 75% and 50% of CPH in bioplastic with only 15.02% and 14.71% of water absorption, respectively. There was a sharp decrease in the percentage of water absorption for these two ratios. These results indicate that the kenaf fibres help improve water absorption properties making the plastic more suitable for food packaging.

Water absorption for 25% of CPH gives a value of 24.57%, while 0% of CPH only gives a value of 6.47%, as presented in Figure 3. According to this figure, the bioplastic from 0% of CPH absorbs a small amount of water. However, kenaf fibres are known as natural fibres, consist of hydroxyl groups. These groups can form the intermolecular hydrogen bonds (inside the macromolecule or intramolecular and between another cellulose macromolecule), which indicates that all-natural fibres are hydrophilic, loving water based on Aji et al. (2009). Even though this plastic absorbs a small amount of water, it is enough to destroy the hydrogen bonds to make the plastic melt. Thus, this plastic is not suitable to be applied as food packaging.

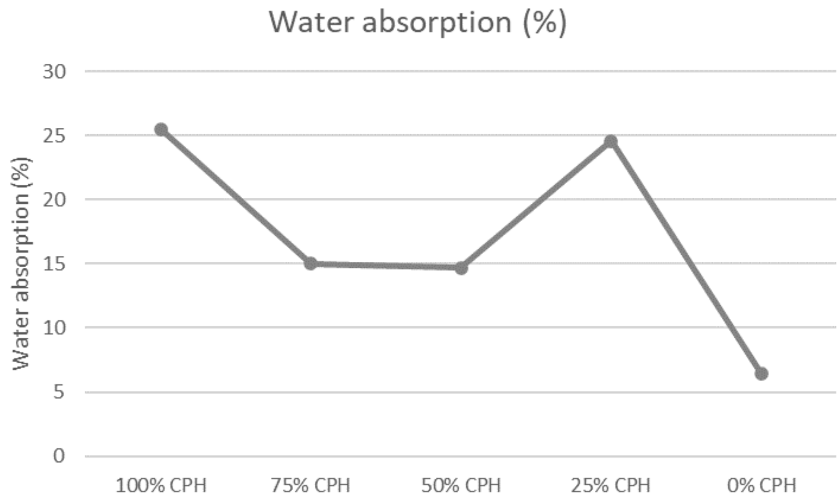


Figure 3. Percentage of water absorption for the developed bioplastic films

Moisture Content of Bioplastic

In this study, moisture content measures the amount of water inside the developed bioplastic films. Moisture content is vitally important because some polymers degrade when wet moulded, resulting in the property changing. The research shows that 100% of CPH moisture content is 20.89%, as presented in Figure 4. The result shows the highest reading as the CPH excellence in absorbing water and moisture.

Water content loss in the 25%, 50% and 75% of CPH bioplastic are 5.89%, 6.92% and 8.36%, respectively. The trend is increasing as the content of CPH increases. However, water content for 0% of CPH bioplastic shows the highest value, 12.87%. According to Mohanty et al. (2000), moisture content for the natural fibres obtained for jute, flax, and hemp was 12.6%, 10.0% and 10.8%, respectively. The moisture content for 100% of kenaf bioplastic is 12.87%, slightly higher than the previous research. The natural fibres show the value of moisture content is more than 10%, and it is parallel with the results from this study.

Furthermore, the moisture content reading decreases as increasing the percentage of CPH extract in bioplastic development. The moisture content reading drops from 12.87% to 5.89% and slightly increases in 50% of bioplastic from cocoa pod husks which are 6.92%. For the result of the bioplastic ratio of 75% cocoa pod husks, the reading shows 8.36% of moisture content and an increase sharply for 100% cocoa pod husks bioplastic. Daud et al. (2013) tested the chemical composition and morphology in CPH and found that the water content for the pure CPH is 14.1±0.05 (%). The moisture content obtained from this research is within 8.38% and 20.89%, which is parallel with the previous study. However, the inconsistent result may be occurred due to the types of additives used or incompatibility of the procedure in bioplastic production.

Sorbitol used in the bioplastic formulation comprises several hydroxyl groups in the molecules, which increase the ability to interact with water by hydrogen bonding (Al-Hassan and Norziah, 2012) as it is immersed in the water. The presence of sorbitol increases the intermolecular forces and lower the capability to interact with water (García-Ayuso et al., 2000). Consequently, the chances of sorbitol interacting with the cellulose chain in CPH are low. This makes the cellulose chain act as a water absorber and creates the high moisture content of a higher concentration of CPH.

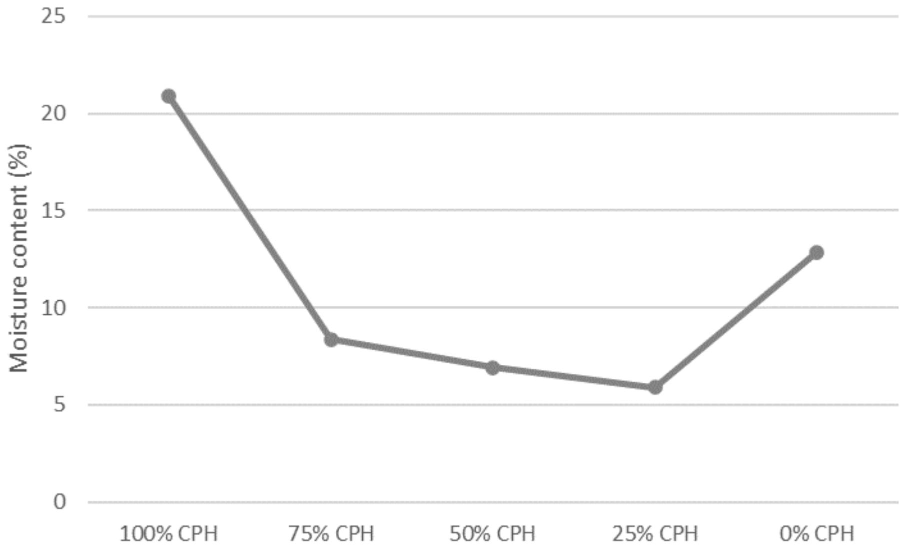


Figure 4. Moisture content for the developed bioplastic films

Water Vapor Permeability (WVP)

Figure 5 presents the results for water vapour permeability for all prepared bioplastic films. Bioplastic with a ratio of 0% CPH shows the highest value of WVP, while bioplastic with a ratio of 50% CPH displays the lowest value of WVP. These WVP values determine the ability of bioplastic to permit the moisture transfer between food and environment, where bioplastic with a low value of WVP exhibits the highest possibility to prevent the moisture transfer between food and environment (Azmin et al., 2020; Luchese et al., 2015). Hence, bioplastic film with a 50% CPH could be applied as food packaging because it showed the lowest value of WVP. Other research found that the edible films and coatings associated with cellulose derivatives could be used for food packaging and preservation (Bertuzzi et al., 2007).

The sorbitol applied in bioplastic formulation modifies the cellulose network's molecular organization. It increases the free volume resulting in a less dense network that results in more permeable films to water (Bourtoom et al., 2006). This phenomenon has been proven (displayed in Figure 5) as increase the ratio of CPH also increase the value of WVP. However, the interaction of the same ratios of CPH and kenaf does not permit the water space in the bioplastic. Adding more kenaf fibre in the bioplastic increases moisture's potential to be absorbed by the plastic. The results in this study are parallel with the research done by McHugh et al. (1993).

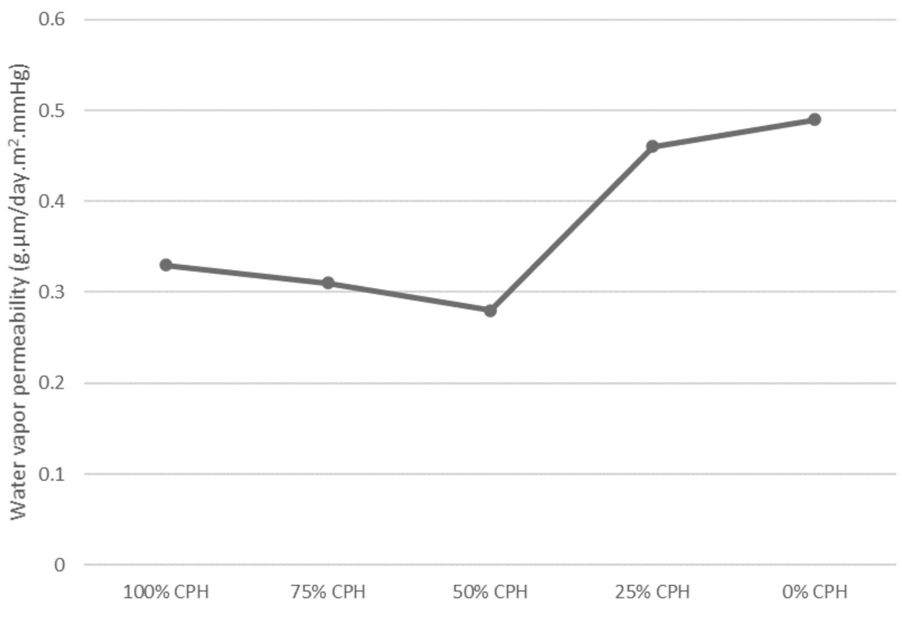


Figure 5. Water vapour permeability for the developed bioplastic films

CONCLUSION

Sensory evaluation and physicochemical properties tests of five bioplastic films containing different ratios of CPH to kenaf fibre were successfully conducted. 50% of CPH bioplastic was found the best film after accessing sensory, drying time, water absorption and moisture content. This plastic showed the best in sensory, moderate in drying time (five days), water absorption (and moisture content (6.92%). 100% of kenaf bioplastic ranked the lowest in sensory but shortest drying time (2%), lowest in the percentage of water absorption (6.47%) but highest in the percentage of moisture content (12.87%). However, this plastic melts in a short time (less than 3 minutes) in a water absorption test. Thus 100% of kenaf bioplastic is not suitable to be utilized as a food packaging container.

Patents: Not applicable.

Author Contribution

For research articles with several authors, a short paragraph specifying their individual contributions must be provided. The following statements should be used "Conceptualization, Siti Nuurul Huda Mohammad Azmin and Nur Syazwin Eleena Mohamad Sharif; methodology, Siti Nuurul Huda Mohammad Azmin and Mohd Shukri Mat Nor; software; validation, Siti Nuurul Huda Mohammad Azmin, Nur Syazwin Eleena Mohamad Sharif and Palsan Sannasi Abdullah.; formal analysis, Nur Syazwin Eleena Mohamad Sharif; investigation, Siti Nuurul Huda Mohammad Azmin, Nur Syazwin Eleena Mohamad Sharif, Mohd Shukri Mat Nor and Palsan Sannasi Abdullah; resources, Nur Syazwin Eleena Mohamad Sharif; writing—original draft preparation, Siti Nuurul Huda Mohammad Azmin and Nur Syazwin Eleena Mohamad Sharif; writing—review and editing, Siti Nuurul Huda Mohammad Azmin; visualization, Siti Nuurul Huda Mohammad Azmin and Nur Syazwin Eleena Mohamad Sharif; supervision, Siti Nuurul Huda Mohammad Azmin; project administration, X.X.; funding acquisition, Siti Nuurul Huda Mohammad Azmin. All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest: No conflicts of interest

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