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Sugar palm nanocrystalline cellulose reinforced sugar palm starch composite: Degradation and water-barrier properties

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Abstract. In this work, sugar palm nanocrystalline cellulose (SPNCCs) nanocomposites were prepared and used as a biodegradable reinforcement material to improve the water vapor barrier properties of the sugar palm starch (SPS)-based films. SPNCCs with different size based on hydrolysis time (30, 45 and 60 minutes denoted as SPS/SPNCCs-30, SPS/SPNCCs-45, and SPS/SPNCCs-60) were incorporated into SPS plasticizers with glycerol and sorbitol using solution casting method. Then the SPS and SPS/SPNCCs bionanocomposites were submitted to biodegradation by means of soil burial experiment and water vapor barrier test. The biodegradation test shows that SPS degrades very quickly than SPS/SPNCCs which lose 61.93% of its weight at the end of 7 days compared to the SPS/SPNCCs-60 bionanocomposite 52.61%. Adding 0.5 wt.% SPNCCs-60 loading significantly improve water vapor permeability (WVP) of the nanocomposite film by 19.94% compared with the neat film. This was ascribed to the high compatibility between SPNCCs and SPS matrices, which was supported by the field emission scanning electron microscopy (FESEM).

1. Introduction

Nowadays many researchers are developing fully biodegradable composites or biocomposite due to the continuing environmental catastrophe caused by non-biodegradable plastics. Generally, these biocomposites are composed of either natural matrices or natural fibres [1]. Thus in order to overcome this problems, natural biopolymers such a starch, polylactic acid (PLA) and polyhydroxyalkanoates (PHAs) are introduced as a potential alternatives to conventional plastics for packaging applications [2].

Starch is one of the potential candidates for the replacement of non-biodegradable plastic due to its ease availability, renewability, affordable and most importantly biodegradable [3]. Besides, these biopolymer also has been explored widely for the potential manufacture of products such as food and fruit packaging films, plastic shopping bags, flushable linter, medical delivery system and device [2,4]. Nevertheless, the utilization of starch polymer as a films have been reported to have low water barrier properties [5-6]. Thus, many studies have been conducted to improve the water sensitivity of starch-based materials without negotiating their biodegradability [7-9]. Consequently to overcome this limitations, the addition of nanosize natural fibres during the preparation of starch biopolymer film is an effective strategy for enhancement of the packaging films properties [10]. Nanosize natural fibres such



as nanocrystalline celluloses (NCCs) and nanofibrillated celluloses (NFCs) can be extracted through the process of chemical and mechanical methods. Hydrolysis is one of the chemical methods that widely used to extract NCCs. Recently, NCCs has attracted a tremendous interest in material science study due its nanodimension, high crystallinity, high aspect ratio, high mechanical strength, low density as well as easy availability along with highly abundant [2, 11]. Moreover, the addition of NCCs has been reported to reduce the water vapor permeability of starch based film. This may due to the morphology of NCCs which increased the tortuosity in the starch based films leading to slower water vapor permeability processes, thus increasing their suitability for food packaging application [12].

Various types of nanosize cellulosic reinforcement have been determined and experienced in biopolymers. Interestingly, it was found that the compatibility between nanosize fibres and starch matrix is high, which is significant for improvement of the film water sensitivity [7]. Nanosize fibre or nanocrystalline cellulose (NCCs) can be isolated from biomass like jute [13], hemp [10], bamboo [14], corncob [15] and sugar palm [16]. Sugar palm is investigated to be a good source of cellulose fibres because of its high cellulose content, which then this cellulose was extracted to gain NCCs [16-17]. Sugar palm tree is versatile tree that grown in mostly tropical countries. Interestingly, the inner part of sugar palm stem contains starch which can be utilized as a biomatrix [18]. In this paper we developed a novel biodegradable bionanocomposites where the matrix (SPS) and nanofibre (SPNCCs) are derived from the similar natural source i.e sugar palm tree.

One of the potential application of SPS/ SPNCCs bionanocomposites is it can be used in packaging industries. The degradation is one of the indicator of packaging durability. Since there are no information on the biodegradability of SPS/SPNCCs bionanocomposites has been reported in the literature, therefore in this study is done to investigate how the addition of the various size of sugar palm nanocrystalline cellulose (SPNCCs) enhances the water vapor permeability and its effect on biodegradability of the sugar palm starch film and bionanocomposites. These basic data therefore are essential for the design and use of the resultant bionanocomposite.

2. Experimental procedure

2.1. Materials

Sugar palms fibre (SPF) and sugar palm starch (SPS) were extracted and collected from sugar palm trees located in Jempol, Negeri Sembilan (Malaysia). Sodium hydroxide, sodium chlorite (80 % purity), acetic acid, sulphuric acid (98 % purity), sorbitol and glycerol plasticizer were supplied by Evergreen Sdn Bhd (Semenyih, Malaysia).

2.2. Preparation of SPNCCs

Sugar palm fibres (SPF) were collected from different part of sugar palm trees (sugar palm frond, trunks, fibre and bunch) as it wrapped around the tree from top to bottom. The knife was used to remove the SPF from the tree. Then a Fritsch pulverisette mill was used for grinding and screening to gain an even size of SPF (2 mm). The cellulose preparation procedures were reported elsewhere [7, 16, 19]. Briefly, the SPF were extracted using two main processes known as delignification and mercerization. The initial and second processes of production of sugar palm cellulose (SPC) were conducted in accordance with ASTM D1104-56 (1978) and ASTM D1103-60 (1977) for the removal of lignin and hemicellulose, respectively, resulted in production of sugar palm celluloses (SPC). Colloidal suspensions of SPNCCs were prepared by acid hydrolysis treatment of SPC. The SPC were mixed with 60 wt% sulphuric acid aqueous solution at variable hydrolysis time of 30, 45, and 60 minutes with the ratio of the SPC to liquor was 5:100 (wt%). The excess sulphuric acid was then removed by centrifuge SPNCCs 5 times with 6,000 rpm, 10 min at 8 °C. Then SPNCCs were dialyzed with distilled water until a constant pH of 7 was gained. The resulting suspension were sonicated using ultrasonic treatment for 30 minutes [20]. Finally SPNCCs were freeze-drying to be used for nanocomposite film.

2.3. Preparation of the SPS/SPNCCs nanocomposite films

The fabrication of plasticizer sugar palm starch/ sugar palm nanocrystalline celluloses (SPS/SPNCCs) composite film was based on a solution casting methods. Starch, sorbitol, glycerol, SPNCCs and distilled water were mixed and sonicated together in order to obtain homogenous nanocomposite film [7]. Briefly, a solution of SPNCCs was prepared by mixing and sonicating it with 190 mL of distilled water with known concentration of SPNCCs (0.5 wt. % on the starch basis). The SPNCCs content was fixed at 0.5 wt. % based on the starch basis. Then, ten gram of SPS and plasticizer (30% on the starch basis) was mixed with the SPNCCs solution and stirred at 1000 rpm for 20 minutes at 85 °C in a disperser for the starch to be gelatinized. The ratio of plasticizer used is 1:1 with combination of sorbitol and glycerol ratio. Then the film-forming suspension was left cooling down and placed it under vacuum to remove air bubble inside the suspension before 45 g of the suspension was cast into each petri dish sized of 15-cm diameter. The petri dishes containing the film-forming solution were placed in an oven at 40°C overnight. SPS films prepared without SPNCCs served as the control (designed as SPS film). Whereas the nanocomposite film with different hydrolysis time of 30 min, 45 min and 60 min were denoted as SPS/SPNCCs-30, SPS/SPNCCs-45, and SPS/SPNCCs-60, respectively. The resulting films were kept in the desiccator at room temperature for one week to ensure the equilibrium of the water content in the films before doing any characterization.

2.4. Morphology analysis by FESEM

Surface characteristics of the SPS and SPS/SPNCCs films were determined by a FEI NOVA NanoSEM 230 machine (FEI, Brno-Černovice, Czech Republic) with an accelerating voltage of 3 kV in order to observe the degradation phenomenon after the biodegradability test. Before being tested by FESEM, samples were cut into 1cm × 1cm and then coated with gold to avoid charging using an argon plasma metallizer (sputter coater K575X) (Edwards Limited, Crawley, United Kingdom) [21].

2.5. Morphology analysis by TEM

The structure and diameter measurement of sugar palm nanocrystalline cellulose (SPNCCs) were determined by Hitachi H-7100 transmission electron microscopy (Hitachi, Tokyo, Japan). These image captured of the sample was obtained from high-resolution transmission electron microscopy (HRTEM). Before being tested by HRSEM, samples were placed on the surface of copper grid coated with a thin carbon film. After that the samples were stained with uranyl acetate for 1 min and allowed to dry at room temperature. The purpose of staining is to get better visualize and contrast image under a TEM.

2.6. Water absorption

Water absorption analysis was run in accordance to ASTM D 570-98. Firstly, the samples were dried for 24 h at 50 °C and cooled in desiccator to gain an even weight. Then the films were weighed and soaked in distilled water at room temperature. After a particular soaking period, the films were taken out of the water and wiped with a cloth and weighed again. The differences between the initial and final masses of the films were calculate using equation [22]:

$$\text{Water absorption (\%)} = \frac{M_{\text{final}} - M_{\text{initial}}}{M_{\text{initial}}} \times 100 \quad (1)$$

Where, M_{initial} = mass before, M_{final} = mass final

2.7. Biodegradability test

Biodegradability was determined by measuring the weight loss of the bionanocomposite buries in compost soil under moisture controlled condition. The size of samples of 30 × 10 mm with triplicate of each samples were buried 100 mm below the surface of soil, which was regularly moistened with distilled water [1]. Each sample was dug out of the compost soil after being buried for 24, 48, 72 and 168 h, respectively, and then washed with water and dried to a constant weight at 60 °C in a vacuum oven. The weight loss was then determined using the equation below:

$$\text{Weight loss (\%)} = \frac{W_0 - W_t}{W_0} \times 100 \quad (2)$$

Where, W_0 =weight before being buries, W_t = weight after being buried.

2.8. Water vapor permeability (WVP) test

The film samples were conditioned in a desiccator with a relative humidity of 50% at room temperature. The WVP test was investigated according to ASTM E96 (1995), with slight modifications according to Sanyang et al. [23]. The experiment was repeated thrice. Initially, the mouth of the cup which was 30 mm was filled with 20 g of silica gel. Circular film samples were cut and placed over the mouth of the cup, leaving about 3 mm vacuum to the top. Then it was sealed neatly. After that, the test cups were measured it mass before placing in a constant relative humidity chamber (25°C, relative humidity 75%). The mass gain values of the cups were measured periodically until the equilibrium state was reached. The mass increment of the test cups were recorded, and WVP was calculated as below,

$$WVP = \frac{(m \times d)}{(A \times t \times P)} \quad (3)$$

Where m (g) is the increment of test cup mass, d (mm) is the thickness of the films, A (m^2) is the area of film exposed, t (s) is the interval time of permeation, and P (Pa) is the water vapor partial pressure across the films. The results are in the unit of $10^{-9} \times g \cdot s^{-1} \cdot m^{-1} \cdot Pa^{-1}$.

3. Experimental result and discussion

3.1. Physical Properties of Sugar Palm Nanocrystalline cellulose (SPNCCs)

The physical properties of the sugar palm nanocrystalline cellulose (SPNCCs) from different treatment was determined using Image J software. The images were collected from the TEM analysis.

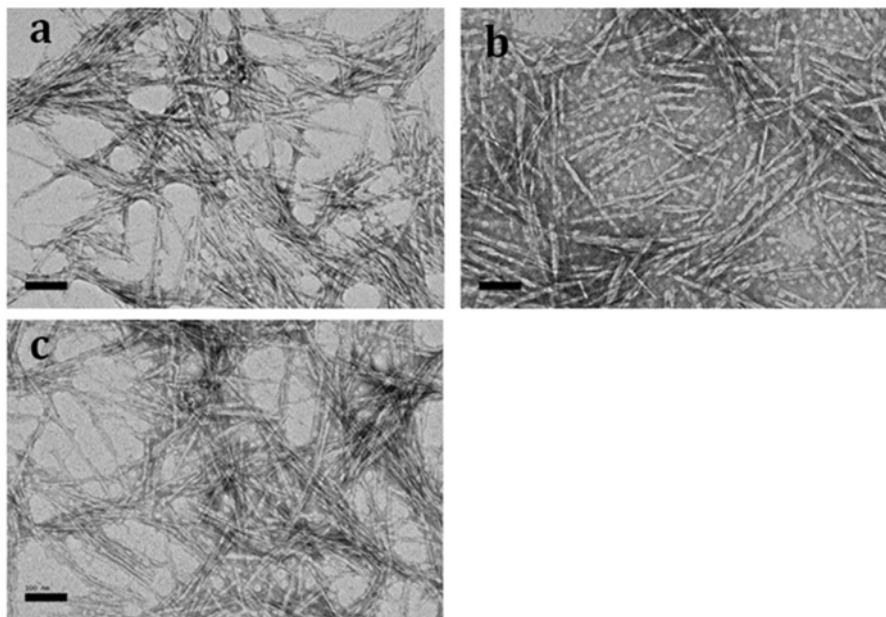


Figure 1. TEM micrographs of the (a) SPNCCs-30, (b) SPNCCs-45 and (c) SPNCCs-60.

Table 1. Physical Properties of SPNCCs-30, SPNCCs-45 and SPNCCs-60.

Fibres	Length (nm)	Diameter (nm)
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Sugar Palm Cellulose (SPC)	-	11870
SPNCCs-30	175 ± 37.01	13 ± 1.73
SPNCCs-45	130 ± 30.23	9 ± 1.96
SPNCCs-60	110 ± 33.69	7.5 ± 1.35

Figure 1 and Table 1 show the TEM micrograph and physical properties of nanocrystalline cellulose, respectively. TEM micrograph of sugar palm nanocrystalline cellulose (SPNCCs) (Figure 1) revealed their nanometric dimension. As shown in Table 1, the average length of the SPNCCs-30, SPNCCs-45 and SPNCCs-60 were approximately 175 ± 37.01 nm, 130 ± 30.23 nm and 110 ± 33.69 nm, respectively. Whereas the diameter of the SPNCCs-30, SPNCCs-45 and SPNCCs-60 were approximately 13 ± 1.73 nm, 9 ± 1.96 nm and 7.5 ± 1.35 nm, respectively. The diameter measured were similar to the nano-sized structures that were extracted from other agro-waste sources such as coconut husk (5.5 ± 1.5 nm) [24], rice straw (5.06 nm) [25], soy hulls (4.43 ± 1.20 nm) [26], sugarcane bagasse (4 ± 2 nm) [27], banana residues (5nm) [28] and smaller than microfibrils from wheat straw (10–80nm) [29], sisal fibres (30.9 ± 12.5 nm) [30] and sugarcane bagasse (30nm) [31]. These resultant images (Figure 1) revealed the aqueous suspensions contained sugar palm NCCs residing mostly of individual crystal and some aggregates.

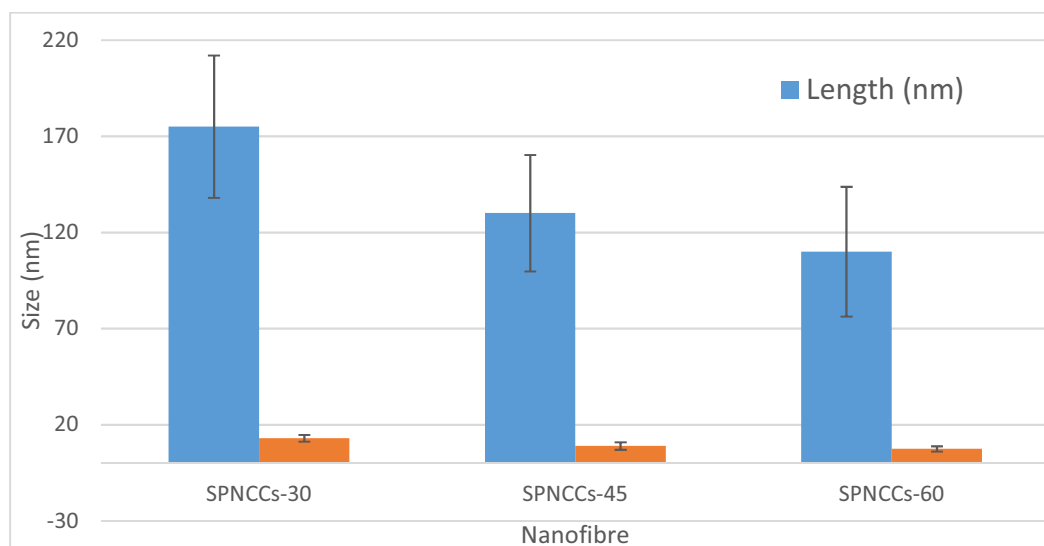


Figure 2. Length and diameter of SPNCCs-30, SPNCCs-45 and SPNCCs-60.

Figure 2 shows the length and diameter of SPNCCs-30, SPNCCs-45 and SPNCCs-60. The differences between the SPNCCs-30, SPNCCs-45 and SPNCCs-60 were in the reduction of size of length and diameter. Based on Table 1, the reduction of the SPNCCs-30 compared to SPC was 99.89%. This was attributed by the removal of the amorphous region via the hydrolysis treatment on SPC [16]. The process of the hydrolysis continued for the SPNCCs-45 and SPNCCs-60 for 45 minutes and 60 minutes, indicated the changes in the size of the SPNCCs which was reduce by 31% and 42% compared to SPNCCs-30, respectively. This was caused by the longer reaction time of H_2SO_4 via hydrolysis treatment on the fibres, which removed amorphous region from the nanofibres. Besides, longer hydrolysis time could irritate the structure (length and diameter) of SPNCCs. Thus, the longer the reaction times of hydrolysis, the smaller the length and diameter of the nanofibres [32].

3.2. Water absorption

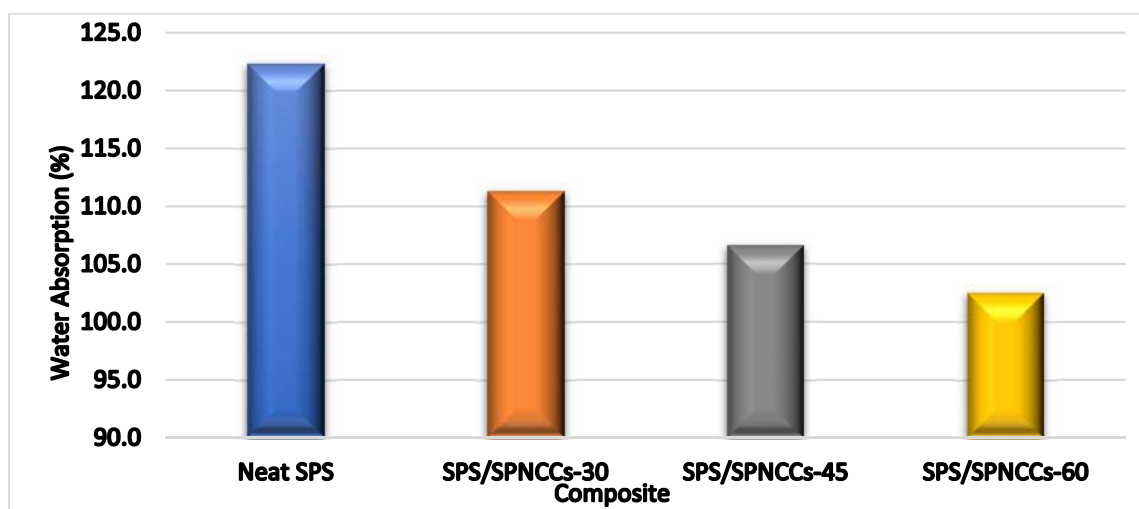


Figure 3. Water absorption of composite.

Water absorption is a significant study in SPS film due to water acts as a plasticizer. Figure 3 displays the percentage of water absorption of SPNCCs-30, SPNCCs-45 and SPNCCs-60 nanocomposite films. As been shown in Figure 3, it can be seen that the neat SPS gave the highest percentage of water absorption, followed by SPS/SPNCCs-30, SPS/SPNCCs-45, and SPS/SPNCCs-60, i.e. 111.3 %, 106.6 % and 102.5 %, respectively. This indicated that film reinforced with SPNCCs has lower hydrophilicity. Moreover, the water absorption of the film is high due to the presence of hydroxyl groups in film molecules. Film with higher concentration of starch is tend to absorb more water compared to film that have low concentration of starch [1]. Besides that, as the size of SPNCCs decreased, the percentage of the water absorption also decreased. Additionally, film with big particle size of SPNCCs exhibited greater tendency to absorb water which possibly due to a lower response to stress [33]. In this study, water absorption tests revealed that stabilizing the mixtures with SPNCCs decreased the water absorption capacity. This is due to the adhesion between the SPNCCs and SPS in which they tend to form strong hydrogen bonding via the physical interlocking with non-polar matrices and the OH- groups if hydrophilic polymer matrices [34-35]. The result of this experiment conducted are quite similar to those from previous work [1].

3.3. Biodegradation of bionanocomposites

Biodegradation is the decomposition of materials by the action fungi, bacteria and microorganism or by other biological means. Generally, the decomposition of polymer start when these microbial organism contact with the biodegradable polymer [36]. These microbial organisms transform the polymer through enzymatic or metabolic process that broken down the polymers into smaller compound which have lower average molecular weight. Therefore, this favors material's decomposition in the environment. When the biodegradation is complete, the process is called mineralization [37]. Figure 4 shows the weight loss of the SPS and SPS/ SPNCCs bio-nanocomposite after biodegradation testing. At the end of day 7, the weight of neat SPS had lost 61.94%, whereas the SPS/SPNCCs bionanocomposites had lost 56.88 %, 55.74 % and 52.61 % weight for SPS/SPNCCs-30, SPS/SPNCCs-45, and SPS/SPNCCs-60, respectively. The average degradation rate is 8.85 %/day, 8.13 %/day, 7.96 %/day and 7.52 %/day, respectively for the neat SPS, SPS/SPNCCs-30, SPS/SPNCCs-45, and SPS/SPNCCs-60, respectively. It is observed that the weight loss of SPS/SPNCCs composite is lower than that of the SPS at any given time points.

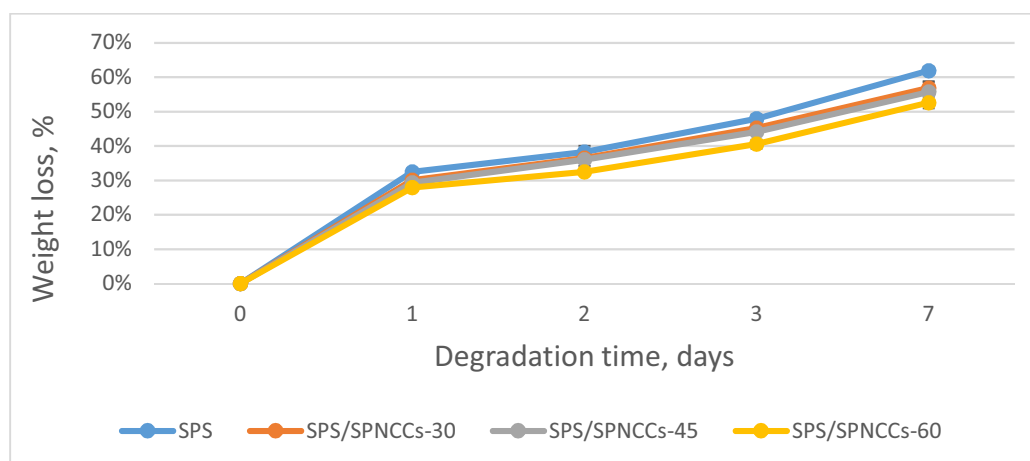


Figure 4. Weight loss of SPS/SPNCCs composite films as a function of soil burial time.

The control film, SPS film was totally degraded after 9 days while it took bio-nanocomposites films 12 days to degrade completely. The weight loss of for the neat matrix was higher compared to the bio nanocomposite films. There are two factors that may be attributed to this situation to occur which are water absorption by film and degree of crystallinity of SPNCCs in bio-nanocomposite films. SPNCCs facilitated the polymer matrix with a stabilization effect by creating a three-dimensional cellulosic network which restricted the chain mobility and reduced the number of hydroxyl groups, and thus reduced water absorption of SPS/SPNCCs nanocomposite films [38-39]. The weight loss for the control film, SPS was higher compared to the SPS/ SPNCCs bionanocomposite for the entire successive degradation test. This may be attributed to the physical properties of SPS which SPS absorb more water than SPS/ SPNCCs films, making it more exposed to microbial organisms attack [41]. These microbial organisms attack the SPS in the presence of water medium [42]. This can be related to the water absorption properties of starch films, where the water absorption for neat SPS was about 122.3 %, whereas for SPS/SPNCCs-30, SPS/SPNCCs-45, and SPS/SPNCCs-60 bionanocomposite films were 111.3 %, 106.6 % and 102.5 %, respectively. This properties could be attributed to the hydrophilic behavior of SPS [41]. Thus, the samples that have higher starch contents would have better biodegradability properties.

Generally cellulose is comprised of amorphous (less-oriented molecules) and crystalline region (highly oriented molecules). When it was treated with strong acid via hydrolysis process, the amorphous region broke up, thus producing NCCs with higher degree of crystallinity. The capability of cellulolytic microorganisms to degrade cellulose differs greatly with the physic-chemical characteristics of the substrate such as polymerization of cellulose and the degree of crystallinity [43], where the degree of crystallinity cellulose is major structural parameter [44]. It has been described in a previous work that crystalline region are more difficult to degrade [45]. Compared to control film SPS, SPS/SPNCCs contained higher crystallinity, which makes it a higher resistance to the microbial organism attacks than starch. This may attributed the difference in weight loss between SPS and the SPS/ SPNCCs bionanocomposite. Moreover, the variances in resistance to microbial organism attacks between SPS/ SPNCCs and starch SPS exhibited that in SPS/SPNCCs bionanocomposite, microbial organism attacks start with starch. In the case of SPNCCs/ starch, the amorphous region favors the microbial organism accessibility to the matrix (mainly to the deterioration of starch) and SPNCCs have the minor role, as can be concluded from the difference in weight loss suffer by the SPS and the SPS/SPNCCs matrix [1].

When the microbial organisms consume the surrounding starch, the bionanocomposite film loses their structural integrity. Thus lead to the deterioration of the mechanical properties, hence allowing the attack of SPNCCs by microbial organism [46]. Indeed, the result gained herein shows that the SPS/SPNCCs bionanocomposite film will not cause any deterioration/ ecological impact, where in other words, the SPS/SPNCCs bionanocomposite film are fully biodegradable.

3.4. Water Vapor Permeability

Hydrophobicity are one of the most important criteria that should be emphasize in the process of material selection for food packaging application, since it may affect the performance of the end products whether the product is water-sensitive product or not. Therefore, films with low water vapor permeability are suitable for food packaging applications, including the reducing and preventing of moisture transfer between surrounding and the food [47]. Hence, minimizing the water vapor permeability (WVP) of SPS films is important for their tremendous potential application. Figure 5 shows the WVP of neat SPS and SPS/SPNCCs composite film as a function of soil burial time. It can be seen that the highest WVP is SPS film which is $9.58 \times 10^{-10} \times g.s^{-1}.m^{-1}.Pa^{-1}$. This is because of their hydrophilic nature, besides their sensitivity to moisture contain and surrounding humidity, a factor that is hard to control [48]. The reinforcement of starch with SPNCCs significantly improved the WVP of the neat SPS films. The addition of 0.5% SPNCCs into neat SPS films reduced their WVP value by 11.17 %. This decreased is due to the tortuous path caused by the presence of rigid crystalline structure and the dispersion of SPNCCs in the starch polymer, which prolongs or hinders the path of water molecules through the film matrix [7,49]. The WVP of the film showed an improved of 11.17 %, 14.72 % and 19.94 %, when altering the size of the SPNCCs reinforced bio matrix, SPS/SPNCCs-30, SPS/SPNCCs-45, and SPS/SPNCCs-60, respectively. This increments trending showed that the smaller the size of the nanofibres the harder the water to pass through the matrix SPS, thus improved the WVP of the neat SPS films.

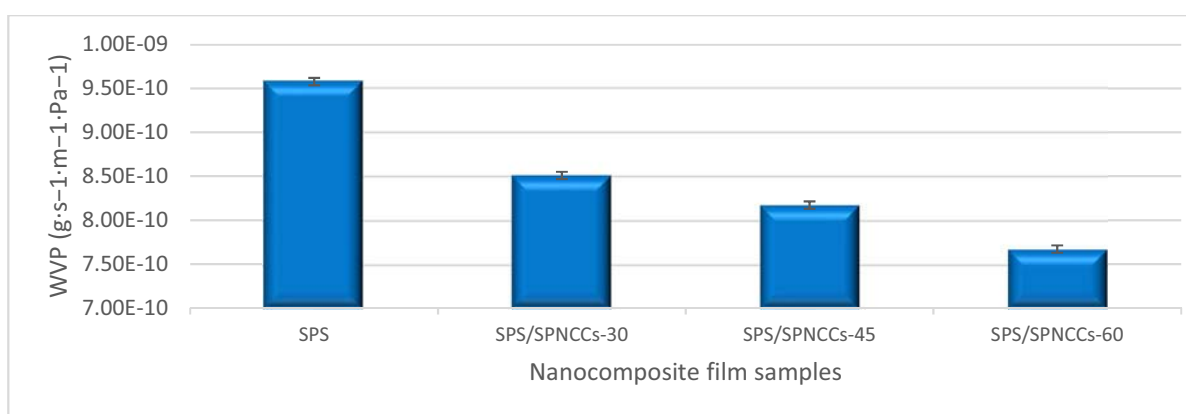


Figure 5. Water Permeability of SPS/SPNCCs composite films as a function of soil burial time.

3.5. Surface Morphology of Bionanocomposite Film

Figure 6 displayed the surface morphology of SPS-based films with and without the addition of SPNCCs after being degraded in soil test for 168 h. From the figure 6, it can be observed that most of the starch had degraded in both SPS and SPF/SPS bionanocomposite which was observed using FESEM. Nevertheless, this effect of degradation can be observed on the weight loss of the bionanocomposite, as explained earlier and observed in figure 4. Therefore it can be concluded that the addition of SPNCCs delays the degradation of the bionanocomposite. The micrograph of the neat SPS films showed a smooth and continuous surface with no trace of starch granular or cracks and agglomerations of SPNCCs. Similar observations were reported by Sanyang et al. (2016) and Dias et al. (2011) for neat sugar palm starch and rice flour films, respectively. Thus, the high dispersion of SPNCCs (Figure 6(b)) is a good sign of strong interfacial adhesion between the two components of the SPS-SPNCCs nanocomposite film. These good distributions translate into high WVP properties. Bilbao-Sainz et al. (2011) described that the functional properties of the composite improved when the fibres well-dispersed into the polymeric matrix. Besides, in Figure 6 (b) of neat SPS film after the process of degradation take place, the surface of the film become rough and wavy due to the microbial attack within the soil. Whereas in Figure 6 (b) of SPS/SPNCCs film observed the present of excessive numbers of SPNCCs distributed on

the surface of SPS/SPNCCs film. This is due to the attacks of microbial on the surface of film which then revealed the image of SPNCCs that adhere within the film [1].

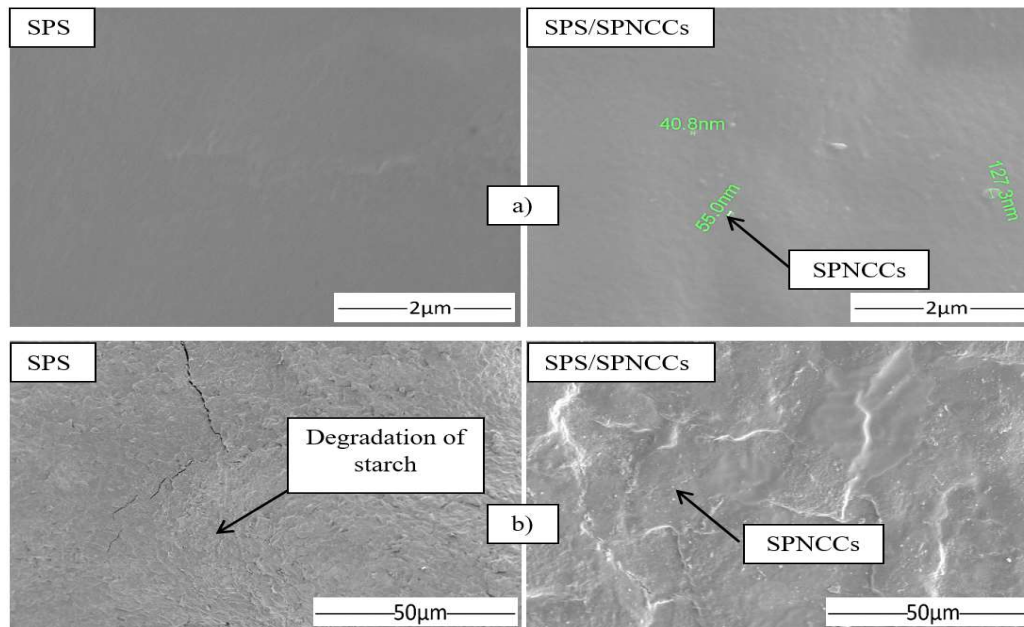


Figure 6. Surface morphology of SPS and SPF/SPNCCs bionanocomposite; a) before and b) after being buried.

4. Conclusion

An innovative bionanocomposite with combination of both matrix and natural fibres are obtained from less utilize of the sugar palm tree was successfully developed. It was determined that the WVP effect on the SPS and SPS/SPNCCs bionanocomposite had brought a significant improvement when decrease the size of the SPNCCs reinforced bio matrix, at the end of 168 h of WVP test. Whereas, the biodegradation test revealed that the SPS degraded faster as compared to SPS/SPNCCs bionanocomposites. Thus, this bionanocomposite shows good WVP and biodegradable properties for packaging materials application.

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