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Water Transport Properties of Bio-Nanocomposites Reinforced by Sugar Palm (*Arenga Pinnata*) Nanofibrillated Cellulose



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ARTICLE INFO	ABSTRACT
Article history: Received 18 June 2018 Received in revised form 2 August 2018 Accepted 13 November 2018 Available online 15 November 2018	In present work, sugar palm nanofibrillated cellulose (SPNFCs) nanocomposites were casted and utilized as biodegradable reinforcement materials to improve the water barrier properties of the sugar palm starch (SPS)-based films. SPNFCs with three different sizes based on cycling time (5, 10 and 15 cycles denoted as SPS/SPNFCs-5C, SPS/SPNFCs-10C, and SPS/SPNFCs-15C) were incorporated into SPS plasticizers together with glycerol and sorbitol using solution casting method. Then, the SPS and SPS/SPNFCs bionanocomposites were submitted for water absorption and solubility test. Adding of 0.5 wt. % SPNFCs-15C loading had significantly improved water absorption and water solubility properties of the nanocomposite films by 18.84% and 39.38%, respectively, compared with the neat film. This was ascribed by the high compatibility between SPNFCs and SPS matrices, which was supported under the observation of the field emission scanning electron microscopy (FESEM).
Keywords:	
Sugar Palm, Nanofibrillated Cellulose, Nanocomposites, Water Absorption,	
Water Solubility	Copyright © 2018 PENERBIT AKADEMIA BARU - All rights reserved

1. Introduction

Growing environmental awareness along with strict environmental policies have forced plastic manufacturing industry to discover new materials that can substitute synthetic petroleum-based plastic. The utilization of products that were made by conventional petroleum-based polymer had created many potential problems due to their non-renewable sources and ultimate disposal as reported by Ilyas *et al.*, [13]. Therefore, one of the alternatives is to substitute this conventional material with biodegradable biocomposites. This is due to their advantages, for instance, sustainable sources, environmentally friendly, more economical, light weight and simplified end-of-life disposal

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issues. Besides, biocomposite also have become substitute materials in some engineering applications. Briefly, biocomposite can be defined as combination of two or more elements, where at least one of the elements come from natural resources, either plant, animals or microorganism [25]. Concisely, biocomposites are comprised of reinforcement of natural fibre and matrix, either in the form of thermosetting or thermoplastic polymer. Whereas, bionanocomposite is a novel class of nanosized materials, in which they contain the constituent of biological origin and particles with at least one dimension in the range of 1–100 nm. The design of nanoscale structures could enhance the morphology, functional properties and stability of the polymer matrix used in the films and coating [12]. Cellulose is the most abundant renewable polymer in the world as it is found in plant cell walls (i.e. flax, hemp, kenaf and ramie), and it can also be synthesized by some bacteria (i.e. gluconacetobacter, agrobacterium, pseudomonas, rhizobium, and sarcin) [14]. Besides, it has remarkable reinforcing properties due to its abundant hydroxyl group within large surface area. There are two types of nanoreinforcements that can be obtained from plant cellulose; nanofibrillated cellulose (NFC) and nanocrystalline cellulose (NCC). These nanocellulose can be obtained by synthesizing the cellulose chain using either mechanical or chemical or both combined treatments. The NFC have nanosized diameter (2-20 nm, depending on the origin), with several micrometer length. Each NFC is formed by aggregation of elementary fibrils, which are made of alternating crystalline and amorphous regions. The crystalline region, also known as nanocrystalline cellulose (NCC) can be isolated via strong acid hydrolysis. NCC has nanosized diameter ranging 8-20 nm, with length ranging from 100 nm up to 1-2 μ m, resulting in high aspect ratio. Moreover, these nanofibre were reported to have high modulus (150 GPa) and high strength (10 GPa), in which they are very suitable for reinforcement purposes.

Recently, sugar palm fibre (SPF) has gained many attentions from many researchers due to its high proportion of cellulose which can be readily isolated from other component, such as hemicellulose and lignin. The cellulose was obtained from SPF by delignification and mercerization process [11]. Ilyas *et al.*, isolated nanocrystalline cellulose from SPF via hydrolysis treatment for the purpose of reinforcement with polymer matrix [10]. Malainine *et al.*, studied the effect of nanofibrillated cellulose from *Opuntia ficus-indica* parenchyma cell reinforced on starch matric and had revealed that the reinforcement of NFC enhanced their mechanical and water barrier properties [19]. However, no previous research works had been devoted to the development of sugar palm nanofibrillated cellulose (SPNFCs) reinforced thermoplastic starch matrix of which was derived from natural resources. In this study, biopolymer used for the matrix is extracted from sugar palm tree (sugar palm starch) and the nanofiller SPNFCs was also isolated from sugar palm fibre via taken from the same tree (*Arenga pinnata*). A truly environmental-friendly biocomposites are obtained from a single tree.

It is well acknowledged that biocomposites provide superb benefits to human mankind. Nevertheless, there are some problems associated with natural polymer matrices, which are associated with moisture absorption due to the abundance of hydroxyl groups in their structure. The moisture that was built-up in biocomposites can lead to physical degradation such as thickness swelling and dimensional changes, as well as lowering the mechanical properties of film. The thickness swelling can lead to reduction in the adhesion between the fibres and matrix. As a consequence, the moisture absorption test is generally performed for quality control purposes of bionanocomposite materials. In the present study, nanocomposite materials were prepared from sugar palm nanofibrillated cellulose obtained from sugar palm fibre and a sugar palm starch via solution casting method. The goal of this work is to study the effect of nanocellulose size on the water absorption properties and the water solubility behaviour of thermoplastic starch. Sugar palm starch



was used as a matrix with water, sorbitol and glycerol as a plasticizer and with SPNFCs-5 cycles, SPNFCs-10 cycles, and SPNFCs-15 cycles, as reinforcements.

2. Experimental

2.1 Materials

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

2.2 Preparation of SPNFCs

Sugar palm fibres (SPF) were collected from different part of sugar palm trees (sugar palm frond, trunks, ijuk and bunch), as it wrapped around the tree trunk from top to bottom. A knife was used to remove SPF from the tree. Then, a Fritsch pulverisette mill was used for grinding and screening to gain an even size of SPF particles, about an average of 2 mm. The cellulose preparation procedures were reported elsewhere [26]. Briefly, the SPF were extracted using two main processes, known as delignification and mercerization. The initial and second processes 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). SPNFCs were prepared via mechanical treatment. Sugar palm cellulose (SPC) was refined for 20,000 revolutions in a PFI-mill according to ISO 5264-2:2002. Then, the fibres were isolated by the process of high pressurized homogenization (HPH). Typically, 1.8 % fibre suspension in water was processed in a high pressurized homogenizer (GEA Niro Soavi, Panda NS1001L, Parma, Italy) at 500 bars. Samples were passed with various cycle of 5, 10 and 15 times through an intensifier pump. The nanofibre with different cycles of 5, 10 and 15 times were denoted as SPS/SPNFCs-5C, SPS/SPNFCs-10C, and SPS/SPNFCs-15C, respectively. Through this process, fibres were broken down from macro-sized structure to nanosized structures, forming slurries of nanofibrillated cellulose. The NFCs suspensions were then collected and were freeze-dried at -110 °C using ethylene gas and stored at cool place prior to sample analysis and nanocomposite films preparation.

2.3 Preparation of the SPS/Spnfcs Nanocomposite Films

The fabrication of plasticizer sugar palm starch/ sugar palm nanofibrillated celluloses (SPS/SPNFCs) composite film was based on a solution casting methods. In order to obtain homogenous nanocomposite film, starch, sorbitol, glycerol, SPNFCs and distilled water were mixed and sonicated together [26]. Briefly, a solution of SPNFCs was prepared by mixing and sonicating it with 190 mL of distilled water with known concentration of SPNFCs (0.5 wt. % on the starch basis). The SPNFCs content was fixed at 0.5 wt. % based on the starch basis. Then, 10 g of SPS and plasticizer (30% on the starch basis) were mixed with the SPNFCs 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 was 1:1 with combination of sorbitol and glycerol ratio. Then, the film-forming suspension was left to cool down and was placed under vacuum to remove air bubbles inside the suspension prior to casting of the suspension by putting 45 g of the suspension into petri dishes sized of 15-cm diameter. The petri dishes containing the film-forming solution were placed in an oven at 40°C overnight. SPS films were prepared without SPNFCs served as the control (designed as SPS film), whereas the nanocomposite film with different cycles of 5, 10 and 15 times were denoted as SPS/SPNFCs-5C, SPS/SPNFCs-10C,



and SPS/SPNFCs-15C, respectively. The resulting films were kept in the desiccator at room temperature for a week to ensure the equilibrium of the water content in the films prior any characterization tests.

2.4 Transmission Electron Microscopy (TEM)

The structure and diameter measurement of sugar palm nanofibrillated cellulose (SPNFCs) were determined by Hitachi H-7100 transmission electron microscopy (Hitachi, Tokyo, Japan). The images were obtained from high-resolution transmission electron microscopy (HRTEM). Prior to the testing 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 the staining step was to get better visual and contrast image via TEM.

2.5 Field Emission Scanning Electron Microscopy (FESEM)

Surface characteristics of the SPS and SPS/SPNFCs 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 coated with gold to avoid charging using an argon plasma metallizer (sputter coater K575X) (Edwards Limited, Crawley, United Kingdom) [27].

2.6 Water Absorption

Water absorption analysis was carried out in accordance to ASTM D 570-98. Firstly, the samples were dried for 24 h at 50 °C and cooled in desiccator to gain 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 (1) [7].

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

2.7 Water Solubility

Three samples from each formulation of the nanocomposite films (30 mm × 10 mm) were dried in an vacuum oven 105 $^{\circ}$ C for 24 h, following the method of Shojaee *et al.*, [28]. The samples were weighed to determine the initial dry matter of each film (W_i). Each sample was immersed in 50 mL of distilled water in a beaker that was sealed and periodically stirred under constant agitation at 23 ± 2 $^{\circ}$ C for 6 h. Finally, the insoluble portion of the film sample was removed, oven dried at 105 $^{\circ}$ C for 24 h and weighed to determine the weight of the solubilized dry matter (W_o). Water solubility of each film was determined by Equation (2).

Solubility (%) =
$$\left[\frac{W_i - W_o}{W_i}\right] \times 100$$
 (2)



3. Experimental Result and Discussion

3.1 Physical Properties of Sugar Palm Nanofibrillated Cellulose (SPNFCs)

The physical properties of the sugar palm nanofibrillated cellulose (SPNFCs) from different cycle of High Pressurized Homogenizer (HPH) treatment were determined using Image J software, Origin 7.5 software and a Gaussian line shape to the peaks of the images collected from the TEM analysis.

Table 1 and Figure 1 show the physical properties of nanofibrillated cellulose and TEM micrograph, respectively. TEM micrograph of sugar palm nanofibrillated cellulose (SPNFCs) (Figure 1) revealed their nanometric dimensions. The diameter of the SPNFCs-5C, SPNFCs-10C and SPNFCs-15C were approximately 21.37 ± 6.91 nm, 11.54 ± 2.77 nm, and 5.5 ± 0.99 nm, respectively. The diameter analyzed for SPNFCs-15C were similar to the nano-structures that were isolated from other agrowaste sources such as such as banana (5 nm) [34], flax fibre (5 nm) [6], potato tuber cell (5 nm) [3], and wider than prickly pear fruit (2-5 nm) [9]. Besides that, SPNFCs-10C have similar diameter with cassava bagasse (2-11 nm) [30] and raw cotton linter (*Gossypium hirsutum*) (12 nm) [22]. Whereas, the SPNFCs-5C have similar diameter with pineapple leaf (30 nm) [4], sugarcane bagasse (35 nm) [20], wheat straw (30-70 nm) [15], rutabaga (80 nm) [6]. These resultant images revealed the use of high pressurized homogenization treatment on sugar palm fibres and certified that the aqueous suspensions contained sugar palm NFCs residing mostly of individual and some aggregates.

Table 1

Physical Properties of SPNFCs-5C,

SPNFCs-10C and SPNFCs-15C		
Fibres	Diameter (nm)	
Sugar Palm Cellulose (SPC)	11870	
SPNFCs-5C	21.37 ± 6.91	
SPNFCs-10C	11.54 ± 2.77	
SPNFCs-15C	5.5 ± 0.99	



Fig. 1. TEM micrographs of the (a) SPNFCs-5C, (b) SPNFCs-10C and (c) SPNFCs-15C



Figure 2 shows the diameter of SPNFCs-5C, SPNFCs-10C and SPNFCs-15C. The differences between the SPNFCs-5C, SPNFCs-10C and SPNFCs-15C were the reduction of diameter [35]. Based on Table 1, the reduction of the SPNFCs-5C diameter from SPC was 99.82%. This was attributed by breaking the interfibrillar hydrogen bond between nanofibril via high pressurize homogenizer treatment on SPC. The process continued for SPNFCs-10C and SPNFCs-15C for 10 cycles and 15 cycles, indicated the changes in the size of the SPNFCs which was reduced by 46.00 % and 74.26 %, respectively compared to SPNFCs-5C. This was also caused by the longer reaction HPH cycling via mechanical treatment on the fibres, which loosening the interfibrillar hydrogen bonding within nanofibres. Besides, longer cycling time could reduce the structure (diameter) of SPNFCs [36]. Thus, the longer the cycling times of HPH, the smaller the diameter of the nanofibres produced. Thus, in order to defibrillate the sugar palm nanofibrils, the interfibrillar hydrogen bond energy should be beaten separately rather than to cut them. The differences of the diameter obtained was depending notably on the homogenization pressure, number of passes, sample pretreatment as well as on the fibres origin [2,5,37].



Fig. 2. Length and diameter of SPNFCs-5C, SPNFCs-10C and SPNFCs-15C

3.2 Surface Morphology of Bionanocomposite Film

Figure 3 showed the surface morphology of SPS-based films reinforced SPNFCs by using FESEM. The Figure 3 (a) micrograph of the SPS/SPNFCs films showed a smooth and continuous surface with no trace of starch granular or cracks and agglomerations of SPNFCs. Besides that, high dispersion of SPNFCs (Figure 3(c)) is a good sign of strong interfacial adhesion between the two components of the SPS/SPNFCs nanocomposite film. These good distributions translate into high resistance of water barrier properties, which can be indicated through the water absorption and water solubility test. Moreover, the functional properties of the composite was enhanced when the fibres well-dispersed into the polymeric matrix.





Fig. 3. Surface morphology of SPF/SPNFCs bionanocomposite (a) surface, (b) cross-sectional (100 μ m), and (c) cross-sectional (2 μ m)

3.3 Water Absorption

One of the most crucial study in biofilm is water absorption which indirectly attributed to the properties of water that would acts as a plasticizer in biofilm. Figure 4 revealed the percentage of water absorption of SPNFCs-5C, SPNFCs-10C and SPNFCs-15C nanocomposite films. In Figure 4(a), it can be seen that the neat SPS gave the highest percentage of water absorption, followed by SPS/SPNFCs-5C, SPS/SPNFCs-10C, and SPS/SPNFCs-15C, with value of 119.79%, 100.72%, 99.18% and 97.23%, respectively. This phenomenon showed that biofilm reinforced with SPNFCs has lower hydrophilicity. The water absorption of the neat SPS biofilm was higher than reinforced SPNFCs that was due to the presence of hydroxyl groups in film molecules. Film with higher concentration of starch matrix tend to absorb more water compared to biofilm that have low concentration of starch [24]. Moreover, as the diameter size of SPNFCs reduced via HPH cycling process, the percentage of the water absorption also decreased. Film with big particle size of SPNFCs exhibited greater tendency to absorb water, which possibly due to a lower response to stress [7,40]. In this work, water absorption tests revealed that stabilizing the mixtures with SPNFCs decreased the water absorption capacity. This was attributed to the strong adhesion between the SPNFCs and SPS in which they tend to form strong hydrogen bonding via the physical interlocking with non-polar matrices and the OHgroups of hydrophilic polymer matrices [18,21]. The result of this experiment conducted are quite similar to those from previous work [24].





Fig. 4. (a) Water absorption and (b) water uptake at equilibrium (■) and diffusion coefficient (★) of the SPS and SPS/SPNFCs nanocomposite film as function of SPNFCs content.

Besides that, the weight gain because of water absorption of all bionanocomposite films were linearly increased with the increase of immersion time and became more constant after 1 h, following a Fickian diffusion process. Based on the Figure 4 (a) also, the water uptake-*t* curves show two well-separated zones, in which at shorter times, t < 1 h, the kinetic of absorption is very fast, however at extended times, t > 1 h, the kinetic of absorption is very slow and lead to a flat line, equivalent to the water uptake equilibrium. Generally, water absorption is influenced by fibre orientation, permeability, surface protection, exposed surface area, diffusivity, temperature, and fibre content [33,39,41]. However, in this work, fibre content and temperature were kept constant, whereas the SPNFCs nanofibres orientation in the SPS biofilm was assumed to be identical due to the same processing parameters.



The water uptake at equilibrium of neat SPS biofilm and SPS/SPNFCs nanocomposite films as a function of SPNFCs cycling process was plotted in Figure 4(b). From the Figure stated, it can be seen that neat SPS film absorbed about 119.79 wt. %. It can be simply explained that 1 g of starch could absorbed 2.198 g of water. The water uptake equilibrium decreased non-lineally with increment of SPNFCs nanofibres HPH cycle, and about 97.23 wt. % of water uptake was observed for the SPS/SPNFCs-15C nanocomposites film. Compared with neat SPS, the nanocomposite film showed a lowered swelling capacity. The kinetic study of the sorption in polymers as a mean of resolving the diffusion coefficient has been widely used. The partial differential equation for mass transfer (diffusion) is expressed as [32].

$$\frac{\partial C}{\partial t} = D(\frac{\partial^2 C}{\partial x^2}) \tag{3}$$

Where, C is the concentration at time *t*, and distances *x*, and distance *x*, from the polymer surface. For an infinite slab with a constant *D* and at short times.

$$\frac{M_t - M_0}{M_\infty} = \frac{4}{L} \left[\frac{Dt}{\pi} \right]^{1/2} \tag{4}$$

Where, M_t and M_0 are the weight of the sample at time t and the initial weight of the sample, respectively. M_{∞} is the equilibrium water sorption, and L is the thickness of the sample.

The ratio of the amount of water absorbed by the specimen at time t, $(M_t - M_0)/M_{\infty}$, was plotted as a function of $t^{1/2}$, and the diffusion coefficient (D) was, therefore, calculated from the slope of the initial linear part of the resulting curves. As shown in Fig. 4(b), the neat SPS film showed the highest D value of about $1.586 \times 10^{-8} cm^{-10} s^{-1}$. The neat SPS was observed to have higher D value compared to nanocomposite films, in which this phenomena could be attributed to the hydrophilic behavior of SPS, in which chemical properties of starch itself is composed of multi-hydroxyl polymer with three hydroxyl groups per monomer. Interestingly, with an increase of nanofillers cycle in the SPS biopolymer from 5 cycles to 15 cycles, the D value of the nanocomposites decreases from $1.586 \times 10^{-8} cm^{-10} s^{-1}$ to $1.442 \times 10^{-8} cm^{-10} s^{-1}$. This spectacle occurred due to the high surface area and abundant hydroxyl group of SPNFCs which indirectly caused strong adhesion between the nanofiller/ matrix adhesion (SPNFCs and SPS). As a result, the water becomes harder to penetrate and stored within the starch polymer matrix. Therefore, increase in the SPNFCs concentration, decreased the hydrophilic of SPS/SPNFCs nanocomposite films. Moreover, these strong hydrogen bonding interactions within the nanocomposite films tend to stabilize the starch matrix when it is exposed to highly moist atmosphere. High crystallization of SPNFCs concentration in the nanocomposite also might be attributed to the decrement of the diffusion coefficient and water uptake at equilibrium of the biofilms [16]. Therefore, the increase of HPH cycling of SPNFCs nanofiller, would decrease the SPNFCs diameter size, which indirectly resulted in low water uptake when reinforced with thermoplastic starch. This summarized that the biofilms reinforced SPNFCs are less water sensitive that neat SPS.

Besides that, the interface between glycerol plasticizer and starch matrix could promote moisture adsorption because of the dimensional changes in the area where the adhesion between interfaces (glycerol-starch) is poor, hence forming a path that speed up the water molecules accumulation. Interestingly, due to the chemical similarity between SPNFCs and SPS, structural and interface defects was reduced dramatically, hence forming a strong resistance for water molecules to pass through or stored in the film. It can be concluded that the nano-reinforcement process that arise within the structure of polymer matrix had influenced the permeation process that depends mainly on the



formation of the tortuous path by SPNFCs and diffusion coefficient, whereas at high moisture contents still produces a barrier effect on the movement of water molecule [29].

3.4 Film Solubility in Water

One of the main property of films for food packaging applications is film solubility. Several potential uses may require water insolubility to improve water resistance and product integrity for storage of food safety. The problems arouse in using starch as film packaging is the water resistance properties, explained by water solubility, in which the film has high water solubility. Thus, the reinforcement of SPNFCs within SPS matrix polymer is predicted to overcome this limitation. The film solubility of neat SPS and SPS/SPNFCs nanocomposite films are displayed in Figure 5. It can be seen that all biofilms did not remain intact once they are immersed in water.



Fig. 5. Water solubility of the SPS and SPS/SPNFCs nanocomposite films.

The water solubility behavior depends on the diameter size of nanofiller, in which the more the HPH cycle, the smaller the SPNFCs diameter. The water solubility decreased with the diameter of SPNFCs. The addition of SPNFCs reduced the films solubility from 33.36 % to 20.22 %. The SPS/ SPNFCs-15C showed the lowest film solubility value of 20.22%, which proved that the films have good water stability. Besides, it was also an indication of strong interactions between nanofiller-SPNFC and SPS starch chains within the biofilm matrix. The terrific ability of SPNFCs in interacting with starch chains were attributed to the interaction of abundance of hydroxyl groups of SPNFCs and starch hydrogen bond. Hence, these interactions provide resistance and stability to SPS/SPNFCs films. Moreover, according to Tunc et al., these interfaces enhanced the cohesive properties of the biopolymer matrix and reduced the sensitivity of water due to water molecules were not able to break these strong bonds [31]. Rhim and Ng in different report, reported that the water solubility resistance were improved when it was reinforced with nanofillers, in which this was attributed to the strong formation of hydrogen bond between hydroxyl groups of the biopolymer and nanofiller [23]. These results were also supported by Abdollahi et al., in their research project on alginate nanocomposite films [1]. It can be summarized that the reinforcement of SPNFCs improved the water resistance properties of the films which was a relevant characteristic for food packaging materials to improve shelf-life of food products [38].



4. Conclusion

Sugar palm fibre (*Arenga pinnata*) were successfully individualized via the method of delignification, mercerization and high pressurize homogenizer process under a shear action to produce nanofibrillated cellulose suspensions. Individualized nanofibrillated cellulose possessed 'spaghetti-like' form with high aspect ratio fibres almost 5-20 nm in diameter. Increasing HPH cycling process would decrease the SPNFCs diameter. The resulting suspension was utilized to process bionanocomposite with combination of both matric and natural fibres that are obtained from less utilized parts of sugar palm tree. It was observed that the water absorption effect on the SPS and SPS/SPNFCs bionanocomposite had brought a significant improvement when the size of the SPNFCs were reduced. Whereas, the water solubility test revealed that the SPS soluble faster as compared to SPS/SPNFCs bionanocomposites. Thus, this bionanocomposite shows improvement in water absorption and water solubility properties for packaging materials application.

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