Research Article

Oil-palm Based Nanocellulose Reinforced Thermoplastic Polyurethane for Plastic Encapsulation of Biomedical Sensor Devices: Water Absorption, Thickness Swelling and Density Properties

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Abstract
Oil palm nanocellulose has been demonstrated to display a wide range of unique properties for many fields. They are suitable for biomedical applications and have been used in this domain for decades. The current variety of nanocellulose fibers allows the development of new nanocomposites. This work fabricated oil palm nanocellulose with variations of fiber loading (1, 2, 3, 4, and 5 wt%) and thermoplastic polyurethane (TPU) polymer matrix by using a mechanical stirring followed by hot pressing methods. The physical characters of nanocellulose oil palm reinforced TPU nanocomposites, such as water absorption, thickness swelling, and density were characterized. The fiber loading of oil palm nanocellulose content at 5 wt% shows the highest water uptake, thickness swelling, and the lowest density properties of oil palm nanocellulose reinforced TPU nanocomposites.

Keywords: Oil-palm nanocellulose, Thermoplastic polyurethane nanocomposites, Plastic encapsulation, Physical properties dimensional stability

1 Introduction

The existence of polymer encapsulation of biomedical sensors, such as polyethylene naphthalene (PEN) [1], polydimethylsiloxane (PDMS) [2], [3], polyethylene terephthalate (PET) [4], epoxy [5] shows that this polymer material was demanded for encapsulation of biomedical fields, mainly for the sensor. For instance, the sensor that used plastic encapsulation, such as temperature sensor, humidity sensor, pressure sensor, flow sensor, acoustic sensor, etc. Many scientists and researchers used developed plastic in flexible electronics [6], [7]. Since some of the sensors do not contain metal, it can be made by the low cost plastic to diagnose or monitor a wide range of health conditions, including surgical complications.

The plastic encapsulation can be incorporated with other nanofillers, either inorganic or organic materials [8]. The inorganic or synthetic materials can be harmful and higher cost, while organic or natural fillers can be substantial alternatives to inorganic materials that are sustainable and environmentally friendly [8]–[11]. Incorporating natural fillers, such as nanocellulose oil palm demonstated the potential of nanocellulose from plant-based [12]–[16]. The oil palm nanocellulose improved mechanical properties of nanocomposites and has been observed using different polymer matrices, such as PVA [13] and thermoplastic starch/PVA [17]. It has been reported that thermoplastic polyurethane (TPU) is being used in some important applications, such as automotive components and medical equipments. This is due to the material's excellent mechanical properties, transparency, and resistance to oil, grease, and abrasion, among other characteristics. TPU is an elastomer composed of linear chains that are separated into hard and soft segments of the copolymer. The combinations between TPU with other natural fibers, such as kenaf, sugar palm, coir were reported in microcellulose size and very few works reported with the nanocellulose size, mainly with oil palm nanocellulose fiber [18], [19].

The use of lower water-resistance of nanocellulose has limited its application. A study by Amri *et al.* [20] conducted a research in which they investigated the water absorption of cellulose nanofibrils (CNF)/ Jatropha oil-based waterborne polyurethane (WBPU) nanocomposite films by using the epoxidation and ring-opening methods. The contact angle reduces as the CNF concentration rises, however, this has only a slight effect on the film's water absorption (around 3.5%). Even after being immersed in water for 5 days, the difference between the neat WPBU and the maximal CNF loading film was less than 1%. In another research, Kępa *et al.* [21] examined cellulose nanofibres (CNF) produced from spinifex grass/epoxidized soybean oil (ESO) via the process of nanopaper production. According to this work, the water absorption of the nanocomposites may reduce the amount of absorbed water vapor by up to 53%. The effects of ZnO/cellulose nanocrystals (CNCs) inclusion on the structure–property relationship of acrylonitrile-butadiene rubber were investigated (in NBR films) by Ogunsona *et al.* [22]. It was discovered that the inclusion of the 0.5 phr CNC significantly decreased the water absorption of the plain CNC by 250%. Water absorption of the nanocomposite films was much lower than that of the plain NBR due to CNC consolidation of the rubber particles, which reduced free volume in the NBR structure, resulting in significant reduction of water absorption. De Souza *et al.* [23] fabricated nanocellulose (NC) / poly(butylene adipate-co-terephthalate) (PBAT) by using the melting process and evaluated the water uptake properties. The water absorption results showed that the pegylated-NC has a low propensity to absorb water, which increases its usability as a packaging material. Kahavita *et al.* [24] developed a nanofibrillated cellulose (CNF) reinforced polypropylene composite that water uptake was carried out in their research. According to the research findings, the inclusion of both treated and untreated CNF slightly raised water absorption, while simultaneously decreasd the processability of the final product.

Furthermore, Ben Shalom *et al.* [25] fabricated cellulose nanocrystals (CNC)/polyvinyl alcohol (PVA) composite films with and without 1,2,3,4-butane tetracarboxylic acid (BTCA) using the conventional solution casting technique. They reported that the water absorption of crosslinked CNC and CNC-PVA nanocomposite films is substantially decreased, while the transparency of the films is significantly enhanced as a function of the PVA and crosslinker concentration. Besides, Zhang *et al.* [26] investigated nanocrystalline cellulose (NCC) and polyethylene glycol (PEG) on the hydrolytic degradation behavior of poly(lactic acid) (PLA). As a result of the presence of hydrophilic NCC and PEG in the composites, the hydrolytic degradation of PLA was significantly accelerated. This was attributed to the rapid dissolution of PEG, which allowed easy access of water molecules to the composites and resulted in the initiation of rapid hydrolytic chain scission.

The limitation of the utilization of this nanocellulose is that the hydrophilic structure may impair the potential of plant-based nanocellulose as nanofiller in plastic encapsulation [27], [28]. Therefore, the importance of studying dimensional stability is a must for the plastic encapsulation of nanocomposites. There is still a lack in the literature on the oil palm nanocellulose reinforced TPU regarding water uptake, thickness swelling, and density properties. This work introduces the study on the dimensional stability of nanocellulose oil palm reinforced TPU. The changes of physical properties with the variation of oil palm nanocellulose (1, 2, 3, 4 and 5 wt%) were observed for 216 h. Table 1 represents the previous work working on dimensional stability properties of nanocellulose composites

2 Materials and Methods

2.1 *Materials*

The materials used for this study are oil palm-based (NOP) nanocellulose suspension, Thermoplastic Polyurethane (TPU), Grade 58311 from Lubrizol, Belgium, in crystal pellets form were purchased through ZoepNano Sdn. Bhd., Serdang, Selangor and Innovative Pultrusion Sdn. Bhd was obtained from Seremban, Negeri Sembilan, Malaysia. The details of TPU were shown in Table 2.

Table 1: Previous works on water absorption, thickness swelling and density properties of nanocellulose composites

Type of Nanocelluose Fiber	Type of Matrix	Immersion Media	Immersion Time	Water Absorption (WA)	Thikcness Swelling (TS)	Density	Ref.
(0.1–0.5 wt%) Cellulose -nofibrils (CNF)	Jatropha oil-based waterborne polyurethane (WBPU)	Deionized water	5 days	Higher content of CNF, the WA <3.5%	NA	NA	[20]
Cellulose nanofibres (CNF) derived from spinifex grass	Epoxidized soybean oil (ESO)	Humidity chmaber	48 h	Reduce WA up to 35%	NA	NA	[21]
(1–5 wt%) Nanocellulose (NC)	Poly(butylene adipate-co-terephthalate) (PBAT	Water	50 h	Higher content of NC, the WA <3%	NA	NA	[23]
Nanofibrillated cellulose (CNF)	Polypropylene	Water	24 h	The silane treated & untreated CNF slightly increased WA	NA	NA	[24]
Nanocellulose fibers from sugarcane bagasse	Epoxy	Water	-	The alkaline treated reduced ~10% WA	NA	NA	[29]
Carboxymethylcellulose (CMC) & nanofibrillated cellulose (CNF) from palmito sheaths pulp	Chitosan	Water	24 h	The addition of 1.5% of CNF resulted a decrease of almost 50% on water absorption	NA	NA	[30]
(0–0.1 wt%) Sugar palm nanofibrillated cellulose (SPNFC)	Sugar palm starch	Water	24 h	1 wt% SPNFCs loading significantly improved the water absorption and water solubility of the composite film by 24.13% and 18.60%, respectively, compared with the control SPS film.	NA	NA	[31]
Silylated cellulose nanocrystal (SCNCs)	waterborne polyurethane (WPU)	Deinoinzed water	70 h	hydrophobicity of the material were improved simultaneously, reaching the percolation threshold at a 0.50 wt % SCNCs as determined theoretically	NA	NA	[32]

Table 2: Properties of Thermoplastic Polyurethane (TPU)

Mechanical Properties	Unit	Value
Hardness	Shore A/D	87/-
Density	g/cm^3	1.12
Tensile strength	MPa	42
Tensile Stress Elongation Tensile stress- 50% Elongation 100% Elongation 300% Elongation	MPA % MPa MPa MPa	42 655 4.9 5.9 9.1
Compression Set (*) 70 h at 22 °C 24 h at 70 °C	 % %	 27 70
Tear Resistance	kN/m	54
Abrasion loss	mm^3	34
Rebound resilience	%	40
Brittle point	°C	–70

2.1.1 Isolation of oil palm (NOP) nanocellulose fiber

The isolation of nanocellulose of oil palm (NOP) fiber was adopted from Ilyas *et al.* [31]. In order to improve fiber accessibility and processing efficiency, a refining treatment prior to the high pressure homogenization (HPH) process was needed. To achieve this, the oil palm cellulose was refined in a beater (PFI-mill, DSG-2000, Regmed, Brazil) for a total of 20,000 revolutions, in accordance with ISO 5264-2:2002. The fibers were subjected to the refining process in order to improve both exterior and interior fibrillations, which resulted in the better fiber flow and the avoidance of clogging during the fluidization procedure. HPH was used to separate NFC from cellulose in oil palm fibers. Typically, 1.8 wt% of the fiber suspension was processed in a high-pressure homogenizer to get the desired result (GEA Niro Soavi, Panda NS1001L, Parma, Italy). A total of 15 passes through an intensifier pump, which increased the pump pressure, were made before passing through the interaction chamber. This is due to the defibrillation of the fibers via shear pressures and impacts against the channel walls and colliding streams. During this process, macro-sized fibers were broken down into nano-sized particles, resulting in the formation of slurries of nanofiber composite. The high-pressured homogenizer was kept operating at 500 bar and with a pH of 7.0. The temperature was not fixed at a certain value. Nonetheless, when the suspension temperature reached about 90 °C, the fluidization process was briefly halted to avoid pump cavitation. After the samples had cooled to about room temperature, the procedure was repeated at 45 °C.

2.1.2 Preparation of NOP/TPU nanocomposites

Firstly, TPU in pellets form was heated in an oven for 24 h at 60–65 °C in order to prevent bubble formation during the mixing. Afterward, five sets of 15 g TPU were weighted and prepared five different samples. Each sample contained different amounts of NCF and was labeled as 1NOP/TPU, 2NOP/TPU, 3NOP/TPU, 4NOP/TPU, and 5NOP/TPU as shown in Table 3. Sample 1NOP/TPU was prepared by placing 15 g of TPU on isotemp hot plate stirrer at low temperature. As the TPU pellets started to melt, 14.85 g of NCF were added, stirred well, and then quickly poured into an Aluminium mold sheet with size of 2 cm × 2 cm × 0.2 cm. The Teflon papers were used to make it easier during demolding process. Lastly, mold was placed inside the oven for 10 min, around 170 °C, then was cooled under room temperature for another 10 min. Finally, the sample was demolded and cut into three sets of 1 cm × 1cm × 0.2 cm sizing. The steps were repeated for 4 samples, including 2NOP/TPU, 3NOP/TPU, 4NOP/TPU, and 5NOP/TPU. The formulation of oil palm nanocellulose /TPU is shown in Table 3.

Table 3: Formulation of oil palm nanocellulose /TPU

Sample Designation	TPU (wt%)	NOP (wt%)	Mass of TPU (g)	Mass of NOP (g)
1NOP/TPU	99	1	14.85	0.15
2NOP/TPU	98	2	14.70	0.30
3NOP/TPU	97	3	14.55	0.45
4NOP/TPU	96	4	14.40	0.60
5NOP/TPU	95	5	14.25	0.75
*NOP= Nanocellulose oil palm, TPU= Thermoplastic Polyurethane				

3 Characterization

3.1 *Water absorption testing*

The properties of nanocomposites were studied by measuring the water absorption rate according to ASTM 570-98 [14]. The experiment was conducted by immersing three samples for different NOP contents (1, 2, 3, 4, and 5 wt%) in distilled water. The initial weight of the samples was recorded as in 0 h and the next 24 h of the immersion period marked as final weight, the value of was taken as final thickness for 24 h, 48 h, 72 h, 96 h, 120 h, 144 h, 168 h, 192 h, and 216 h. The formulation was used to identify the rate of water absorption test as in Equation (1).

$$\text{Water absorption (\%)} = (W_f - W_i)/W_i) \times 100 \quad (1)$$

where, W_f = Final weight and W_i = Initial weight of NOP/TPU

3.2 *Thickness swelling testing*

The test was carried out in accordance with ASTM D 570 standards by immersing the sample in distilled water, and the physical changes could be observed in thickness swelling. The initial thickness of the sample was measured using vernier caliper and recorded as 0 h. For the next 24 h, the value was taken as final thickness for 9 days (216 h). Following Equation (2) was used as the formulation to calculate the percentage of thickness swelling.

$$\text{Thickness swelling (\%)} =(T_f - T_i/T_i) \times 100 \quad (2)$$

where T_f = Final thickness and T_i = Initial thickness of NOP/TPU

3.3 *Density properties*

The test was carried out in accordance with ASTM D4018 standards, and the data were measured using a weighted scale densitometer, MD-200S Mirage (Japan) and a Mitutoyo digital vernier calliper, respectively. The density was calculated using density formula as shown in Equation (3):

$$\text{Density, } \rho = m/v \quad (3)$$

where ρ, = Density (g/cm^3), m = Mass (g), v = Average volume (cm^3)

4 Results and Discussion

4.1 *Water absorption properties*

Water absorption of nanocellulose oil palm reinforced thermoplastic polyurethane nanocomposite versus time was presented in Figure 1. Sample with 5 wt%

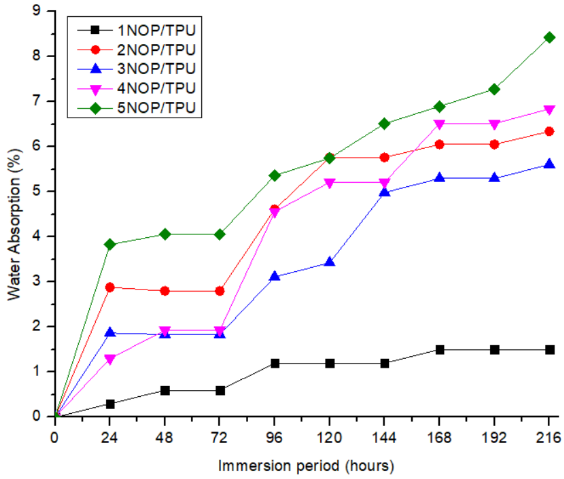

Figure 1: Water absorption of nanocellulose oil palm/ TPU.

NOP had the highest water absorption due to its hydrophilic properties. The increase of water absorption is due to the increase of NOP content in 5NOP/TPU, 4NOP/TPU, 2NOP/TPU, 3NOP/TPU, and 1NOP/ TPU. As the immersion time increased from 0 to 216 h, the mass and water absorption were increased. The percentage of water absorption does not exceed 10%, showing that incorporating TPU reduced the water uptake compared to other nanocellulose composites at 60–90% [33]. Incorporating the TPU polymer matrix reduced the degree of swelling in the nanocellulose oil palm nanocomposites.

4.2 *Thickness swelling properties*

Figure 2 provides a summary of obtained results of thickness swelling analysis. It was found that the NOP/TPU nanocomposites exhibited an increase in thickness swelling when immersed in distilled water for 216 h. The reduced water uptake and swelling of the incorporation of TPU are due to hydrophobic properties, which is water-resistant polymers are materials that are not soluble in water or other polar solvents. When the TPU does not absorb the water, the less water uptake and less swelling properties of the polymer. Besides, the lower density of TPU polymer matrix compared to nanocellulose oil palm at the value of (TPU:1.07 g/cm^3) also contributes to the lower water uptake and thickness swelling properties [34], [35].

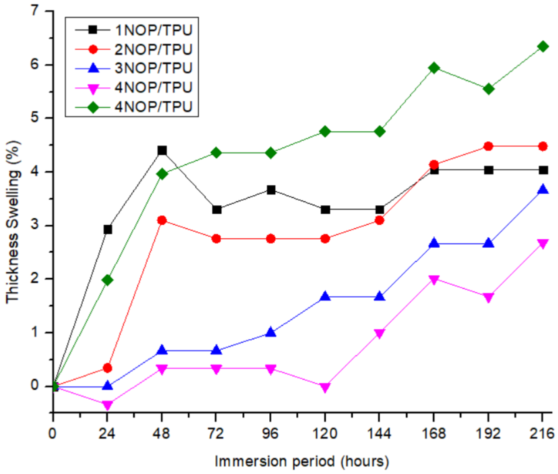

Figure 2: Thickness swelling properties of nanocellulose oil palm/TPU.

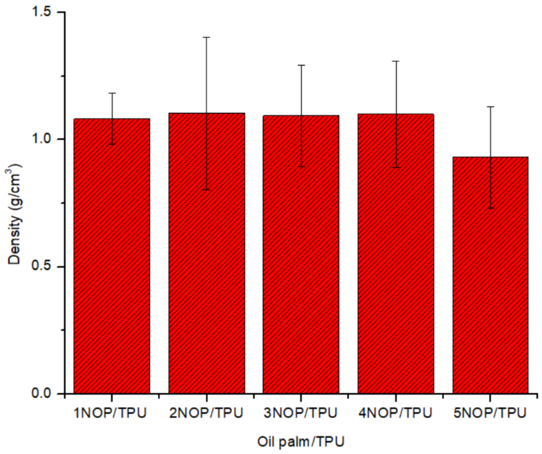

Figure 3: Density properties of nanocellulose oil palm/TPU.

However, the nanocellulose oil palm content may change the physical properties of nanocomposites even the higher addition of 5 wt% of NOP. Oil palm nanocellulose exhibited higher hydrophilic properties, which is poor resistance to swelling conditions. The improper specimen preparation could be one reason for the inconsistency in thickness swelling of 4 wt% NOP that has lower thickness properties than the other samples. Nevertheless, the range of thickness swelling percentage was still in control less than 10%, which aligned with the moisture absorption result.

4.3 *Density properties*

The density properties of nanocellulose oil palm/TPU with variations of 1, 2, 3, 4, and 5 wt% of nanocellulose oil palm are presented in Figure 3. The higher content of nanocellulose oil palm exhibited lower density compared to other formulations. The nanocellulose oil palm is known as low density compared to the TPU polymer matrix. Overall, the highest density of the five formulations was 1NOP/TPU followed by 2NOP/TPU, 4NOP/TPU, and 3NOP/TPU. It was found that the incorporation of 5 wt% NOP led to the reduction of nanocomposites density. The similar findings agreed with the higher nanofiller content, such as nanocellulose sugar palm [33].

5 Conclusions

The investigation of dimensional stability properties of nanocellulose oil palm reinforced TPU were assessed under water absorption, thickness swelling and density testing. The physical properties show that adding the nanocellulose oil palm from 1% until 5 wt% exhibited increase water absorption and sample's density. The analysis of physical properties show that adding the nanocellulose oil palm from 1% until 5 wt% exhibited increase water absorption and thickness swelling, but reduced density properties. Their unique properties can be proposed as the plastic encapsulation for biomedical sensor devices. This material can be alternative material to be encapsulated, such as in wearable sensor devices that used plastic encapsulation to protect the electronic components.

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