

Preparation and Characterisation of Electrospun Composite Nanofibre Polyvinyl Alcohol/ Nanofibrillated Cellulose Isolated from Oil Palm Empty Fruit Bunches

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Composite nanofibre polyvinyl alcohol (PVOH) was prepared in this work reinforced with nanofibrillated cellulose (NFC) using an electrospinning technique. NFC was isolated from oil palm empty fruit bunches (OPEFB) by steam explosion method followed by acid hydrolysis. A 12 wt% PVOH solution in distilled water was prepared under constant reflux at 80 °C for 5 h. Several concentrations of NFC were added to the polymer solution of 2.5, 5.0, and 7.5 wt%. The nano-dimension of NFC was analysed using transmission electron microscopy (TEM). The morphology of electrospun nanofibre was characterised using scanning electron microscopy (SEM). The morphology of PVOH nanofibre was smooth and uniform without beads. The presence of NFC in the PVOH nanofibre decreased the diameter. The morphology of PVOH/NFC nanofibre at a concentration of NFC 2.5% was fairly uniform with good quality. However, the preparation of PVOH/NFC nanofibre at concentrations of NFC 5.0% and 7.5% resulted in elliptical beads. The crystallinity of NFC and electrospinning nanofibre was investigated using X-ray diffraction (XRD). In addition, the thermal properties of the samples were analysed using thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC). The results showed that NFC improved the thermal stability of electrospun PVOH nanofibre.

Keywords: Oil palm empty fruit bunches; Nanofibrillated cellulose; Steam explosion; Electrospinning; Thermal stability

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INTRODUCTION

Electrospinning is a simple method to produce ultrafine nanofibre. The equipment required consists of a power supply, a syringe pump, and a collector (Haider *et al.* 2018). Electrospun nanofibre has excellent properties such as high specific surface area, high aspect ratio, and biomimetic potential; hence, it has broad applications with specific requirements such as scaffolds (Fomby *et al.* 2010), membranes (Ray *et al.* 2016), textile products (Mirjalili and Zohoori 2016), and sensors (Chinnappan *et al.* 2017). In the electrospinning process, an electrically charged polymer solution is ejected through a narrow syringe under a high voltage differential as a fine jet. Rapid evaporation occurs when the charged polymer jets are impinged on to a collector surface. In this process, the jet

elongates, and the solid fibres are formed on the surface of the collector. There are several parameters used in electrospinning that influence the process, including polymer and solution properties (viscosity, molar mass, surface tension, pH, and electrical conductivity), processing conditions (tip to collector distance, applied electric voltage, and feed rate), and ambient conditions (air flow, temperature, and humidity) (Gonçalves *et al.* 2017; Maftoonazad *et al.* 2019).

Polyvinyl alcohol (PVOH) is a hydrophilic and semi-crystalline polymer that is produced by hydrolysis of polyvinyl acetate. PVOH is one of the most commonly used polymers for electrospinning due to its excellent properties (good electro-spinnability, solubility in water, thermal stability, chemical resistance, processability, biodegradability, non-toxicity, and biocompatibility) (Seo *et al.* 2019). Therefore, it has a wide range of applications in tissue engineering, filtration, optics, protective clothing, membranes, drug release, *etc.* (Peresin *et al.* 2010). However, the application of PVOH is limited when it is applied in water because of its high hydrophilicity, low mechanical strength, and poor integrity. Some chemical and physical modification methods have been applied to PVOH spun-fibres to solve the problems, such as chemical cross-linking, post-treatment, blending, heating, and irradiation. However, the methods involve the use of toxic material, time-consuming preparation, and specific equipment (Sutka *et al.* 2015).

Currently, material from natural resources is being used as a reinforcement. Nanofibrillated cellulose (NFC), which is prepared from the most abundant biopolymer, is a hydrophilic material and has high mechanical properties of 80 to 143 GPa (Habibi 2014). The biodegradability, renewability, and superior mechanical properties of NFC has attracted attention as a potential reinforcement for PVOH-based fibres. Both polymers have a huge number of hydroxyl groups so that they have a satisfactory intermolecular interaction by hydrogen bonding (Braga *et al.* 2019). Furthermore, PVOH reinforced with NFC increases thermomechanical properties as well as reduces water absorption (Sutka *et al.* 2013). It has also been reported that the presence of NFC in a PVOH polymer matrix improves the tensile strength by 80% and the storage modulus by threefold (El Miri *et al.* 2015).

Oil palm empty fruit bunches (OPEFB) is a solid waste from oil palm plantations. This material consists of 65% cellulose, 29% lignin, and 28% hemicellulose. Potential applications of cellulose-based OPEFB have been reported, such as pulp production and fertiliser for oil palm plantation (Gea *et al.* 2020). However, the use of NFC isolated from OPEFB for reinforcing of PVOH composite nanofibre has been not reported.

The present study produced a composite nanofibre with PVOH-based fibres reinforced with NFC by electrospinning technique. The NFC from OPEFB was isolated by steam explosion method followed by acid hydrolysis. Several concentrations of NFC were added to PVOH polymer solution to see the effect in the electrospinning process. The morphology, thermal properties, crystallinity, and chemical structure of PVOH/NFC spun-fibres were investigated.

EXPERIMENTAL

Materials

The materials used in this experimental were PVOH powder ($M_w = 60,000$ g/mole fully hydrolysed), NaOH pellets ($\geq 98\%$), CH_3COOH glacial (100%), NaOCl (2.5%), H_2O_2 (30%), and HCl (37%), which were purchased from Merck (Darmstadt, Germany). The

PVOH was dried at 80 °C for 5 h in a vacuum oven before used. Distilled water was used as a solvent of PVOH. NFC was isolated from OPEFB by steam explosion and acid hydrolysis.

Isolation of Nanofibrillated Cellulose

The isolation of NFC was conducted by the steam explosion method. First, OPEFB fibre was prepared by grinding the samples into small pieces, which were immersed in 2% NaOH overnight. The residue obtained from the filtration of the solution was autoclaved at a pressure of 168.9 kPa at 130 °C for 4 h. To remove the lignin, the fibre was washed with distilled water. Afterward, the fibre was washed with 17.5% NaOH: 7.4% CH₃COOH: 1.75% NaOCl solution with ratio of 1:1:6. The fibre was bleached with 10% H₂O₂ and neutralised using distilled water. The sample was filtered, and the residue was hydrolysed with 10% HCl and ultrasonication for 3 h at room temperature. It was then neutralised with distilled water and stirred with high shear homogenizer for 15 min at 8,000 rpm at room temperature until the fibre was suspended. The suspension was filtered and was dried in a vacuum oven at 50 °C for 4 h. The result was coded as NFC.

Preparation of Polyvinyl Alcohol/ Nanofibrillated Cellulose Solution

A 12 wt% PVOH solution in distilled water was prepared, and NFC was added with a concentration of 2.5, 5.0, and 7.5 wt%. The polymer solution was refluxed for 2 h at 80 °C. The suspension was allowed to cool under the same vigorous stirring until reaching room temperature. After that, it was homogenized using ultrasonication for 5 min at room temperature. The resulting mixture was stored at 3 °C for no more than three days prior to use.

Electrospinning

The electrospinning set-up included a syringe pump, disposable 18-G syringes, and a high voltage unit (0 to 50 kV). PVOH solution and PVOH/NFC suspension were electrospun by horizontal electrospinning (basic series electrospinning unit Brand NLI, Nanolab Instruments Sdn Bhd, Malaysia) at room temperature with the following conditions: (i) a voltage of 17.5 kV; (ii) a polymer solution feed rate of 0.3 mL/hour; (iii) a needle tip-to-collector distance of 10 cm; and (iv) a speed of collector 110 to 112 rpm. The electrospun-fibre mat was collected on aluminium foil, and the collected spun-fibre was dried in a vacuum oven at 40 °C for 3 h to remove residual water from the fibre. The spun-fibres were stored in a desiccator containing silica gel.

Characterisation

A very dilute of NFC solution in distilled water was prepared for the imaging with transmission electron microscopy (TEM). A few drops of NFC were deposited on carbon-coated electron microscope grids and negatively stained with a drop of uranyl acetate. A tip of filter paper was used to remove water excess, and then the specimens were allowed to dry at room temperature. The grid was observed with a JEOL/EO JEM-1400 (Tokyo, Japan) operated at an accelerating voltage of 100 kV. The morphology of the samples was analysed using a scanning electron microscopy (SEM) Hitachi TM3030 (JEOL, Ltd., Tokyo, Japan). The sample was first coated with a thin layer of gold to reduce the charging during analysis. The functional groups present in the samples were studied using a Fourier-transform infrared (FTIR) spectrometer (Nicolet 380, Thermo Scientific, Boston, USA). The instrument was operated in a transmission-mode with a resolution of 4 cm⁻¹ and 50

scans. The crystallinity of NFC and electrospun fibres were studied using an X-ray diffraction (XRD) Bruker D8 advanced X-ray diffractometer (Bruker Optik GmbH, Ettlingen, Germany). The instrument was operated at 35 kV and 25 mA. Thermogravimetric analysis (TGA) was performed using a DTA/TG Exstar SII 7300 (Hitachi medical system, Tokyo, Japan). A 10 mg sample was placed in a clean platinum pan and closed tightly. The sample was analysed between 30 and 650 °C at a heating rate of 10 °C/min. The thermal properties of the spun-fibres were studied via a differential scanning calorimetry (DSC) X-DSC7000 (Hitachi medical system, Tokyo, Japan) in a range temperature from 30 to 250 °C using a heating rate of 10 °C/min.

RESULTS AND DISCUSSION

Morphological Characterisation of Nanofibrillated Cellulose

The TEM micrographs of the NFC dried at 80 °C for 5 h in a vacuum oven are shown in Fig. 1 with magnification of 200 nm and 500 nm. The NFC shows interconnected webs of tiny fibre with diameter approximately 50 nm and lengths of several micrometres. In addition, TEM micrographs shows that NFC looks like whisker. The same result was reported by other authors (Cherian *et al.* 2010; Gea *et al.* 2018). The appearance of the NFC due to hemicellulose, lignin, pectin and other substance was removed during chemical treatment.

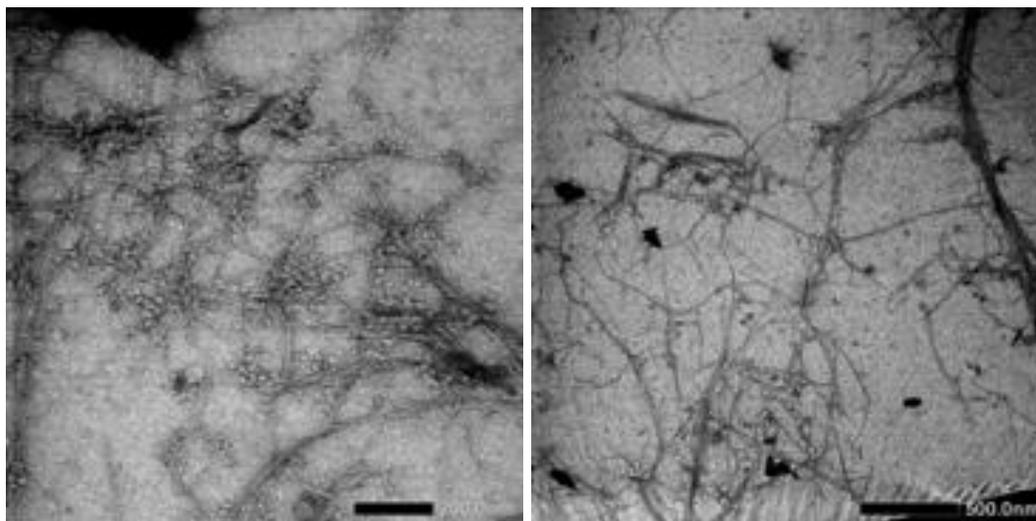


Fig. 1. TEM micrographs of NFC isolated from oil palm empty fruit bunches. The scale bar corresponds to 200 nm (left) and 500 nm (right)

Morphological Characterisation of Electrospun Composite Nanofibre Poly(Vinyl Alcohol)/ Nanofibrillated Cellulose

Figure 2 shows the morphology of the electrospun composite nanofibre PVOH/NFC with various concentrations of NFC, as observed by SEM. The morphology of electrospun PVOH nanofibre was smooth and uniform without any beads (Fig. 2 a,b). The average diameter of the nanofibre was approximately 112 nm. The addition of 2.5 wt% NFC to PVOH produced a uniform nanofibre with a very small amount of beads (Fig. 2 c,d), and the diameter of the nanofibre was approximately 104 nm.

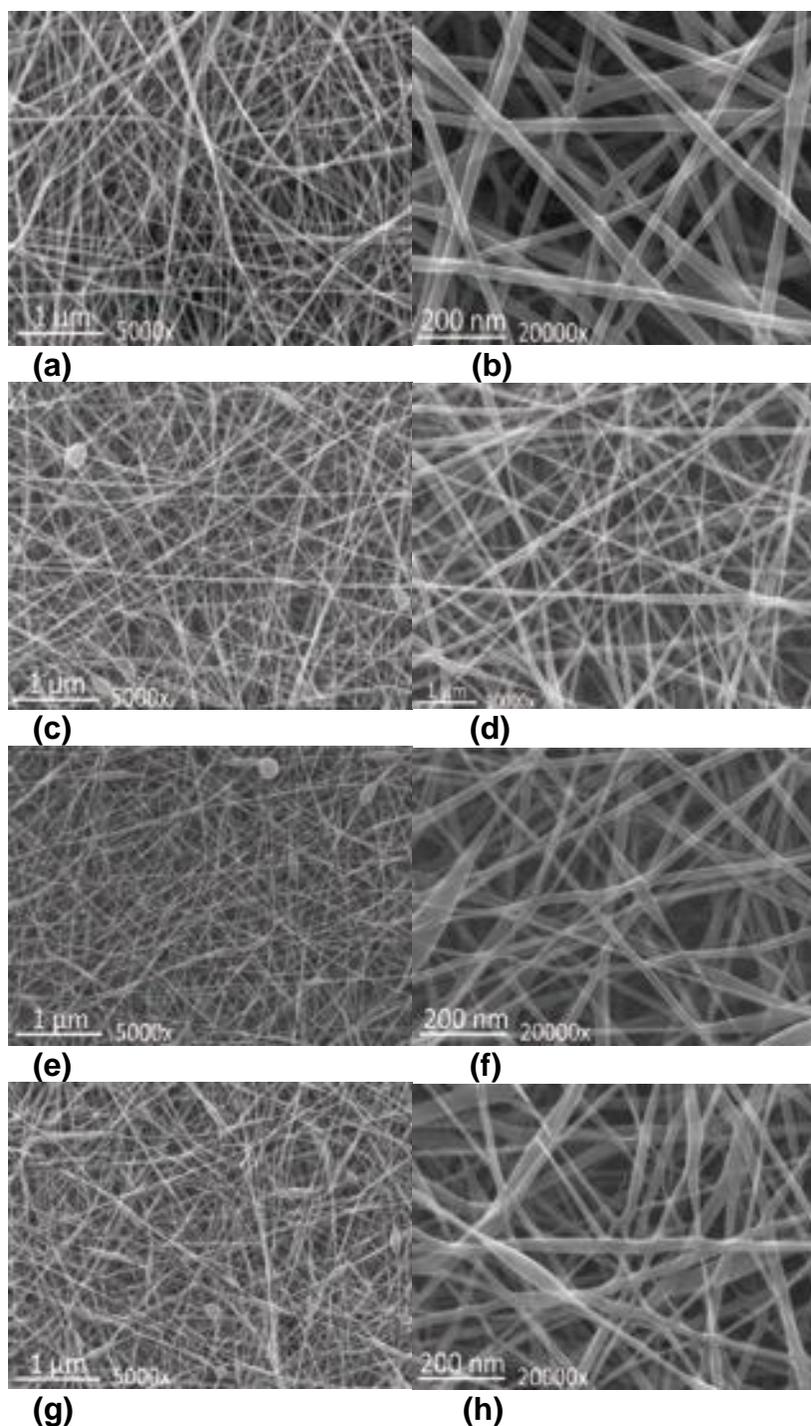


Fig. 2. SEM micrographs of electrospun PVOH/NFC nanofibre at different NFC concentrations with magnification of 500x and 20000x: (a, b) 0% NFC; (c, d) 2.5% NFC; (e, f) 5.0% NFC; and (g, h) 7.5% NFC.

As the concentration of NFC increased to 5.0 wt% (Fig. 2 e, f) and 7.5 wt% (Fig. 2 g, h), the beads increased and formed elliptical shapes. Beads were observed at high concentration of NFC due to low compatibility of NFC to the PVOH. The average diameter also decreased to 96 nm and 84 nm with the concentration of 5 wt% and 7.5 wt% NFC, respectively. The decreasing diameter of electrospun nanofibre after the addition of CNF

was reported previously (Braga *et al.* 2019; Park *et al.* 2014). However, a greater amount of nanofibrillated lignocellulose in PVOH spun-nanofibre is known to result in beads and agglomerated spun-fibre (Seo *et al.* 2019).

In general, the addition of NFC increased the viscosity of electrospinning polymer solution, which decreased the fibre diameter (Park *et al.* 2014). Furthermore, the conductivity of polymer solution was improved by the NFC. Increasing conductivity led to decreased fibre diameter. This can be attributed to the electrostatic charge density of the polymer solution increasing with higher conductivity. When the conductivity is high, the elongation of the jet between the needle and collector is weaker, producing a thicker fibre (Peresin *et al.* 2010). The conductivity of PVOH and PVOH/NFC with the concentration of NFC 2.5%, 5.0%, and 7.5% is shown in Fig. 3. It is shown that the viscosity of 12% PVOH was 107 mPas. The viscosity of PVOH with the present of NFC 2.5% was 139 mPas, and it decreased to 126 and 94.8 mPas, respectively, with the addition of 5.0% and 7.5% NFC. However, the conductivity of PVOH increased with the presence of NFC. It was 231.8 $\mu\text{S}/\text{cm}$, 238.7 $\mu\text{S}/\text{cm}$, 251.3 $\mu\text{S}/\text{cm}$, and 252.6 $\mu\text{S}/\text{cm}$ with the presence of 0%, 2.5%, 5.0%, and 7.5% NFC, respectively.

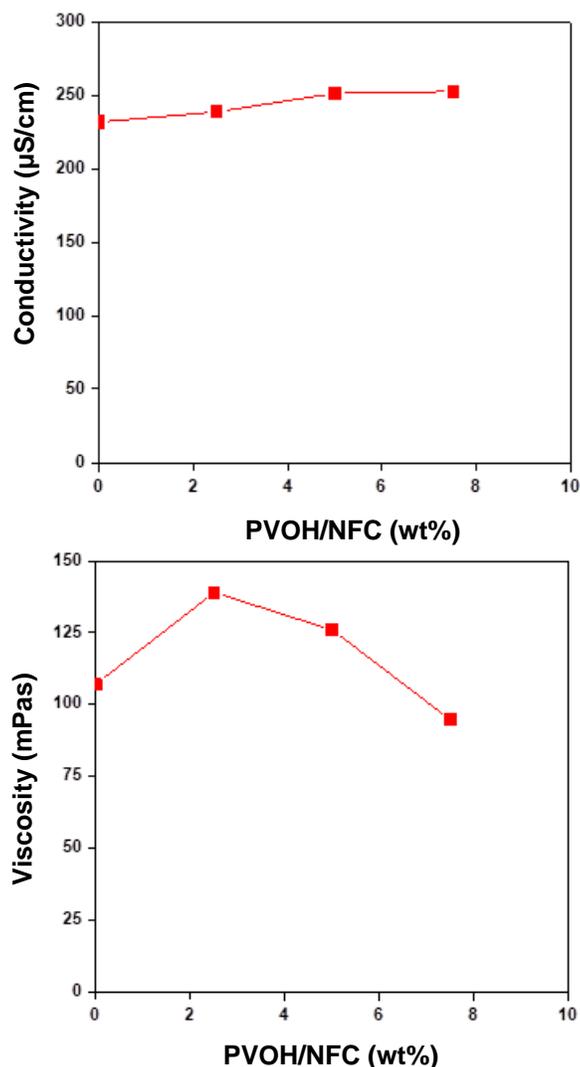


Fig. 3. The conductivity and the viscosity of 12 wt.% PVOH solution in distilled water at different NFC concentrations 0%, 2.5%, 5.0%, and 7.5%

FTIR Spectra

FTIR spectra confirmed the functional groups and the purity of NFC as well as the presence of NFC in the PVOH nanofibre. Figure 4 shows the main absorption bands of NFC at 1427 cm^{-1} (HCH and OCH bending vibration), 1370 cm^{-1} (CH deformation vibration, CH_3 or OH in plane), 1270 to 1232 cm^{-1} (C-O-C, ether bond, aryl-alkyl ether), 1160 to 1083 cm^{-1} (C-O-C antisymmetric bridge stretching, pyranose ring skeletal), 1105 cm^{-1} (OH (C-OH)), 1041 to 1054 cm^{-1} , C-O symmetric stretching (C-O of primary alcohol)), and 896 to 915 cm^{-1} (COC, CCO, and CCH deformation and stretching vibration). These absorptions correspond to cellulose I. In addition, a band at 1642 cm^{-1} is associated to adsorbed water; 3278 to 3495 cm^{-1} and 2890 cm^{-1} absorption correspond to OH stretching and H-C-H stretching (alkyl, aliphatic), respectively (Hatch *et al.* 2019; Harahap *et al.* 2019).

Figure 4 confirms the chemical structure of neat PVOH nanofiber, showing dominant absorption peaks at 3323 cm^{-1} , 2941 cm^{-1} , 1733 cm^{-1} , 1428 cm^{-1} , 1090 cm^{-1} , and 844 cm^{-1} , which are attributed to OH, CH_2 , C=O, CH-C-H, C-O, and C-C. The acetate group in the PVOH was represented in the band at 1959 cm^{-1} (Braga *et al.* 2019).

The electrospun composite nanofibre PVOH/NFC spectrum showed the characteristics neat PVOH fibre due to the small number of NFC particles in the PVOH matrix. The main shift can be seen in the band at 3317 cm^{-1} corresponding to stretching of OH groups, which increased because of the interaction of PVOH-free OH groups with NFC structure by hydrogen bonding. The absorption band from 1087 to 1026 cm^{-1} of the C-O of the alcohol group of NFC can be clearly seen in the electrospun composite nanofibre PVOH/NFC.

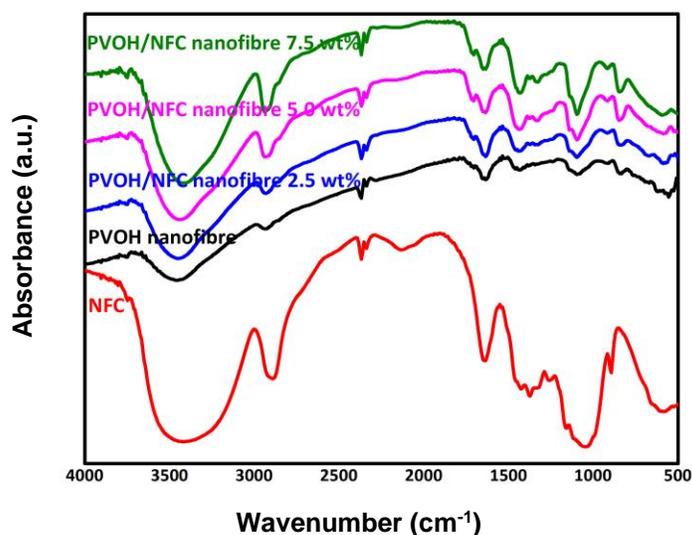


Fig. 4. FTIR spectra of NFC and PVOH/NFC nanofibre with concentration of NFC 0%, 2.5%, 5.0%, and 7.5% (wt)

X-ray Analysis Results

X-ray diffraction (XRD) patterns of PVOH, NFC, and PVOH/NFC spun-fibre at concentration of 2.5 wt% NFC are plotted in Fig. 5.

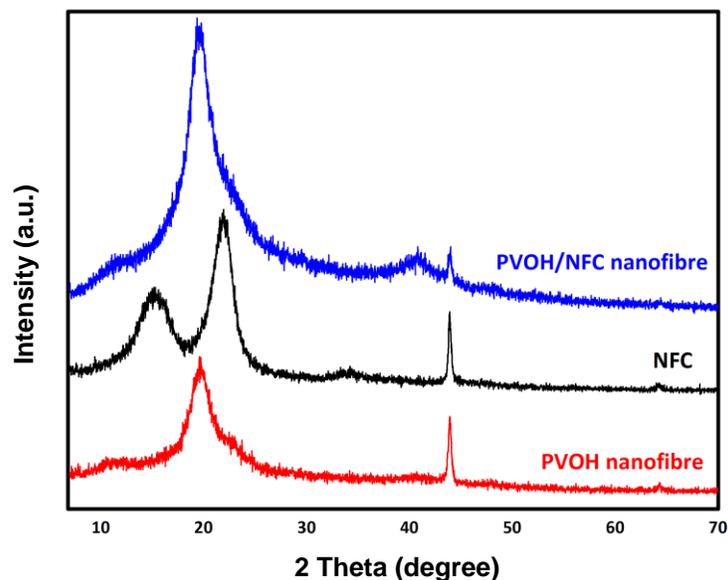


Fig. 5. XRD pattern of PVOH, NFC, and PVOH/NFC nanofibre

The NFC shows a diffraction pattern with a sharp peak at $2\theta = 25^\circ$, which corresponds to the crystallographic plane 002, and $2\theta = 15.5^\circ$, assigned to the cellulose I structure (Hammiche *et al.* 2016). This structure confirmed the FTIR result. The NFC produced in this study was cellulose I. The diffraction pattern of PVOH is characterised at $2\theta = 11.5^\circ, 19.5^\circ, 22.6^\circ, 32.1^\circ,$ and 40.5° assigned to the semi-crystalline nature of the polymer (Li *et al.* 2013). The XRD pattern of PVOH/NFC spun-fibre showed a slightly decreased of crystallinity of PVOH. This is due to the interaction between PVOH and NFC and the electrospinning process affecting the crystallinity of both polymers. Several studies reported that rapid processes in electrospinning hinder development of crystallinity; electrospinning nanofibre results in a low degree of crystallinity (de Dicastillo *et al.* 2017). However, the PVOH/NFC diffraction pattern shows a more intense peak at 22.6° , confirming that NFC acts as nucleating agent.

Thermal Properties of Electrospun Composite Nanofibre Polyvinyl Alcohol/Nanofibrillated Cellulose

The TGA curves for PVOH nanofibre, NFC, and electrospun composite nanofibre PVOH/NFC are presented in Fig. 6. The degradation of all samples shows three well-differentiated steps. The first degradation for all samples occurred from room temperature to 100°C , which is associated with an initial mass loss due to evaporation of moisture content (Santos *et al.* 2014). In this step, the mass loss was 9%, 7%, and 9% for PVOH nanofibre, NFC, and PVOH/NFC nanofibre mat, respectively. The degradation of NFC reported for depolymerisation, dehydration, and decomposition of glycosyl units occurred at the lowest (earliest) temperatures. The biggest mass loss of NFC occurred between 280°C and 400°C (70%), which corresponds to the decomposition of NFC to the monomer of D-glucopyranose.

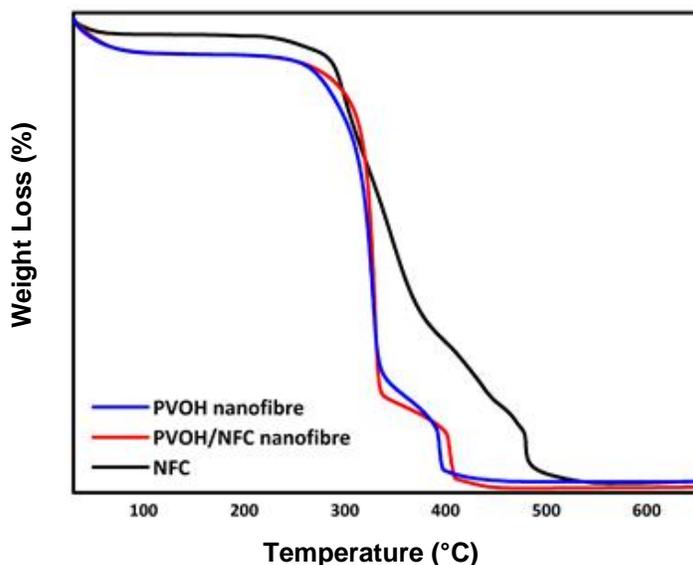


Fig. 6. TGA curve of NFC, PVOH nanofibre and PVOH/NFC nanofibre with a heating rate 10 °C/min

For electrospun PVOH nanofibre, the second degradation occurred from 270 °C to 330 °C with a mass loss 65% corresponding to the thermal degradation of crystalline PVOH. The residual mass of PVOH at 600 °C was 2%. For electrospun composite nanofibre PVOH/NFC, the degradation thermal was shifted to higher temperature, which can be attributed to the presence of NFC improve its thermal stability. It is reported that NFC acted as a thermal barrier because of the formation of hydrogen bonds between NFC and polymer (de Dicastillo *et al.* 2017).

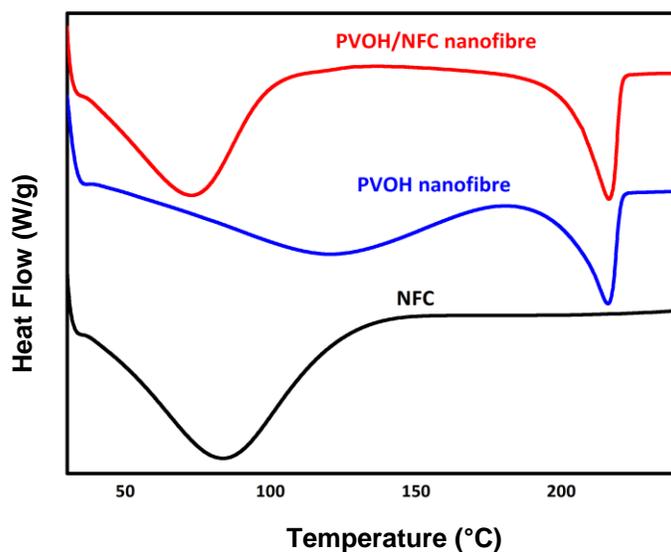


Fig. 7. DSC thermogram for NFC, PVOH nanofibre and PVOH/NFC nanofibre with a heating rate 10 °C/min

In the DSC curve (Fig. 7), NFC showed a broad endotherm peak at 40 °C to 140 °C associated with water in the sample. PVOH nanofibre exhibited an endotherm peak with a melting temperature (T_m) at 215 °C and T_g at 120 °C. After the addition of NFC to PVOH nanofibre, T_m appeared at 216 °C, and T_g decreased to 72 °C. The same result was reported in a previous work (Han *et al.* 2017).

CONCLUSIONS

1. Nanofibrillated cellulose (NFC) isolated from oil palm empty fruit bunches by steam explosion and acid hydrolysis was found to have a diameter 50 nm with lengths of several micrometres.
2. The introduction of NFC into the poly(vinyl alcohol) (PVOH) nanofibre decreased its diameter. However, it produced elliptical beads at high concentration of NFC.
3. The X-ray diffraction (XRD) pattern of electrospun composite nanofibre PVOH/NFC showed a slightly decrease of crystallinity of PVOH. However, the PVOH/NFC diffraction pattern showed a rise intensity of peak at 22.6°, confirming that NFC acts as a nucleating agent.
4. The presence of NFC improved the thermal stability of electrospun PVOH nanofibre. The electrospun composite nanofibre PVOH/NFC potentially can be used as an ultrafiltration membrane in future application.

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