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# Fabrication and characterization of thick films made of chitosan and nanofibrillar cellulose derived from pineapple leaf

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**Abstract.** Research on the development of natural polymers is becoming increasingly important because of environmental reasons. In this study, the thick films manufactured using chitosan and reinforced with nanofibrillar cellulose that is derived from pineapple fibers have been successfully fabricated. Nanofibrillar cellulose is obtained by isolating the cellulose from pineapple fibers through chemical treatments and ultra-fine grinding. Thick chitosan-nanocellulose composite films are prepared by mixing the nanocellulose with a chitosan solution at concentrations of 1%, 3%, 5%, 7%, and 10%. The structures and properties of the obtained films are examined by X-ray diffraction (XRD), Ultraviolet and Visible (UV/Vis) spectroscopy, and Fourier transform infrared (FTIR) spectroscopy. Characterization of the optical properties of the materials by UV/Vis techniques exhibit that the highest transparency can be achieved using 3% nanocellulose. This result is also related to the XRD spectra, which depict that the addition of 1%–3% nanocellulose can effectively increase the degree of crystallinity of the composite films. These results prove that the addition of nanocellulose to a chitosan matrix depicts a good potential to improve the properties of composite films and that these films can be used as biodegradable packaging plastics.

**Keywords:** chitosan, nanofibrillar cellulose, pineapple leaf fiber

## 1. Introduction

Recently, research related to the development of biocomposites has attracted significant attention due to increasing environmental concerns. Biocomposites satisfy the requirements for a “green technology” in which the matrix and reinforcing agents are biodegradable materials that can be easily decomposed in the environment, thereby avoiding pollution [1]

Chitosan and nanocellulose are biodegradable materials that have been extensively developed due to their numerous advantages. Chitosan is one of the strongest natural polymers that are known; it is a hydrophilic cationic polysaccharide that is produced by the deacetylation of chitin, which is one of the most abundant materials on earth [2].

Cellulose is a natural ingredient that is used as a reinforcing agent in the polymer matrix. It is a fiber that can be isolated from certain plants, such as banana, pineapple, flax, cotton, jute, sisal and hemp, through chemical and mechanical processes [3]. Nanocellulose can be classified into three major sub-categories based on its dimensions, utilities, and preparation (depending on the source of cellulose and the processing conditions) [4]. The nomenclatures used are cellulose nanocrystals (CNC), cellulose nanofibers (CNF) and nanofibrillar cellulose (NFC). Cherian *et al.* [3] isolated



cellulose from the pineapple-leaf fibers to ensure usage as a reinforcing agent in polyurethane. In this study, the isolated cellulose, which was obtained from the pineapple-leaf fibers, was used as a reinforcing agent in the chitosan matrix.

## 2. Materials and methods

### 2.1. Materials

The chitosan that was used in this study exhibited a deacetylation level that was greater than 80% and was purchased from Balai Penelitian Bioteknologi & Bioindustri, Bogor. The pineapple-leaf fiber was purchased from CV Hasanah Niaga, Bandung. The chemical reagents that were used included sodium hydroxide, sodium chlorite, hydrochloric acid, and acetic acid, which were ordered from PT. Merck Chemicals and Life Sciences.

### 2.2. Preparation of the composite film

The composite film was prepared by mixing the solution of a 2 % w/v acetated chitosan with NFC for several variation of w/w concentrations of 0%, 1%, 3%, 5%, 7%, and 10%. This solution was further heated for two h at 80 °C upon stirring and was ultrasonicated at 800 watt for 30 min. The chitosan/nanocellulose composite film was casted by pouring a 25 mL solution over a 20 × 20 cm mica, which was then dried at room temperature for 48 h.

### 2.3. Characterization

**2.3.1. XRD analysis.** The XRD characterizations are performed to obtain information about the crystal structures of metal and alloy materials, minerals, inorganic compounds, polymers, and organic materials. In this research, the studies were conducted using a Rigaku Smartlab XRD instrument at a voltage of 3 kV and a current of 30 mA. The source of X-ray radiation was Cu-K $\alpha$  ( $\alpha = 1,54 \text{ \AA}$ ), and the samples were examined within the  $2\theta$  range of 5° to 80° at a scanning rate of 0.02°.

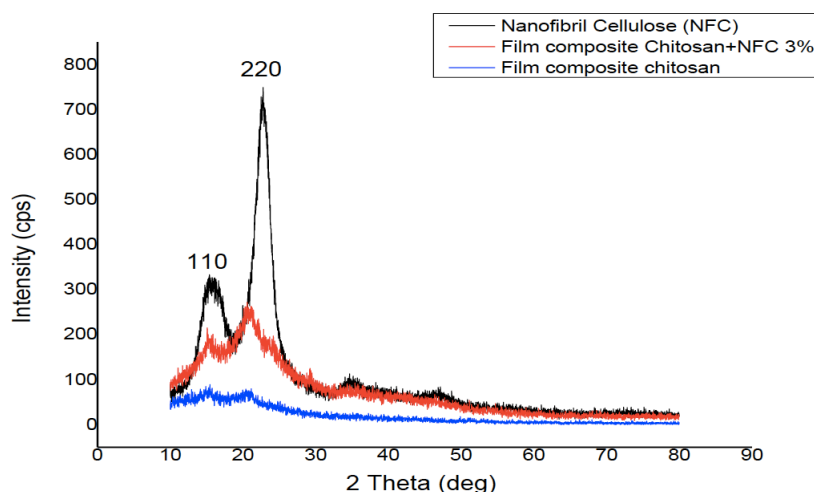
**2.3.2. Optical properties.** The absorbance of the plastic composite films was measured using a Thermo UV/Vis 10 S Genesys spectrophotometer at a resolution of 1 nm in the wavelength range of 200 to 800 nm.

**2.3.3. FTIR analysis.** An FTIR analysis was performed to study the effect of the addition of NFS to the chitosan film and to determine the IR band and its shift due to the reaction between nanocellulose and chitosan. The FTIR spectra were obtained for nanofibrillar cellulose, chitosan films, and chitosan composites containing nanofibrillar cellulose using an FTIR-ATR Thermo Scientific Nicolet iS5 device at 400–4000 cm<sup>-1</sup>.

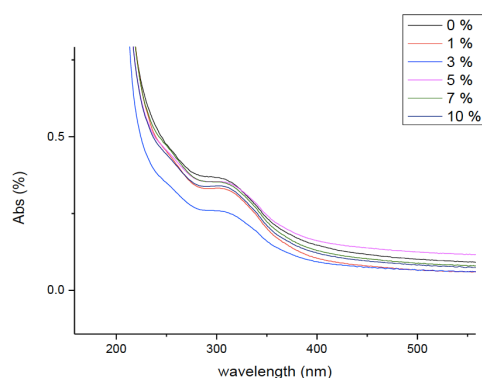
## 3. Results and discussion

### 3.1. XRD studies

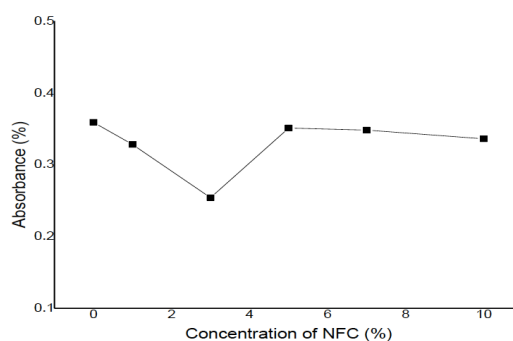
Figure 1 illustrates the diffractograms of NFS, chitosan, and the composite film. It can be observed that NFS has a crystalline structure, exhibiting sharp peaks at  $2\theta = 15.51^\circ$  and  $22.84^\circ$ . According to Cherian *et al.* [5], who also conducted a study on the isolation of nanocellulose from pineapple-leaf fibers, a sharp peak appeared at  $\theta = 22.7^\circ$ . Khan *et al.* [6] also reported the presence of sharp peaks at  $2\theta = 16.8^\circ$  and  $2\theta = 20^\circ$ – $22.4^\circ$  in the diffractogram of nanocellulose. These peaks correspond to the (1 1 0) and (2 2 0) planes of cellulose, respectively. Figure 1 depicts the peaks corresponding to chitosan at  $2\theta = 15.35$  and  $20.80$ . According to Celebi and Kurt [1], the wide peak of chitosan can be observed at  $2\theta = 20^\circ$ , expressing the semi-amorphous properties of this compound. The addition of 3% nanocellulose to chitosan caused an increase in the intensities of the peaks that were observed at  $2\theta = 15.25^\circ$  and  $20.46^\circ$ . This is likely to be caused due to the so-called transcrystallization effect [6], which is defined as the orientation of a semi-crystalline matrix perpendicular to the nanocrystalline cellulose [2].



**Figure 1.** X-ray diffractograms of nanocellulose and film composites.



**Figure 2.** UV-Vis absorption spectra of composite film



**Figure 3.** Absorption peaks of the composite films at 310 nm.

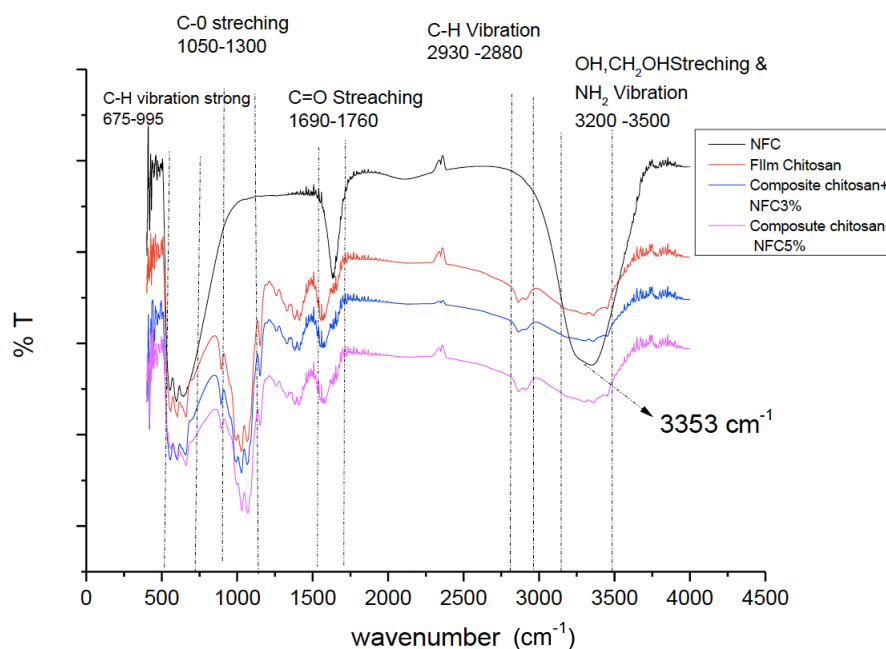
### 3.2. Analysis of the optical properties of the NFC-chitosan composite films

The optical properties of the NFC–chitosan composite films were evaluated between 200 and 800 nm. Figure 2 depicts the absorbance spectra of a pure chitosan film and chitosan–NFC composites with NFC concentrations of 1%, 3%, 5%, 7%, and 10%. It can be observed that the composite film that contains 3% NFC exhibited the lowest absorbance at a wavelength of 310 nm (the other concentrations caused similar absorbance values). This indicated that the chitosan–NFC (3%) composite was the most transparent one.

The absorbance-peak values that were measured for the composite films at a wavelength of 310 nm can be observed from figure. 3. Wu *et al.* [7] performed a study related to the optical properties of nanofibrillar cellulose and observed that the transmittance value of the composite film decreased with the increasing NFC concentrations. In this study, chitosan–NFC (3%) was the most transparent composite film, presumably because of its high crystallinity.

### 3.3. FTIR studies

The FTIR spectra of the pineapple-leaf fibers can be observed from figure 4. The chitosan absorption peaks were characterized by the intra- and intermolecular stretching of O–H and the vibration of CH<sub>2</sub>OH at 3500–3250 cm<sup>-1</sup>, which was overlapped by the stretching of -NH<sub>2</sub> at 3500–3280 cm<sup>-1</sup>. There was also a C–H vibration at 2930–2880 cm<sup>-1</sup>.



**Figure 4.** The FTIR spectra of NFC and the composite films.

In the nanocellulose spectrum, there was a wide transmission band at approximately 3400–3200  $\text{cm}^{-1}$ , indicating the vibration of the O–H strain due to hydrogen bonding of the hydroxyl group of cellulose [8]. A sharp peak was also observed at approximately 3337  $\text{cm}^{-1}$ , which corresponded to O–H vibration due to the occurrence of an intramolecular hydrogen bond [9]. Strong transmission bands are also observed at approximately 675–995  $\text{cm}^{-1}$ , indicating the occurrence of C–H vibrations. At approximately 1160  $\text{cm}^{-1}$ , there was a transmission band indicating a C=O strain.

The sharp peak that was observed in the FTIR spectrum of nanocellulose at approximately 3353  $\text{cm}^{-1}$  cannot be observed in the spectra of pure chitosan films or composite chitosan–NFC films. Additionally, there was no signal corresponding to the C=O strain in the latter spectra. Hence, there were no significant differences between the FTIR spectrum of chitosan and the spectra of the composite films, which was due to the small concentration of NFC that was used.

#### 4. Conclusions

In this study, we successfully prepared biocomposites that were derived from chitosan and nanocellulose obtained from the pineapple-leaf fibers. The composite film that contained 3% nanocellulose was the most transparent one among all the studied materials, as confirmed by the UV/Vis experiments. The results of the XRD analysis proved that the film crystallinity increases when the chitosan matrix is added with NFC. The low absorbance value of the composite chitosan–NFC film was most likely to be caused due to its high crystallinity. Our results demonstrated that biodegradable plastic with good transparency can be obtained from the composite film that has been successfully improved by the presence of NFC in the chitosan matrix.

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