Supporting Information for

Water-soluble nanocrystalline cellulose films with tunable oxygen

permeability

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Mechanical properties

Tensile strength, bursting strength and elongation at break of the film samples were performed by using the computer control of tensile test machine and computer control board bursting instrument (DCP-KZ1000, Changjiang Co. Ltd., SiChuan and DCP-NPY5600, Changjiang Co. Ltd., SiChuan) according to ISO1924-2. Tensile strength (TS), bursting strength (B) and elongation at break (Eb %) of the GG, NCC and GG-NCC nanocomposite films are presented in Figure S1a and Figure S1b. The TS of pure GG films was found to be 20.5 N and the reinforced films with addition of 5.0, 10.0, 15.0, 25.0 and 30.0% (w/w) NCC increased the TS values up to 25.0, 24.9, 25.8, 27.8, 29.6 and 37.4 N, respectively. The bursting strength of the GG film is 100.2 Kpa, when the NCC concentration increased to 35.0% (w/w), the bursting strength value get to the peak 385.3 Kpa. But when the NCC content continues to increase, the TS and Eb value decreased. Figure S1b shows the effect of the NCC content on the Eb (%) of NCC-reinforced GG films. The Eb value was found to be 4.0% for the pure GG film and 3.2,2.5,2.1,1.8,1.3 and 1.1 for 5.0, 10.0, 15.0, 20.0 and 25.0% (w/w) of NCC addition, respectively.



Figure S1. Effect of NCC content on the (a) tensile strength, bursting strength and (b) elongation at break of the films

water soluble properties

The water soluble properties of the films are also be tested. The films have been completely dissolved in water after been stirred for 5 h. The aqueous solution of GG film is very clear, but the aqueous solution of the composite films become more muddy with the increase of NCC concentration , This could ascribe to the highly crystalline of NCC.



Figure S2. The dissolution degree of the films after being stirred for 5h





Figure S3. The cross section of the films

X-ray diffraction

For X-ray diffraction (XRD) analysis, film samples were folded several times to increase the sample thickness. Samples were analyzed between 2 $\theta = 5^{\circ}$ and 40° in a D8 Discover X-ray Diffractometer (D/MAX-TTRIII, Japanese Neo Confucianism, Japan).

XRD analysis was performed in order to analyze the structural property of the GG and GG/NCC films. Figure S4 shows the diffractograms of NCC, GG with 45 and 60% NCC. The diffractogram of NCC showed the characteristic peaks at $2 \theta = 16.8^{\circ}$ and $2 \theta = 22.4^{\circ}$ and corresponding to 110 and 200 planes of cellulose I, respectively. Incorporation of 45 and 60% NCC into alginate resulted in the presence of an additional peak at $2 \theta = 16.8^{\circ}$, relatively to the contribution of NCC that allowed increasing the crystallinity of the films. The main contribution of NCC to induce a higher level of crystallinity was observed in the peak at $2 \theta = 22.4^{\circ}$. These results are in agreement with all the results observed previously that described improvement of mechanical and barrier properties between GG and NCC.



Figure S4. X-ray diffractograms for GG-NCC film

The TEM image of NCC

NCC presents a simple needle-like structure with the length between 100-600 nm.



Figure S5. The TEM image of NCC