Supporting Information

1. Evaluation of the structural and mechanical characteristics of CNC / MEA composite films

The neat CNC exhibits absorption bands at 3338, 2910, and 1016 cm^{-1} due to the stretching vibration of OH, C-H and C-O. The presence of MEA between CNC increases the intermolecular distance and can form hydrogen bonds with the hydroxyl groups on CNC nanoparticles, which reduces the intermolecular force and increases the toughness of CNC nanoparticles, as seen in FT-IR Fig. 5 a). The largest peak shifts towards higher frequencies with 9% MEA addition, probably as a result of hydrogen bonding interactions between the hydrophilic hydroxyl groups on the surface of the CNC nanoparticles and the hydroxyl groups of the MEA chain. If the peak positions of -OH with 9% MEA addition in the figure are compared to those of the neat CNC iridescent film, it can be seen that the peak positions of -OH after plasticization and modification with MEA are shifted to the lower wave number, with the CNC iridescent film with 9% MEA addition most obviously shifting from 3467 cm^{-1} to 3282 cm^{-11}. As MEA is an alcohol amine compound, this is the case. The molecular structure contains these functional groups, including -OH, -C=O and $-NH_2$, where the O and N atoms have strong electronegativity and a small radius, so when these substances are added to the CNC water suspension, a significant amount of the CNC nanoparticles and CNC nanoparticles hydroxyl that was initially formed will be "robbed." In addition to an increase in intermolecular hydrogen bonds and a decrease in the hydrogen connections between CNC and its hydroxyl groups, the plasticizer also forms more hydrogen bonds with CNC's -OH. The improved mechanical characteristics of the CNC iridescent film, which are stated to be a decrease in brittleness and an increase in flexibility, may be achieved by the employment of the plasticizer as an energy dissipating phase.

Fig. 5b) illustrates the XRD characterisation. The typical diffraction peaks of CNC and CNC/MEA composite films are 15.8 ° and 22.6 °, respectively, and correspond to crystallographic planes (101) and (002) crystallographic planes [2]. Our finding shows that the crystalline structure of CNC is mostly conserved with the addition of MEA in the CNC/MEA nanocomposite, and the configuration of the liquid crystal intermediate phase is stable at the MEA concentration. MEA can reduce the tensile modulus of CNC nanoparticles and weaken both intra- and intermolecular hydrogen bonding. It is clear that the crystallinity of CNC iridescent films is

essentially unaffected by the addition of the MEA plasticizer. It follows that MEA can only interact with the hydroxyl groups that are available to the CNC surface in the intercrystalline region of the CNC iridescent film.

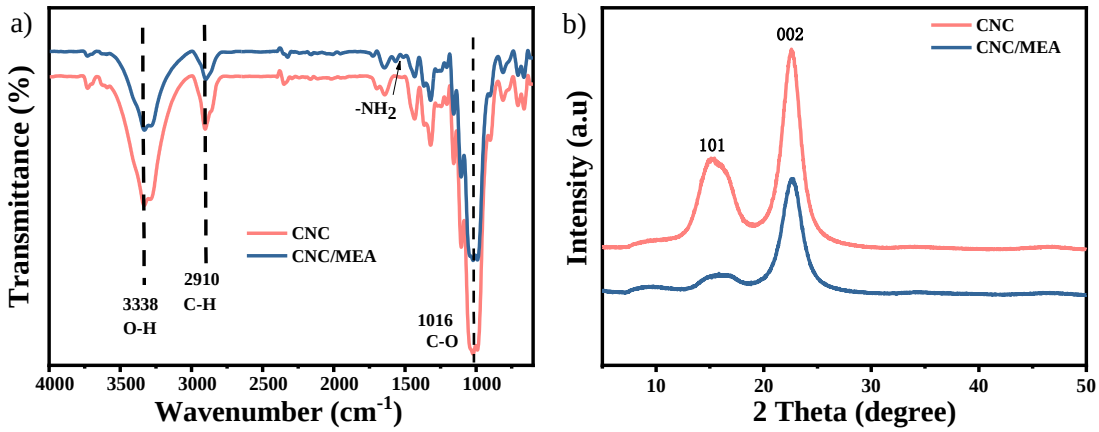

Fig. 5 a) FT-IR and b) XRD diagram of CNC / MEA composite film

2. Evaluation of the structural and mechanical characteristics of ACNC composite films

As seen in Fig. 9 a), a FT-IR analysis of ACNC (which contains 9% KH-792) was performed to determine if the amino group had been added to the CNC surface. The chemical structure of CNC functional groups was not significantly altered by the addition of KH-792; the cellulose -OH stretching vibration characteristic was 3338 cm^{-1}, and distinct characteristic peaks emerged at 2910 and 1029 cm^{-1}, which were the peak of the CH stretching vibration characteristic, the bending vibration peaks of the in-plane of $-CH_2$ and -OCH and the C-O-C stretching vibration peak, respectively. Indicating the effective grafting of the silane coupling agent to CNC, ACNC displayed new distinctive absorption peaks at 1498 cm^{-1}, which corresponded to the bending vibration peaks of $-NH_2$, respectively. The XRD diffraction patterns of the unmodified CNC films and the CNC modified by KH-792 are shown in Fig. 9 b). The findings demonstrate that the same distinctive peaks with the same crystalline cell structure can be found at 2 = 16.3 ° and 22.6 °, respectively, which correspond to the crystallographic planes (101) and (002) [2]. With the use of the aforementioned information, it can be demonstrated that the KH-792 change only affects CNC's surface and does not harm the crystalline area within CNC.

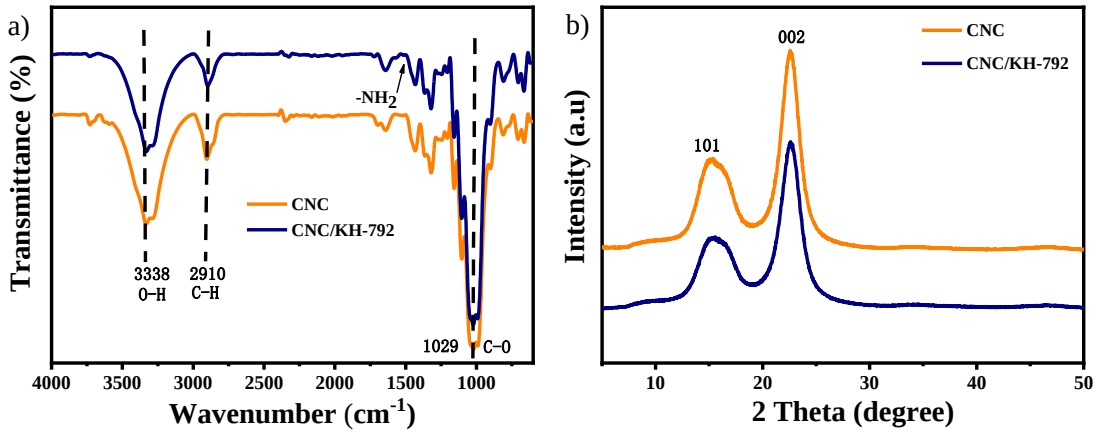

Fig. 9 a) FT-IR and b) XRD diagram of the CNC / KH-792 composite film

2. Particle size diagram of ACNC / MEA / SnO_2 composite films

Fig. 15 shows the CNC size distribution after the addition of 0% a), 2% b), 4% c), 6% d), 8% e) and 10% $SnCl_4$ $5H_2O$ to ACNC / MEA. As the amount of $SnCl_4$ $5H_2O$ increases, the size distribution of the CNC also becomes wider. At this point, the size distribution of the separated CNC is obviously more thorough because the SnO_2 substance is distributed in the CNC within the crevices of the CNC. The gap widens and the CNC size distribution expands as $SnCl_4$ $5H_2O$ content increases, with average particle sizes of 29.3, 75.7, 106.3, 110.4, and 124.1 nm, respectively, between 60 and 255 nm.

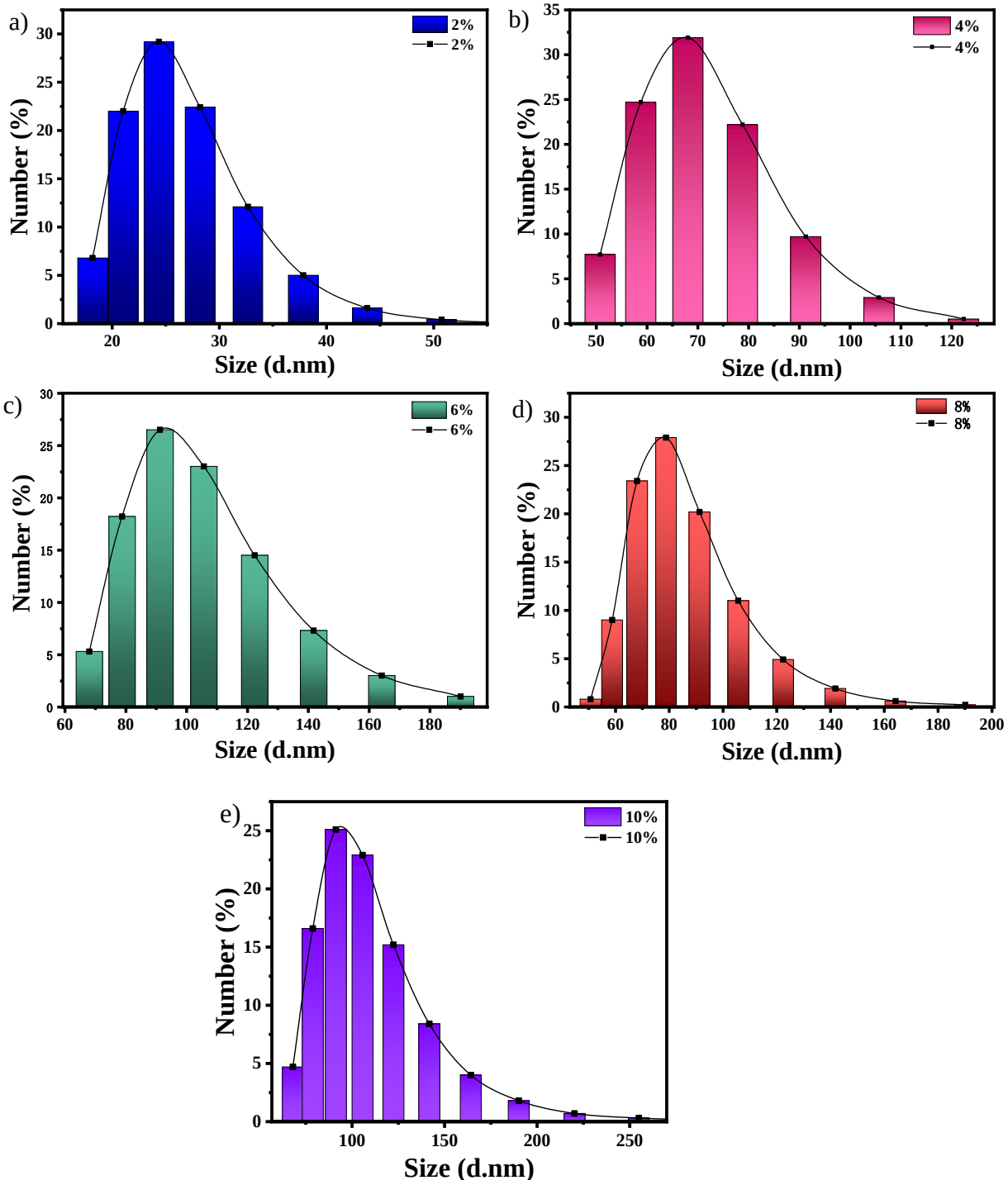

Fig. 15 Particle size diagram of ACNC / MEA / SnO_2 composite films with added $SnCl_4$ $5H_2O$ 0%, 2%, 4%, 6%, 8% and 10%

3. FTIR spectra of ACNC/MEA/SnO_2 composite films in different humidity environments

FTIR spectra provide clear and convincing proof that hydrogen bonds are present during the humidity response of CNC films. Fig. 16 a) shows that the spectral bands at

3300 cm^{-1} and the peaks at 2890 cm^{-1} are indicative of the stretching vibrations of -OH and C-H, respectively, while the peaks at 1428 cm^{-1} and 1314 cm^{-1} are ascribed to the stretching vibration of $-CH_2$ and out-of-plane oscillations, respectively [3]. These are all typical cellulose peaks found in the spectra, and the peaks at 1030 cm^{-1} and 820 cm^{-1} correspond to the stretching vibrations of -C-O and $-C_1$-H. The stretching vibrations of S=O and C-O-S in $-OSO_3$- are also responsible for the peaks in the spectra at 1205 cm^{-1} and 826 cm^{-1}, respectively [4]. The peak attributable to -OH is red-shifted, suggesting the creation of hydrogen bonds between the -OH of CNC and the -OH group of water. According to expectations, when relative humidity increased, the hydroxyl stretching vibrations' maxima gradually redshifted, moving from 3338 cm^{-1} (55%) to 3333 cm^{-1} (75%), 3330 cm^{-1} (85%), and 3325 cm^{-1} (98%), as seen in Fig. 16 b). Similarly to a recent study by Ni et al. [5], it was deduced that more hydrogen bonds between CNC and water molecules formed as relative humidity increased.

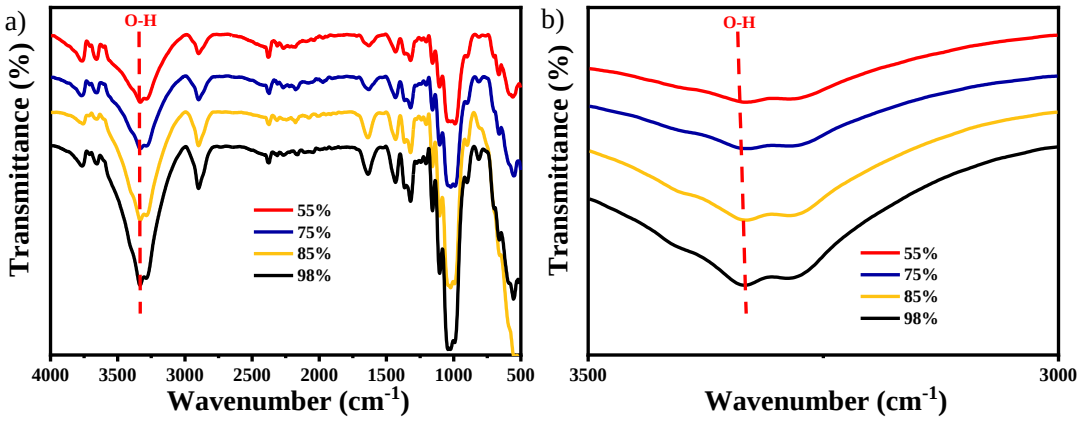

Fig. 16 FTIR spectra of ACNC/MEA/SnO_2 composite films in different humidity environments: a) 4000-500 cm^{-1}; b) 3500-3000 cm^{-1}

4. FTIR map of the ACNC / MEA / SnO_2 composite film under formaldehyde

Similarly, hydrogen bonding may be seen in the FTIR spectra of the CNC film's formaldehyde reaction process. The peak of -OH is red-shifted in Fig. 18 a), which denotes the creation of hydrogen bonds between the -OH of CNC and the -OH of formaldehyde. With an increase in formaldehyde concentration, the stretching vibration peak of -OH was specifically slightly redshifted. The peak position changed slightly from 3337 cm^{-1} (0.15 mg/m^3) to 3335 cm^{-1} (1.5 mg/m^3) and 3333 cm^{-1} (5 mg/m^3), as illustrated in Fig. 18 b). As a result, we may assume that more hydrogen bonds are formed between CNC molecules and formaldehyde as the formaldehyde level increases..

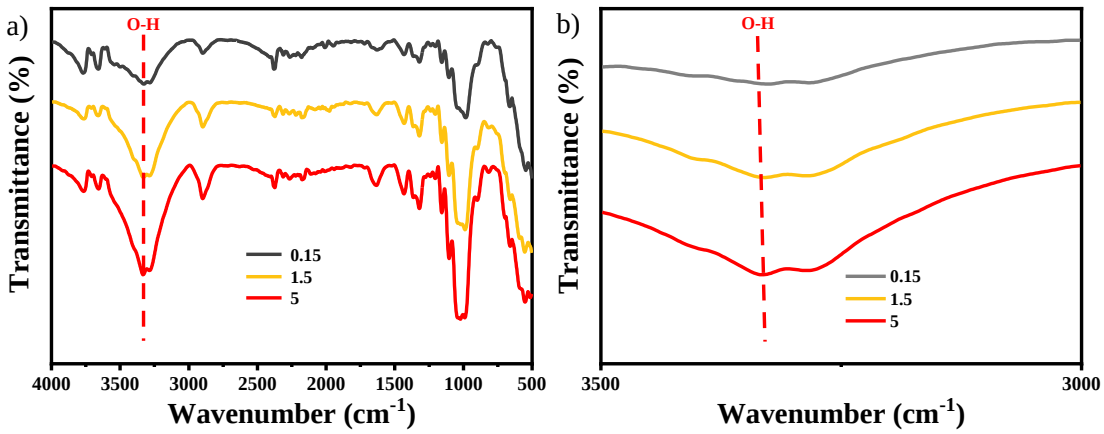

Fig. 18 FTIR map of the ACNC / MEA / SnO_2 composite film under formaldehyde: a) 4000-500 cm^{-1}; b) 3500-3000 cm^{-1}

1. J.-L. Huang, C.-J. Li and D. G. Gray, *ACS Sustainable Chemistry & Engineering*, 2013, **1**, 1160-1164.
2. A. El Oudiani, Y. Chaabouni, S. Msahli and F. Sakli, *Carbohydrate Polymers*, 2011, **86**, 1221-1229.
3. A. Kaushik and M. Singh, *Carbohydrate research*, 2011, **346**, 76-85.
4. P. Lu and Y.-L. Hsieh, *Carbohydrate polymers*, 2010, **82**, 329-336.
5. Y.-P. Ni, Q.-T. Li, L. Chen, W.-S. Wu, Z.-H. Qin, Y. Zhang, L. Chen, X.-L. Wang and Y.-Z. Wang, *Chemical Engineering Journal*, 2019, **374**, 694-705.