Supporting Information for:

Preparation of cellulose nanocrystals coated with polymer crystals and their application to composite films

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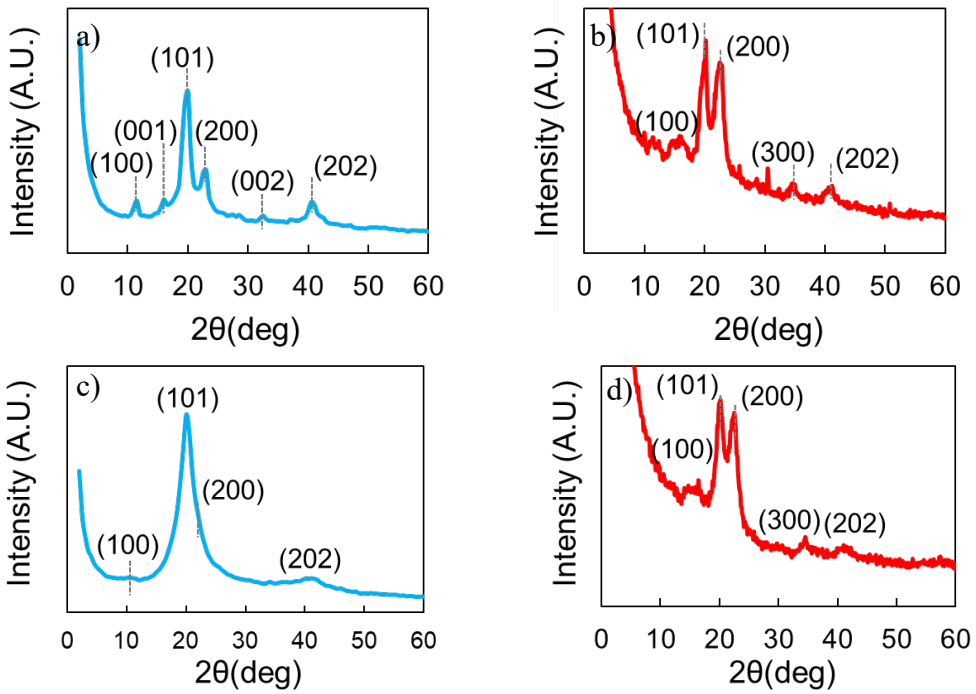

Fig. S1. X-ray diffraction pattern of a) poly(vinyl alcohol) (PVA) crystals, b) nanocomposite fibers $NCF_{(CNC/PVA)}$, c) poly(vinyl alcohol-*co*-ethylene) (EVOH) crystals, and d) the nanocomposite fibers $NCF_{(CNC/EVOH)}$.

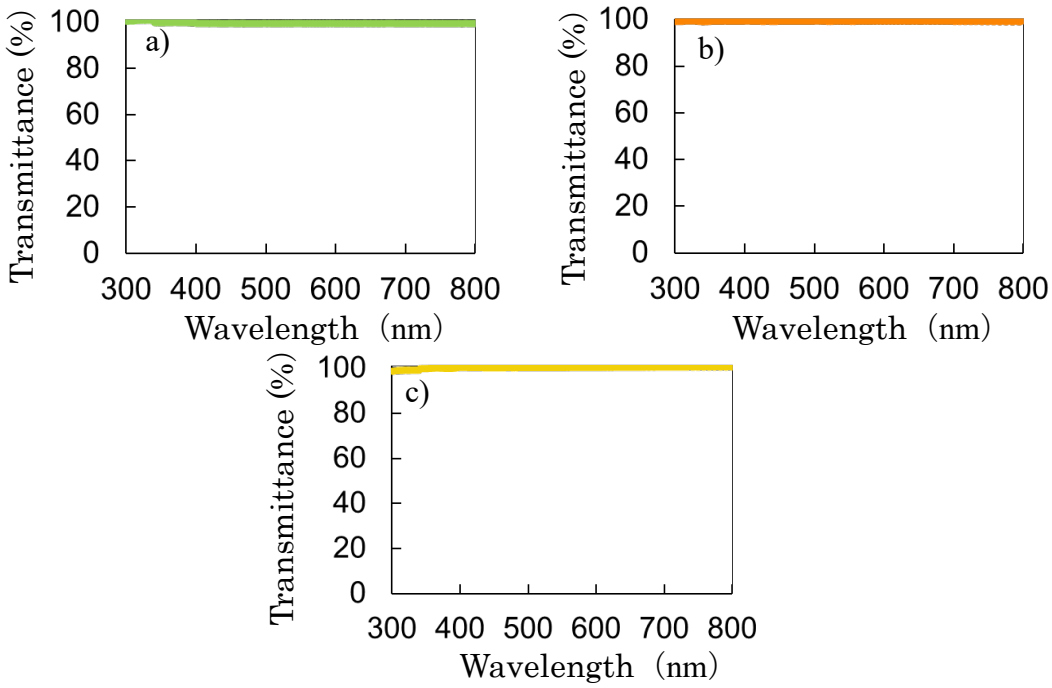

Fig. S2. Ultraviolet–visible spectra of a) CNC 0.1 wt%/PVA composite film, b) $NCF_{(CNC/PVA)}$ 0.1 wt%/PVA composite film, and c) $NCF_{(CNC/EVOH)}$ 0.1 wt%/PVA composite film.

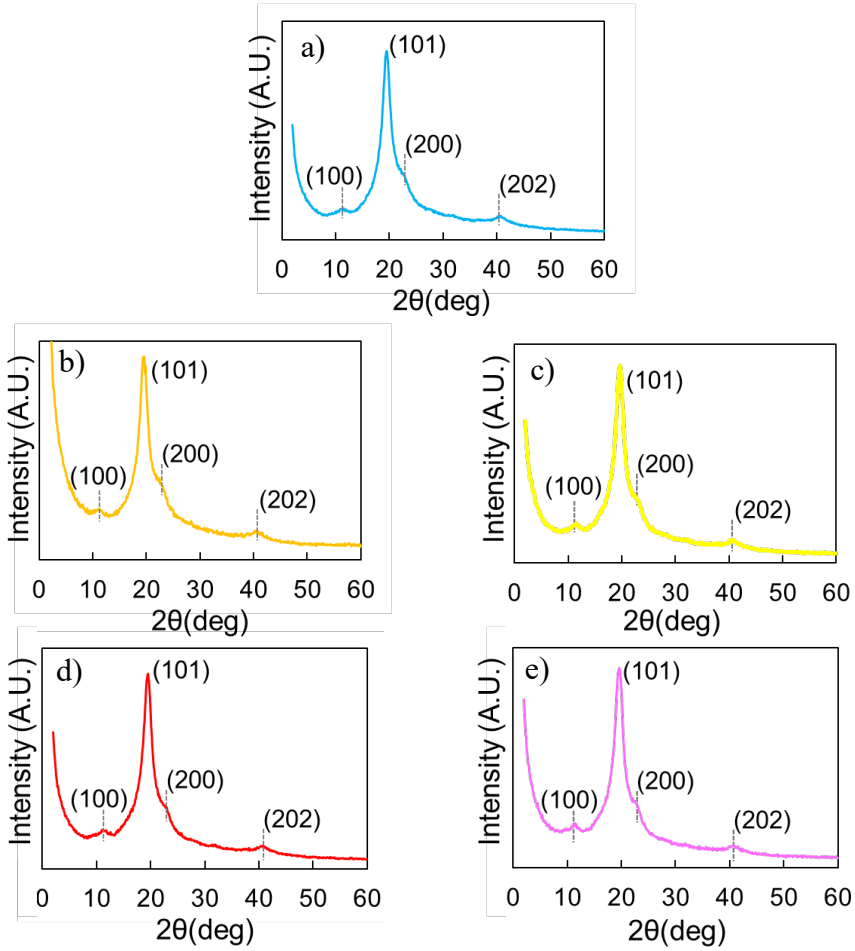

Fig. S3. X-ray diffraction pattern of a) poly(vinyl alcohol) (PVA) film, b) $NCF_{(CNF/PVA)}0.1\%$/PVA film, c) $NCF_{(CNF/EVOH)}0.1\%$/PVA film, d) $NCF_{(CNC/PVA)}0.1\%$/PVA film, and e) $NCF_{(CNC/EVOH)}0.1\%$/PVA film.

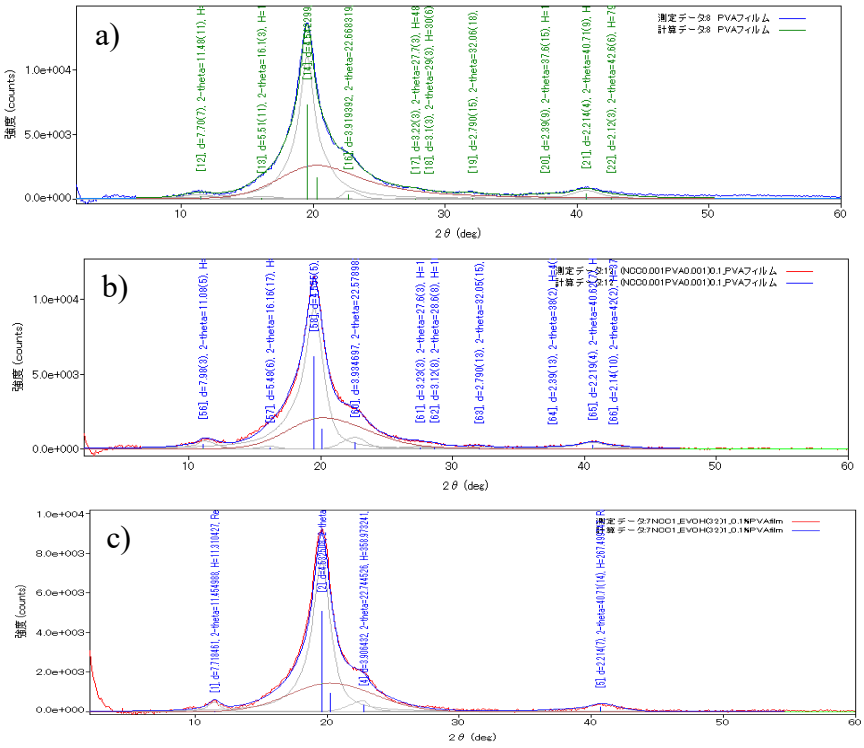

Fig. S4. Deconvoluted X-ray diffraction pattern of a) poly(vinyl alcohol) (PVA) film (Fig. S3(a)), b) $NCF_{(CNC/PVA)}0.1\%$/PVA film (Fig. S3(d)), and c) $NCF_{(CNC/EVOH)}0.1\%$/PVA film (Fig. S3(e)). The red line in the figure is the amorphous peak.

Table S1. Crystallite sizes obtained from resolved X-ray diffraction patterns of PVA and NCF/PVA composite films.

Film	NCF content (%)	CNC content (%)	Crystallite size（nm）/ *hkl* indexing			
			100	101	200	202
PVA	-	-	3.1	5.3	5.7	3.4
$NCF_{(CNC/PVA)}$/PVA	0.1	0.05[a]	4.9	4.8	4.0	3.3
$NCF_{(CNC/EVOH)}$/PVA	0.1	0.05[b]	4.3	5.4	5.0	3.3

[a] Added as $NCF_{(CNC/PVA = 1/1)}$ [b] Added as $NCF_{(CNC/EVOH = 1/1)}$

$NCF_{(CNF/PVA=1/1)}$, $NCF_{(CNF/EVOH=1/1)}$, $NCF_{(CNF/PVA=1/1)}$/PVA composite films, and $NCF_{(CNF/EVOH=1/1)}$/PVA composite films were prepared by previously reported method[S1] and the viscoelasticity of the films was measured using a DMA1 dynamic viscoelasticity measuring device (Mettler Toledo). The measurement conditions were 40 °C–160 °C at a displacement of 8 μm, a frequency of 1 Hz, and a heating rate of 3 °C/min. X-ray diffraction measurements of the composite films were performed in a 2θ range of 2°–60° using an X-ray diffraction apparatus (Rigaku VariMax with RAPID) with a tube voltage of 40 kV and a tube current of 30 mA by irradiating with Cu Kα rays for 20 min. In addition, the crystallinity and crystallite size were calculated based on the measured profile data using the Rigaku X-ray analysis software PDXL.

([S1] Uchida T, Iwaguro F, Yanai R, Dodo H. RSC Adv. 2017;7:19828-19832.)

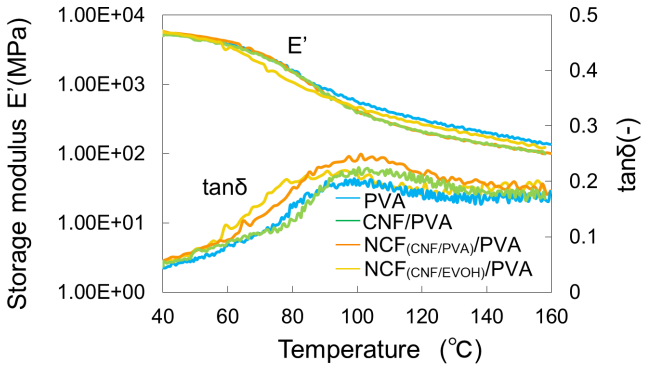

Fig. S5. Dynamic viscoelasticity curve of the poly(vinyl alcohol) and composite films.

Table S2. Tan δ peak temperature of poly(vinyl alcohol) (PVA) and composite films

film	NCF content (%)	CNF content (wt%)	Tan δ peak temp. (°C)
PVA	0	0	99
$NCF_{(CNF/PVA)}$/PVA	0.1	0.05[a]	104
$NCF_{(CNF/EVOH)}$/PVA	0.1	0.05[b]	97

[a] Added as $NCF_{(CNF/PVA = 1/1)}$ [b] Added as $NCF_{(CNF/EVOH = 1/1)}$

Table S3. Crystallinity of poly(vinyl alcohol) (PVA) crystals in the PVA film and composite films

film	NCF content (%)	CNF content (%)	Crystallinity of PVA (%)
PVA	-	-	59
$NCF_{(CNF/PVA)}$/PVA	0.1	0.05[a]	64
$NCF_{(CNF/EVOH)}$/PVA	0.1	0.05[b]	61

[a] Added as $NCF_{(CNF/PVA = 1/1)}$ [b] Added as $NCF_{(CNF/EVOH = 1/1)}$