Supporting Information

Controlling Miscibility of the Interphase in Polymer-Grafted Nanocellulose/Cellulose Triacetate Nanocomposites

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Evaporation Rate of Casting Solvents. In the present study, the evaporation rate was controlled by covering the petri dish with a PTFE sheet with holes and changing the pressure during drying. The detailed conditions are shown in Table S1.

Table S1 Drying Conditions of PEG-TOCN/CTA Composite Films.

	temperature (˚C)	vacuum (MPa)	cover of petri dish during drying of dispersion
DCM	40	−0.03	PTFE with 20 holes
DOX	40	< −0.1	none
DMAc	40	−0.04	none

Crystallinity of CTA Matrix in PEG-TOCN/CTA Composites. The crystallinities of the CTA matrix were calculated from the WAXD data with the Ruland-Vonk or amorphous contribution subtraction method (Table S2).[1,2] Peak positions corresponding to the crystalline and amorphous CTA were determined according to previous studies.[3,4]

Table S2 Crystallinity of CTA Matrix in PEG-TOCN/CTA Composite Films.

	Crystallinity (%)	
	Pristine CTA	PEG-TOCN/CTA
DMAc	35.3	33.9
DOX	27.8	29.6
DMAc	28.8	29.4

Dynamic Mechanical Properties of PEG-NH_2/CTA Blend Films. The glass transition temperatures (T_g) of the CTA matrix in the PEG-NH_2/CTA blend films are shown in Figure S2. The addition of PEG-NH_2 caused a decrease in T_g. This indicates that free PEG-NH_2 chains were miscible with the CTA matrix and had a plasticizing effect on the films regardless of the casting solvent.

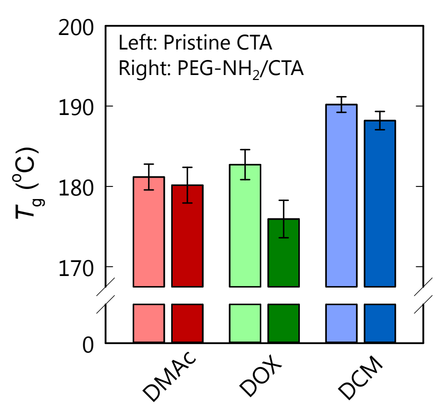

Figure S1. Glass transition temperature of the PEG-NH_2/CTA blend films calculated from tanδ curves. DMAc: *N,N*-dimethylacetamide, DOX: 1,4-dioxane, and DCM: dichloromethane.

Statistical Analysis for Mechanical and Thermal Properties of PEG-TOCN/CTA Composite Films. Welch's unequal variances *t*-test was performed to confirm the statistical significance of differences in mechanical properties and T_g of the pristine CTA and PEG-TOCN/CTA composite films. The *t*-test was performed as a two-tailed test on Microsoft Excel 2010 using Analysis ToolPak. If the *p* value is less than 0.025, it is interpreted that two populations have different means. The results are shown in Table S3. In the case of T_g, the *p* value was greater than 0.025. This is probably because of the small sample numbers (N = 3) in dynamic mechanical analysis.

Table S3 The *t* and *p* Values for Difference in Mechanical and Thermal Properties.

	DMAc		DOX		DCM	
	t	*p* value	*t*	*p* value	*t*	*p* value
Young's modulus	-2.93	0.008	3.31	0.003	-4.70	0.001
Tensile strength	-2.70	0.015	-1.66	0.108	-3.55	0.009
Toughness	-3.55	0.004	-2.87	0.007	-3.75	0.002
Yield strength	-0.99	0.337	-2.18	0.036	-3.54	0.012
T_g	-1.89	0.131	-0.60	0.591	2.65	0.118

Mechanical Properties of PEG-NH_2/CTA Blend Films. Mechanical properties of PEG-NH_2/CTA blend films are shown in Figure S1. No improvement in Young's modulus or yield strength was observed. The tensile strength and toughness of the composite films was slightly decreased by the addition of PEG-NH_2.

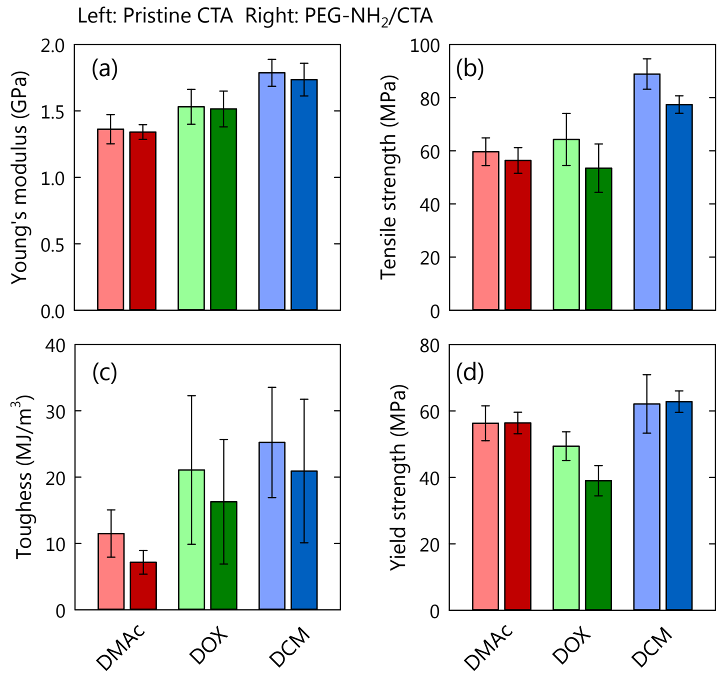

Figure S2. Mechanical properties of the PEG-NH_2/CTA blend films: (a) Young's modulus, (b) tensile strength, (c) toughness, and (d) Yield strength. DMAc: *N,N*-dimethylacetamide, DOX: 1,4-dioxane, and DCM: dichloromethane.

Fundamental Properties of Nanocomposite Films. The densities and water contents of the composite films are shown in Table S4. The density and water content of a nanocomposite film were calculated by weighing the specimens before and after drying at 105 °C for 3 h.

Table S4 Densities and Water Contents of PEG-TOCN/CTA Composite Films.

	density (g/cm^3)		water content (%)	
	Pristine CTA	PEG-TOCN/CTA	Pristine CTA	PEG-TOCN/CTA
DMAc	1.18 ± 0.04	1.18 ± 0.04	1.77 ± 0.51	1.66 ± 0.10
DOX	1.17 ± 0.02	1.15 ± 0.03	1.84 ± 0.22	1.93 ± 0.35

DCM	1.19 ± 0.01	1.21 ± 0.02	1.60 ± 0.21	1.50 ± 0.15

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