

## Supporting Information

Dynamic covalent diarylbibenzofuranone-modified nanocellulose:  
Mechanochromic behaviour and application in self-healing polymer  
composites

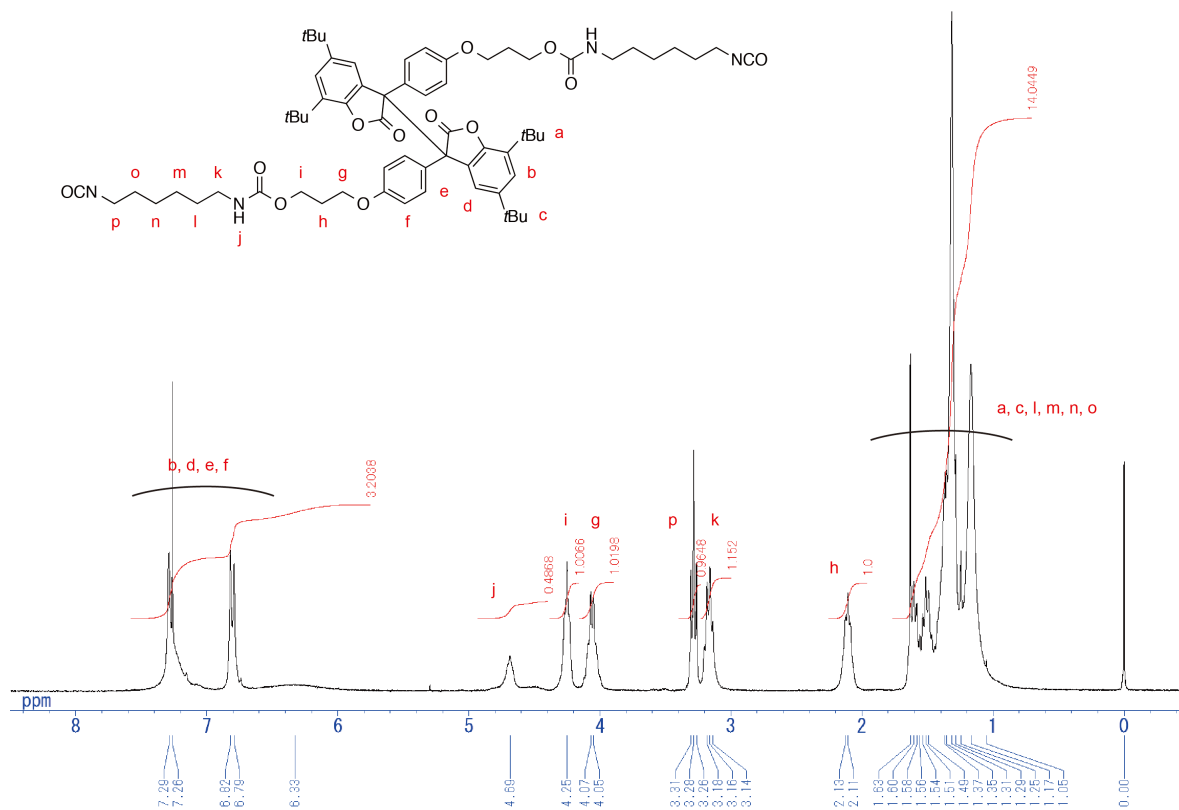
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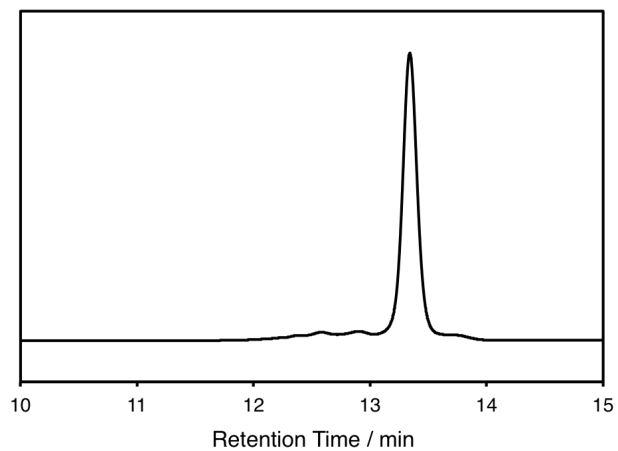
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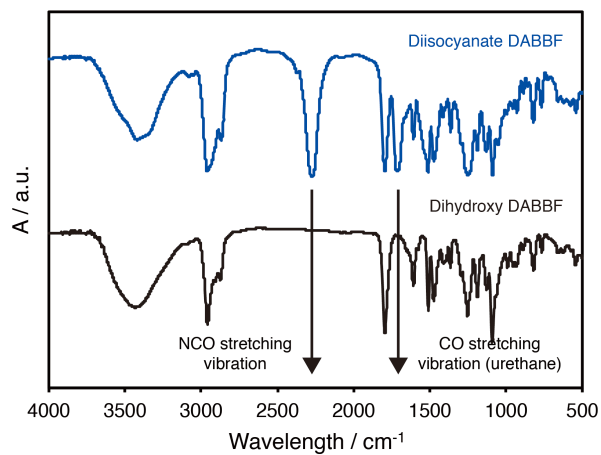
\*E-mail: [otsuka@polymer.titech.ac.jp](mailto:otsuka@polymer.titech.ac.jp)



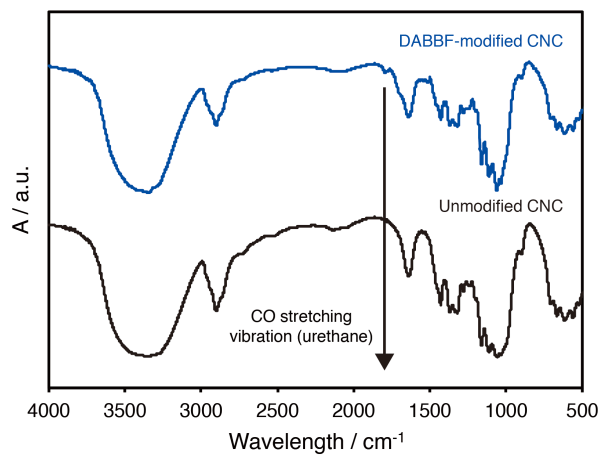
**Figure S1.** Chemical structure and  $^1\text{H}$  NMR spectrum of diisocyanate DABBF (in  $\text{CDCl}_3$ ). The integral of peak p is less than and the integral of peak k is more than those of peaks i, g, and h, indicating that a small fraction of isocyanate groups (approx. 9%) is deactivated.



**Figure S2.** SEC curve of diisocyanate DABBF after purification. A small fraction of higher molecular weight components is observed in the retention time range from 12 to 13 min.



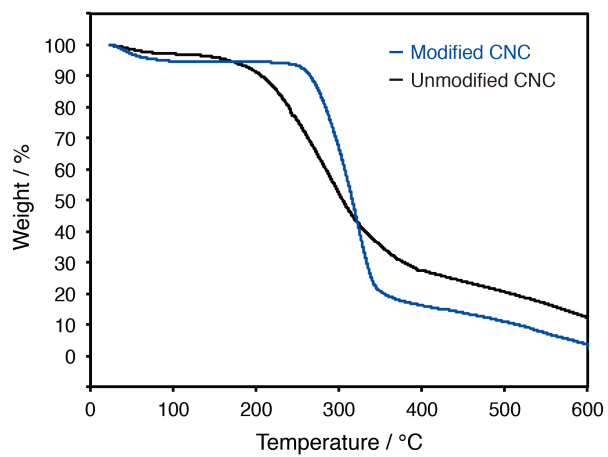
**Figure S3.** FT-IR spectra of dihydroxy DABBF (black) and diisocyanate DABBF (blue). The carbonyl stretching vibration band and the isocyanate stretching vibration band are observed at 1720 cm<sup>-1</sup> and 2270 cm<sup>-1</sup>, respectively.



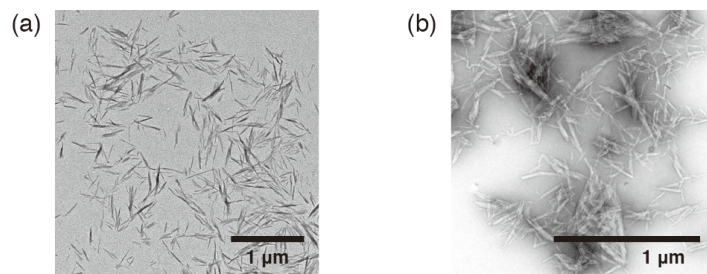
**Figure S4.** FT-IR spectra of unmodified CNCs (black) and DABBF-modified CNCs (blue). The carbonyl stretching vibration band is observed at 1800 cm<sup>-1</sup>.

**Table S1.** Results of elemental analyses for unmodified CNCs and DABBF-modified CNCs

		C	H	N	O
Unmodified CNCs	Fraction / %	41.49	5.93	0.12	47.77
	Composition	12	20	0	10
Modified CNCs	Fraction / %	42.94	5.8	0.56	47.16
	Composition	455	731	4	375

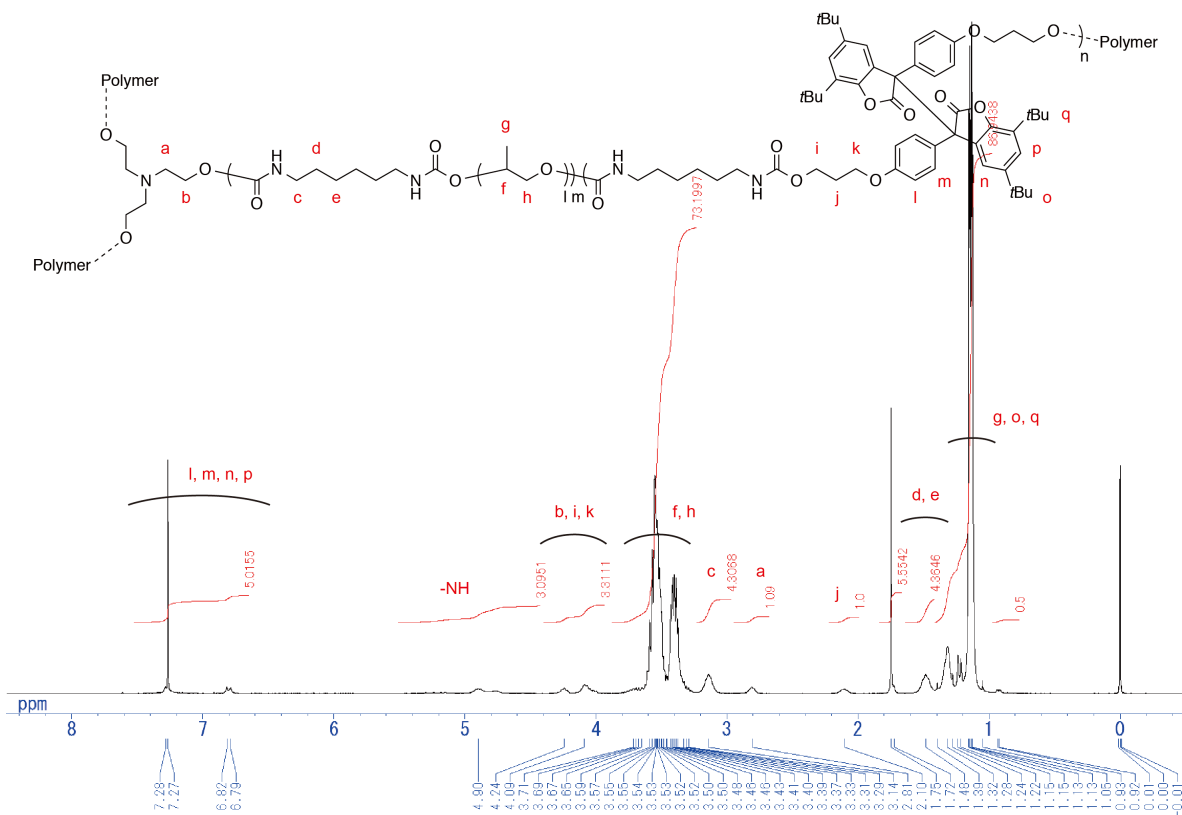


**Figure S5.** Thermogravimetric analyses for unmodified CNCs (black) and DABBF modified CNCs (blue). The weight losses at below 120 °C represent the evaporation of adsorbed water.

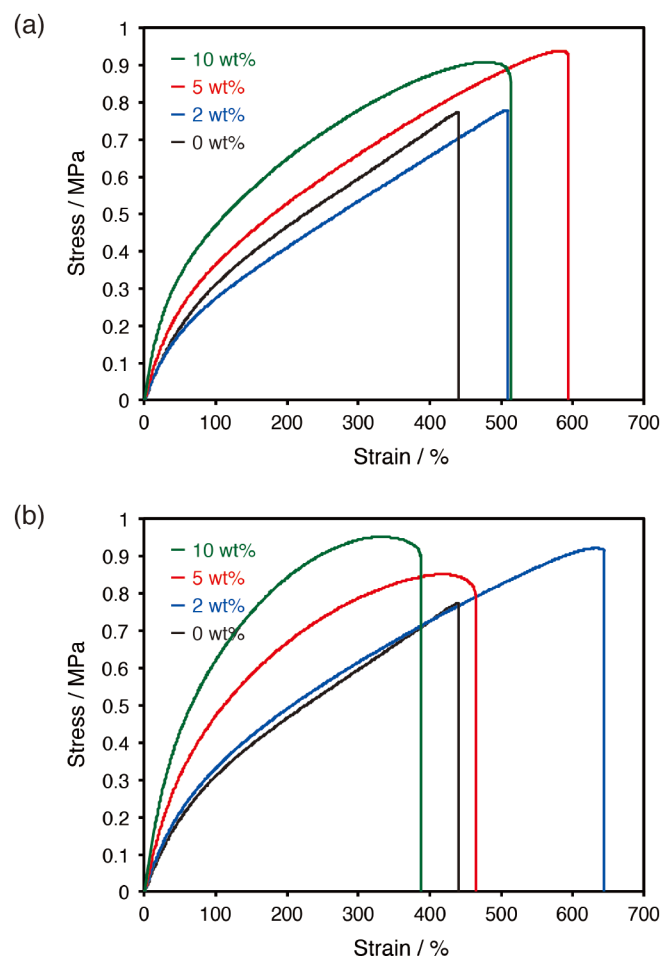


**Figure S6.** TEM images of (a) unmodified CNCs and (b) DABBF-modified CNCs.

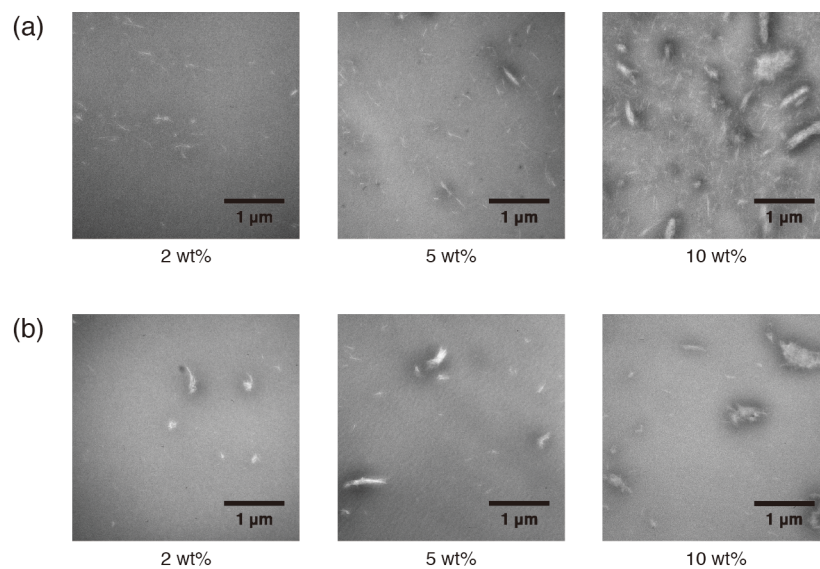




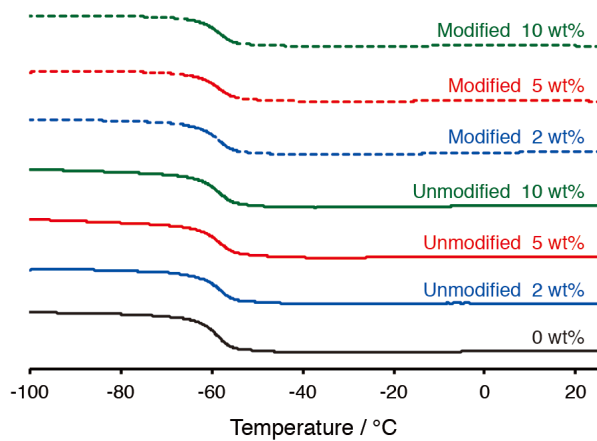
**Figure S7.** Chemical structure and <sup>1</sup>H NMR spectrum of cross-linked polymer with DABBF (in CDCl<sub>3</sub>). The composition ratio was determined to be 1/1.38/5.93/3.05 of TEA/DABBF/HDI/PPG, which is largely similar to the feed ratio (1/1.5/6/3).



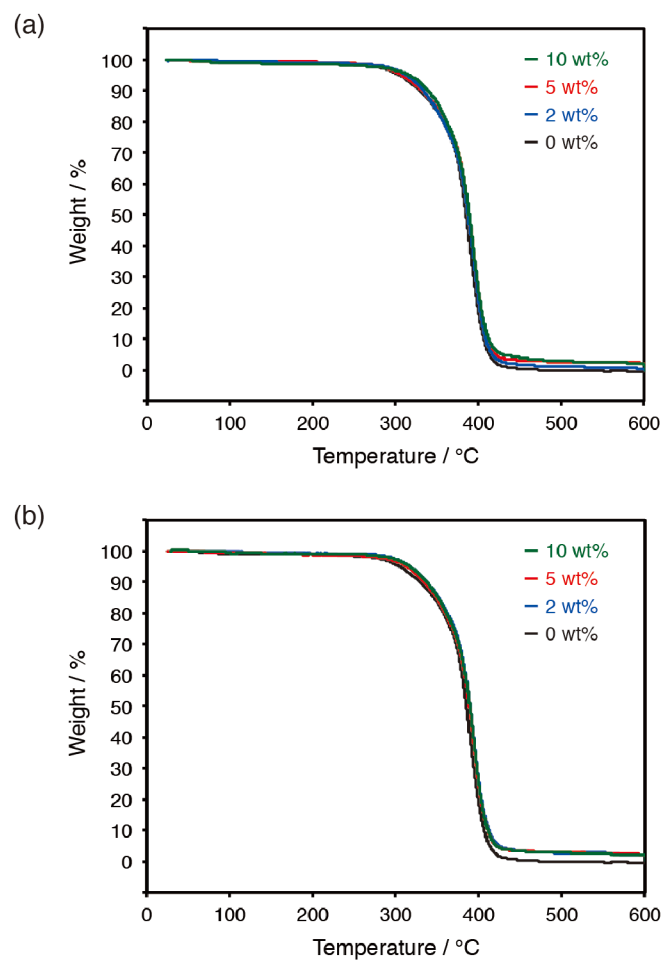
**Figure S8.** Stress-strain curves of composite films with 0 wt% (black), 2 wt% (blue), 5 wt% (red), and 10 wt% (green) of (a) unmodified CNCs or (b) DABBF-modified CNCs.



**Figure S9.** TEM images of composite films with 2 wt%, 5 wt%, and 10 wt% of (a) unmodified CNCs or (b) DABBF-modified CNCs. Dispersed and aggregated CNCs were observed. Large CNC aggregation was observed in the composites with 10 wt% of unmodified and DABBF-modified CNCs.



**Figure S10.** DSC curves of composite films with 0 wt% (black), 2 wt% (blue), 5 wt% (red), and 10 wt% (green) of unmodified CNCs (solid lines) or DABBF-modified CNCs (dotted lines).



**Figure S11.** Thermogravimetric analyses for composite films with 0 wt% (black), 2 wt% (blue), 5 wt% (red), and 10 wt% (green) of (a) unmodified CNCs or (b) DABBF-modified CNCs.

**Table S2.** Glass transition temperatures and degradation (5% weight decrease) temperatures for composite films with unmodified CNCs or DABBF-modified CNCs

	Unmodified CNCs				Modified CNCs		
	wt%	2	5	10	2	5	10
$T_g / ^\circ\text{C}$	-58.6	-58.2	-58.3	-58.4	-58.3	-58.4	-58.2
Degrad. temp. / $^\circ\text{C}$	305	308	313	316	317	314	317