

Electronic Supplementary Information

Flexible cellulose based piezoelectric composite membrane involving PVDF and BaTiO₃ synthesized with the assistance of TEMPO-oxidized cellulose nanofibril

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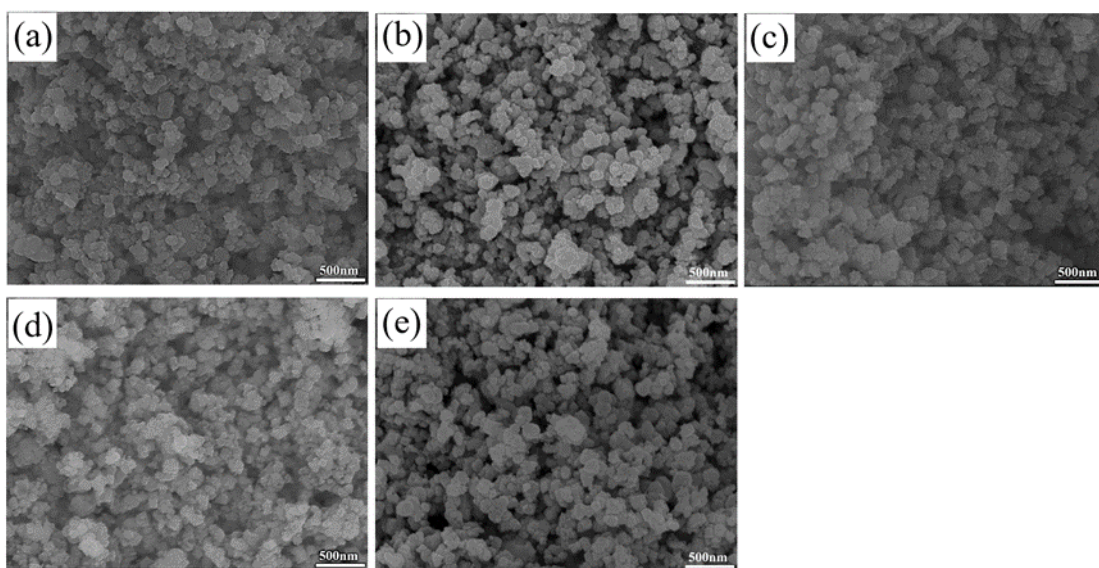


Fig. S1 FE-SEM images of (a) 4CNF-BaTiO₃, (b) 6CNF-BaTiO₃, (c) 8CNF-BaTiO₃, (d) 12CNF-BaTiO₃, and (e) 14CNF-BaTiO₃.

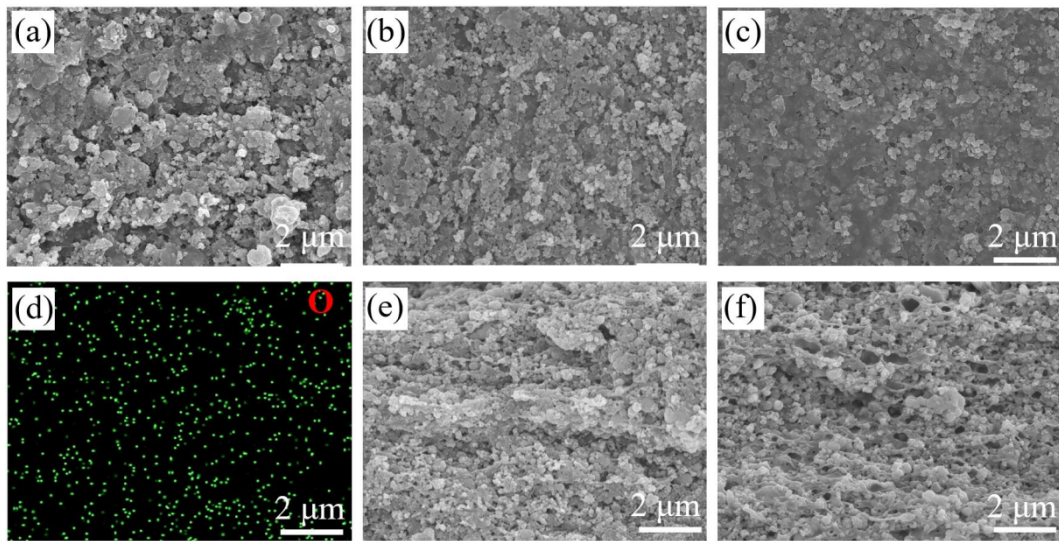


Fig. S2 Surface FE-SEM images of (a) CNF/PVDF/0CNF-BaTiO₃ composite membrane, (b) un-pressing CNF/PVDF/10CNF-BaTiO₃ composite membrane, (c) hot-pressing CNF/PVDF/10CNF-BaTiO₃ composite membrane. (d) EDS diagram of oxygen element at cross section of CNF/PVDF/10CNF-BaTiO₃ composite membrane. FE-SEM at cross section of (e) un-pressing CNF/PVDF/4CNF-BaTiO₃ composite membrane, (f) hot-pressing CNF/PVDF/4CNF-BaTiO₃ composite membrane.

Table S1 Particle size analysis results of CNF-BaTiO₃.

Samples	Grain size	Std Dev	Std Err	PDI
4CNF-BaTiO ₃	276.37	0.52	0.3	0.112
6CNF-BaTiO ₃	255.67	4.35	2.51	0.124
8CNF-BaTiO ₃	281.32	6.88	3.97	0.164
10CNF-BaTiO ₃	304.99	10.67	6.16	0.18
12CNF-BaTiO ₃	262.98	5.3	3.06	0.092
14CNF-BaTiO ₃	294.55	2.87	1.65	0.109
0CNF-BaTiO ₃	320.84	4.97	2.87	0.123

Table S2 XRD analysis results of CNF-BaTiO₃.

Samples	c/a	FWHM of (200)	Average grain size (nm)	Degree of crystallinity (%)
4CNF-BaTiO ₃	1.0044	0.25	38.77±4.91	85.21
6CNF-BaTiO ₃	1.0036	0.25	39.28±5.27	89.68
8CNF-BaTiO ₃	1.0045	0.23	42.07±5.04	78.77
10CNF-BaTiO ₃	1.0029	0.24	41.86±5.06	87.00
12CNF-BaTiO ₃	1.0043	0.24	40.18±3.53	77.47
14CNF-BaTiO ₃	1.0060	0.23	42.71±4.94	82.70
0CNF-BaTiO ₃	1.0032	0.24	40.26±5.54	76.75

Table S3 Tensile performance of different un-pressing composite membranes.

Samples	Tensile strength (MPa)	Elongation at break (%)	Young's modulus E (MPa)
0CNF-BaTiO ₃	14.44±2.7	3.38±1.41	830.94 ± 190.88
4CNF-BaTiO ₃	18.61±3.43	3.08±0.32	1066.75 ± 360.06
6CNF-BaTiO ₃	16.1±2.52	3.15±1.00	974.45 ± 190.00
8CNF-BaTiO ₃	16.62±4.62	3.65±0.94	782.47 ± 143.44
10CNF-BaTiO ₃	18.61±3.75	3.06±1.33	1333.43 ± 618.72
12CNF-BaTiO ₃	15.83±4.63	3.02±0.34	996.30 ± 354.67
14CNF-BaTiO ₃	12.13±3.71	2.92±1.99	852.776 ± 199.93
CNF	24.98±4.17	10.13±3.18	830.94 ± 190.88

Table S4 Tensile performance of different hot-pressing composite membranes.

Samples	Tensile strength (MPa)	Elongation at break (%)	Young's modulus E (MPa)
4CNF-BaTiO ₃	7.46±1.65	5.60±4.34	325.50±157.88
6CNF-BaTiO ₃	6.52±1.70	8.58±4.59	198.68±98.15
8CNF-BaTiO ₃	8.93±1.84	3.85±2.76	444.60±295.24
10CNF-BaTiO ₃	12.91±2.38	5.52±1.74	357.67±162.17
12CNF-BaTiO ₃	17.38±6.11	5.49±1.08	526.86±176.81
14CNF-BaTiO ₃	18.32±3.31	4.66±1.32	664.83±162.60

Table S5 The piezoelectric constant (d_{33}) of PEGs.

Samples incorporating various CNF	4CNF	6CNF	8CNF	10CNF	12CNF	14CNF	0CNF
Thickness of the un-pressing composite membrane (mm)	0.096± 0.005	0.098± 0.005	0.120± 0.004	0.098± 0.011	0.110± 0.009	0.118± 0.012	0.104± 0.004
d_{33} for the un- pressing PEGs ($\text{pC}\cdot\text{N}^{-1}$)	3.00± 0.76	3.88± 1.46	4.13± 1.13	5.25± 1.04	4.50± 0.93	4.38± 1.77	4.28± 1.77

Table S6 The piezoelectric constant (d_{33}) of hot-pressing PEGs.

Samples incorporating various CNF	4CNF	6CNF	8CNF	10CNF	12CNF	14CNF	0CNF
Thickness of the hot-pressing composite membrane (mm)	0.075±0.003	0.083±0.006	0.076±0.002	0.084±0.002	0.087±0.002	0.073±0.002	0.080±0.002
d_{33} for the hot-pressing PEGs ($\text{pC}\cdot\text{N}^{-1}$)	2.60±0.55	3.20±0.45	3.60±0.55	2.80±0.45	2.80±0.44	4.00±1.20	3.20±0.84