Supporting Information

Nanocomposites from Clay, Cellulose Nanofibrils, and Epoxy with Improved Moisture Stability for Coatings and Semistructural Applications

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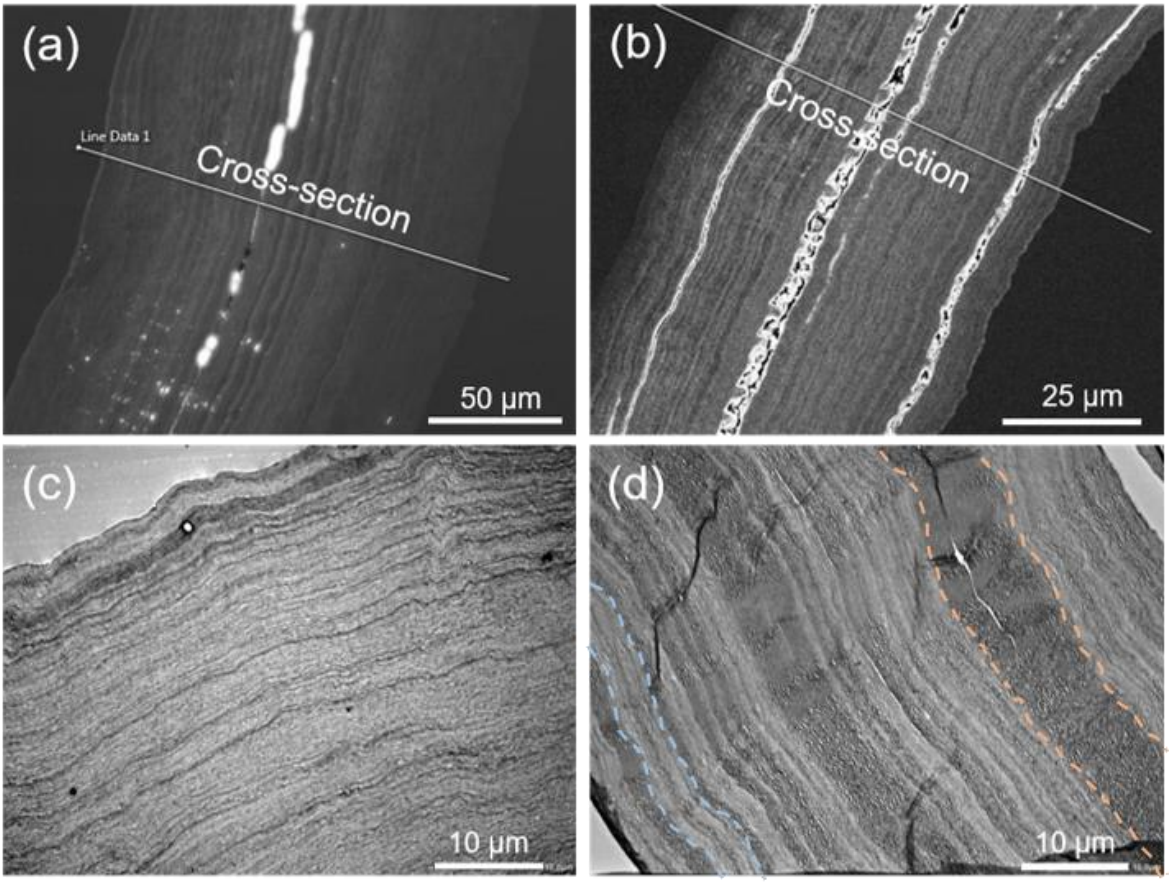

Figure S1. Low-magnification electron images of flat cross-sections. SEM images of (a) CME 30/10/60 (b) CME 35/35/31 composite (5 kV voltage). TEM images of (c) CME 30/10/60 (d) CME 35/35/31 composite. Blue and orange line-delimited regions indicate a polymer-rich and MTM-rich region, respectively. The large cracks are most likely artifacts from the ultramicrotoming method.

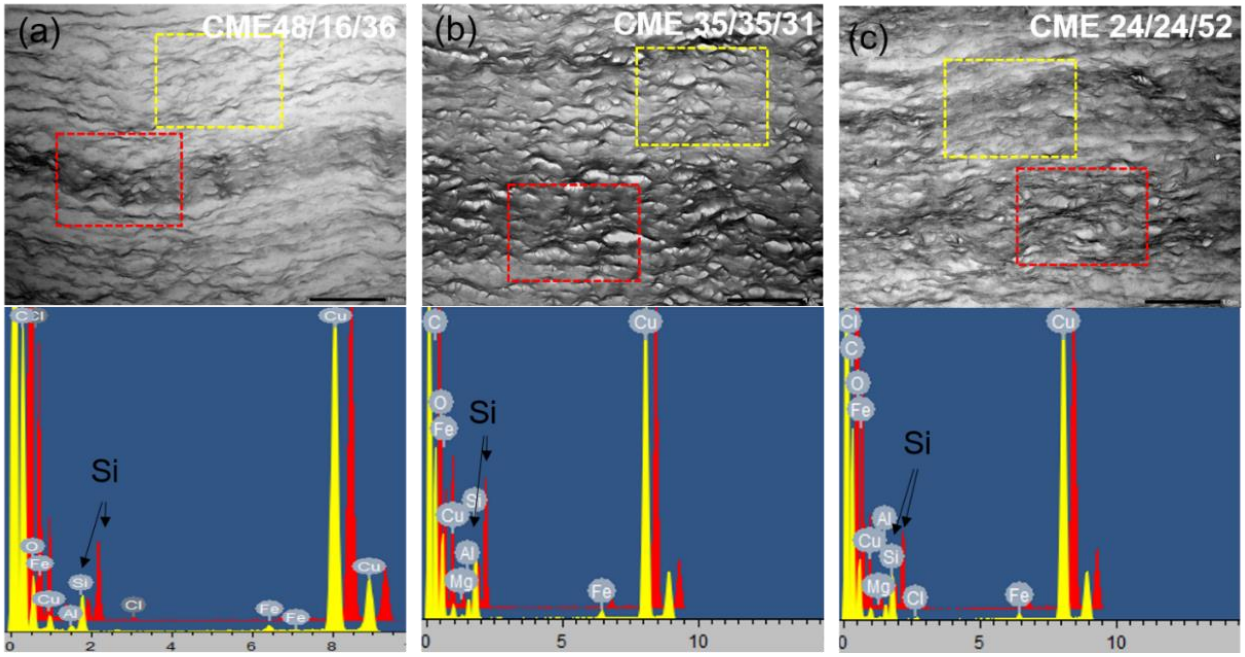

Figure S2. EDX for dark and bright region of TEM image. Si, Al, Fe, Mg are associated with MTM, C is associated with CNF/epoxy, Cu comes from the copper grid used for imaging.

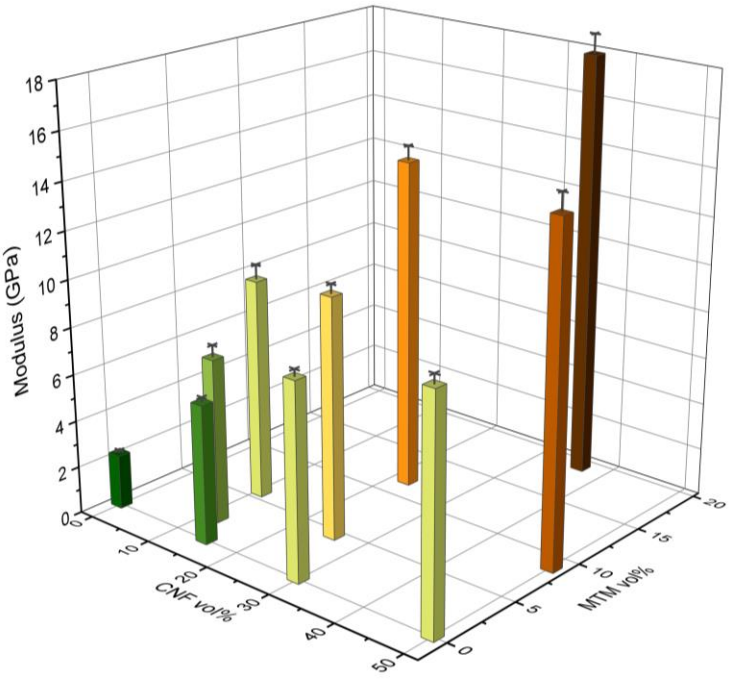

Figure S3. Three-dimensional plot showing the individual effect of MTM and CNF on elastic modulus of the composites, at 50% RH. The 0% MTM data is taken from our earlier work.[1]

Table S1. Mechanical properties for all samples and relative humidities.

Sample	Relative humidity (%)	Young's modulus (GPa)	Ultimate strength (MPa)	Yield strength (MPa)	Strain at break (%)
Neat epoxy	50	2.3 ± 0.1	32 ± 2	*26 ± 3*	35.7 ± 4.9
	90	2.1 ± 0.2	31 ± 6	*25 ± 4*	33.8 ± 6.0
CME 35/35/31	50	18.0 ± 0.8	139 ± 7	*69 ± 1*	2.2 ± 0.1
	90	11.9 ± 0.7	91 ± 8	*54 ± 1*	2.0 ± 0.5
CME 24/24/52	50	14.0 ± 0.6	114 ± 11	*N/A*	1.2 ± 0.3
	90	10.5 ± 1.1	92 ± 4	*75 ± 5*	1.6 ± 0.3
CME 13/13/74	50	9.4 ± 0.6	101 ± 3	*87 ± 3*	1.8 ± 0.2
	90	8.6 ± 0.7	81 ± 2	*77 ± 3*	2.4 ± 0.4
C1M1	50	27.8 ± 0.1	219 ± 12	*156 ± 2*	2.2 ± 0.4
	90	17.9 ± 0.1	154 ± 7	*98 ± 5*	2.4 ± 0.4
CME 48/16/36	50	14.3 ± 0.8	133 ± 1	*59 ± 2*	4.6 ± 0.1
	90	8.5 ± 0.5	111 ± 7	*38 ± 1*	6.6 ± 0.4
CME 30/10/60	50	10.2 ± 0.4	110 ± 3	*84 ± 5*	2.3 ± 0.2
	90	8.5 ± 0.2	94 ± 3	*72 ± 2*	4.6 ± 0.1
CME 15/5/80	50	7.0 ± 0.5	107 ± 6	*80 ± 6*	6.4 ± 0.4
	90	6.9 ± 0.4	98 ± 3	*70 ± 5*	11.2 ± 0.6
C3M1	50	24.7 ± 0.1	280 ± 10	*162 ± 7*	4.3 ± 0.2
	90	15.3 ± 0.1	189 ± 4	*91 ± 4*	4.4 ± 0.3

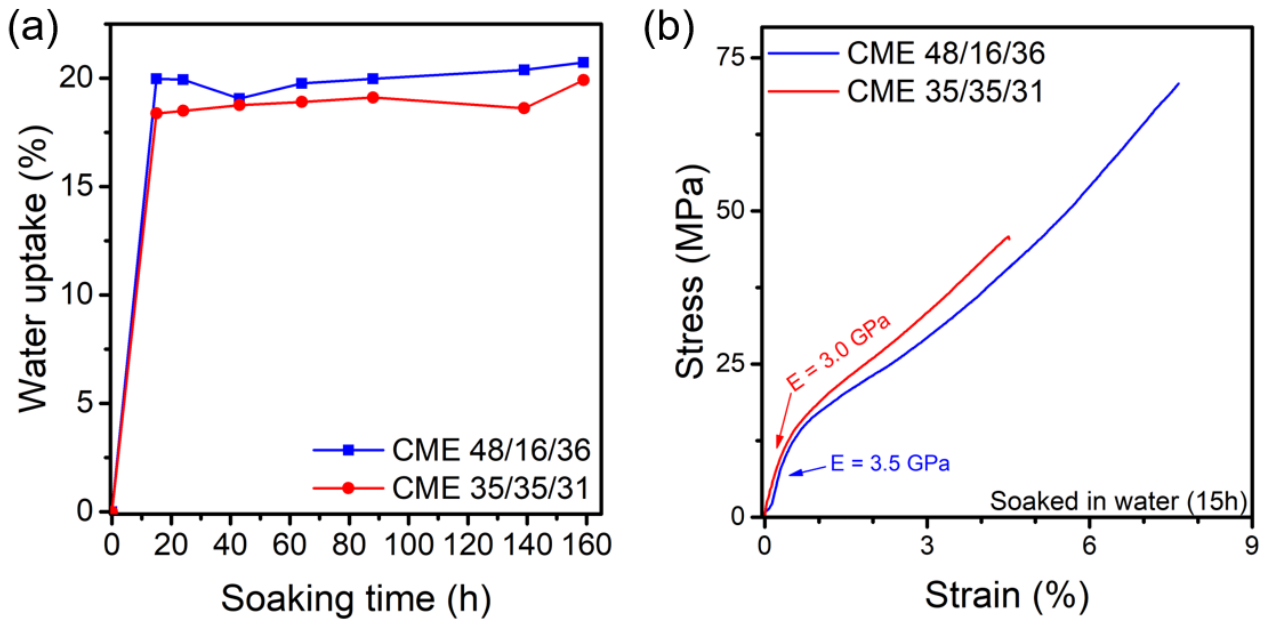

Figure S4. (a) Time-dependent water uptake of composites after immersion in Milli-Q water for 7 days. (b) Representative tensile stress-strain curves of high reinforcement fraction composites after immersion in Milli-Q water for 15 h.

Table S2. TGA peaks and residues in air and nitrogen atmospheres.

Sample	T_{onset} °C	T_{peak} °C	Residue at 800 °C, wt %	Organic residue at 800 °C, wt %
CME 33/33/33 (N_2)	310	349	38	10
CME 50/17/33 (N_2)	307	350	26	12
CME 33/33/33 (Air)	308	349	32	4
CME 50/17/33 (Air)	296	350	20	5

T_{onset} is defined as the temperature at which 10% weight loss is reached. The organic residue was calculated assuming 85% residue for MTM at 800 °C in N_2 and air.

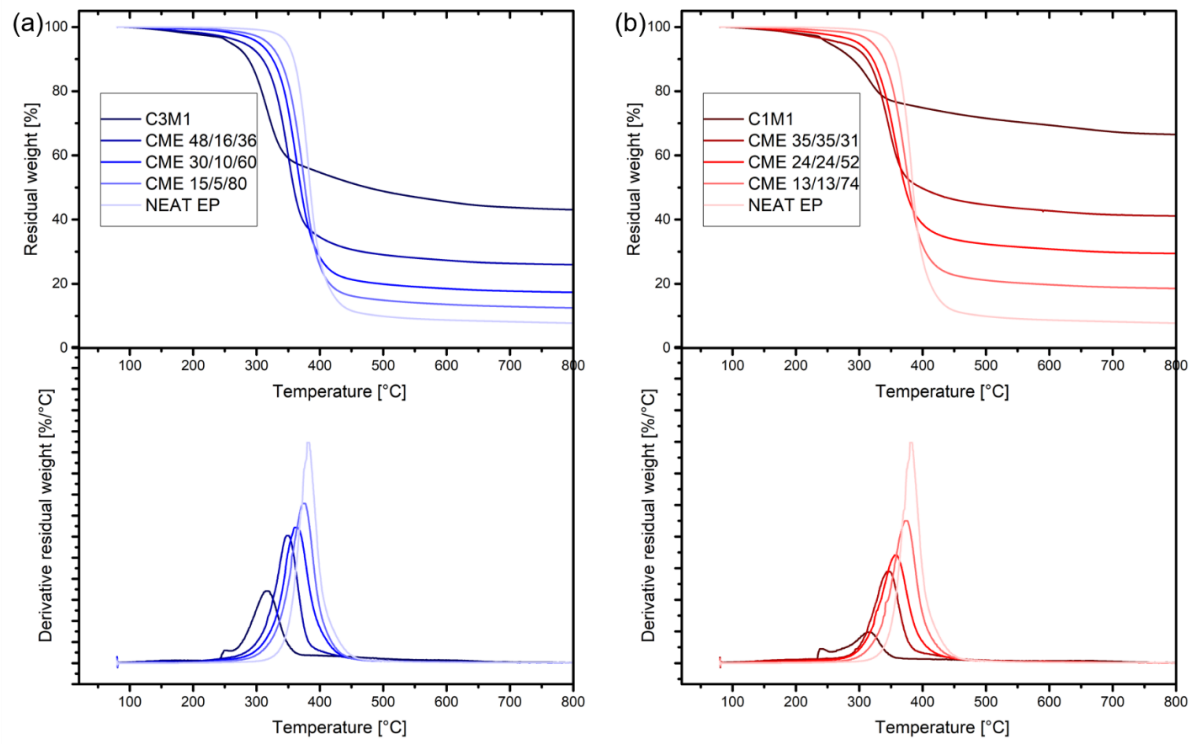

Figure S5. TGA curves in nitrogen atmosphere for all CME composites. (a) C3M1 series and (b) C1M1 series.

Table S3. Vertical flammability burning parameters (burning time and residues).

Sample	Burning time, s	Residues (wt %)
Neat EP	37 ± 12	No residues
CME 33/33/33	4.7 ± 0.6	43 ± 5
CME 50/17/33	5.3 ± 0.6	30 ± 1

REFERENCES

1. Ansari, F.; Galland, S.; Johansson, M.; Plummer, C. J. G.; Berglund, L. A., Cellulose nanofiber network for moisture stable, strong and ductile biocomposites and increased epoxy curing rate. *Composites Part A: Applied Science and Manufacturing* **2014,** *63*, 35-44.