Supplementary Information

Sustainable Barrier Coatings for Food Packaging with a Built-in, Redox-activated Trigger for Surface Hydrophilization

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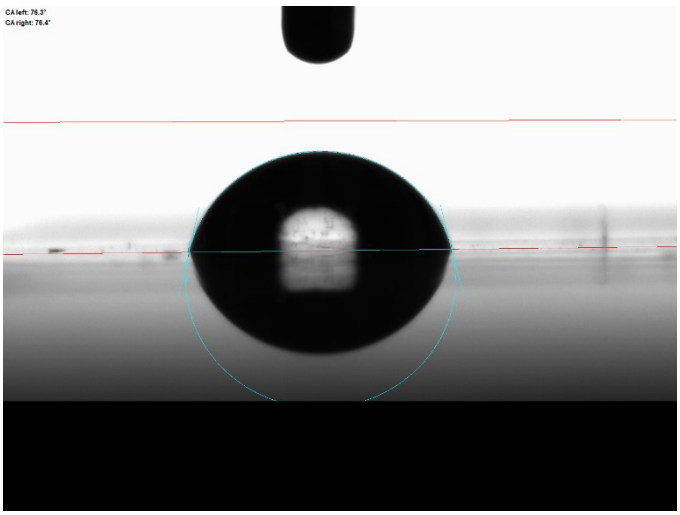

Figure S1. Contact angle of neat PLA substrate.

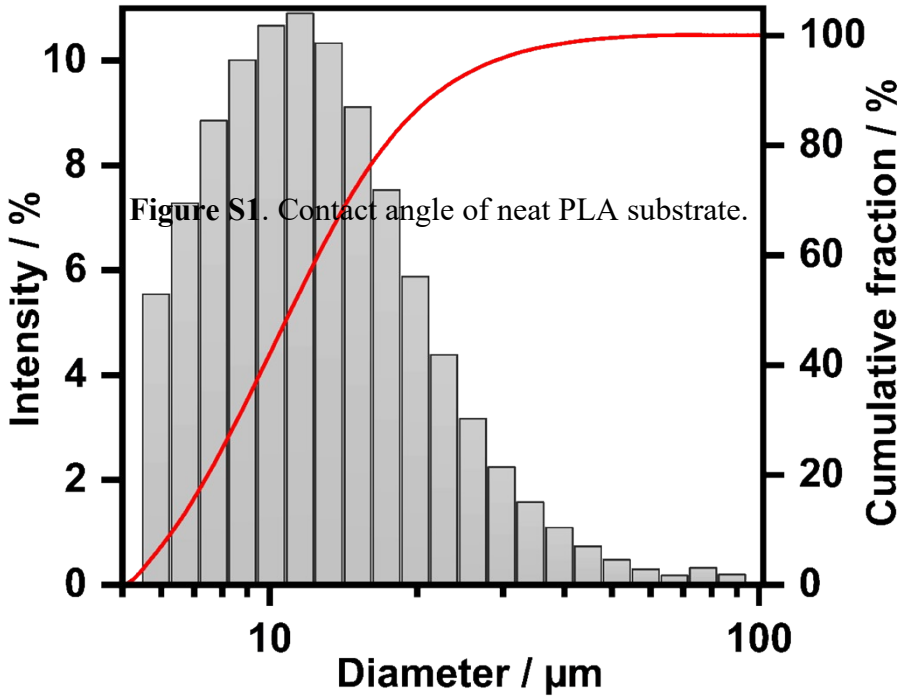

Figure S2. Number weighted size distribution indicating a mean size of 9 µm as determined C4VMT nanosheets by SLS.

Component	CS (mm/s)	QS (mm/s)	Area (%)
Fe^{III}	0.390 ± 0.004	1.550 ± 0.007	78.12 ± 1.14
Fe^{II}	1.330 ± 0.005	2.860 ± 0.009	21.88 ± 1.14

Table S1. Mößbauer hyperfine parameters of spectra of the C4VMT nanosheets after being suspended in the culture medium (corresponding to Figure 3b) with *Geobacter metallireducens*, showing the partial reduction of structural Fe(III) to Fe(II).

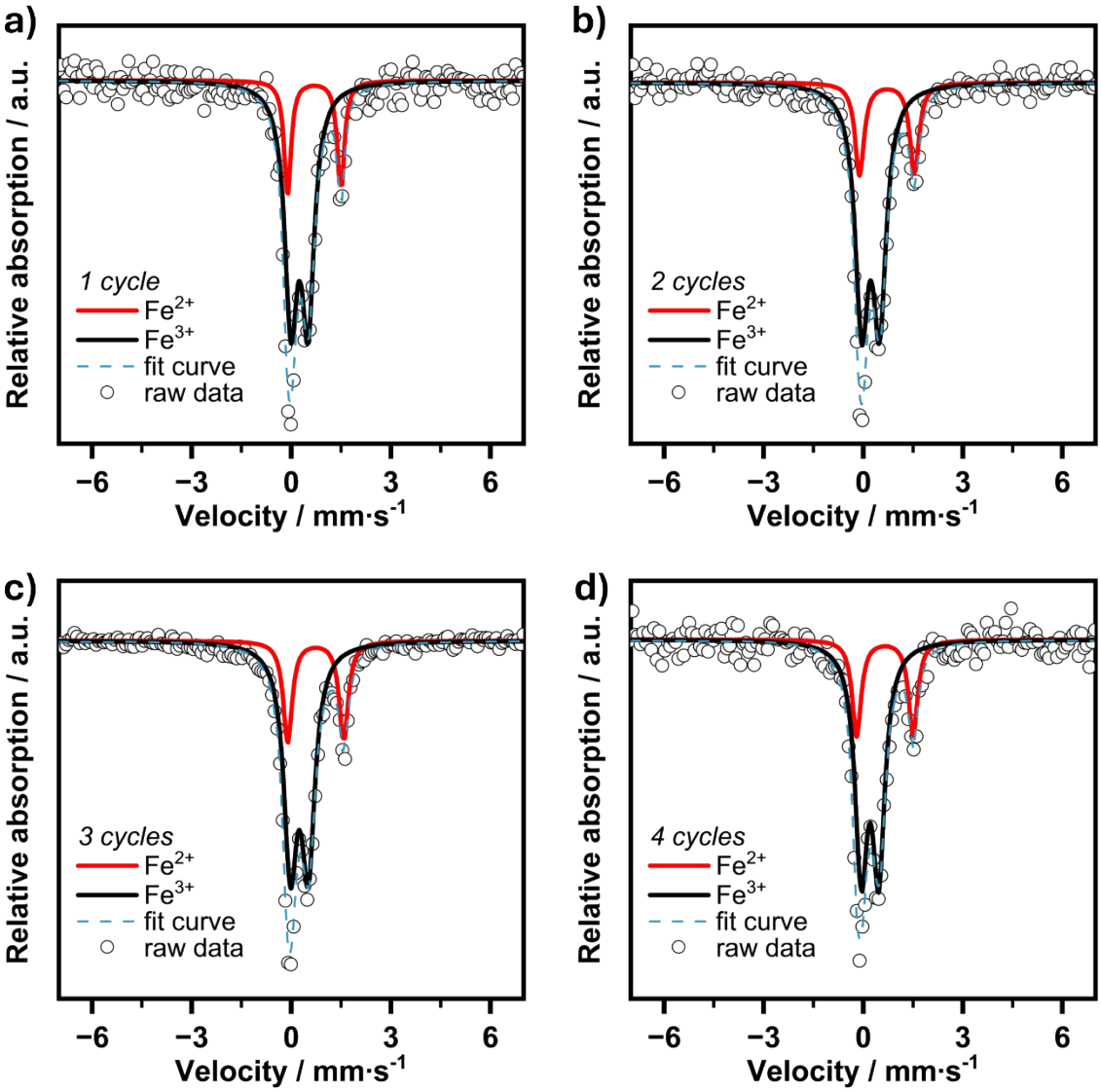

Figure S3. Mößbauer spectra of the C4VMT/$PNAT_{30}$ coatings after being suspended in the culture medium with *Geobacter metallireducens* (corresponding to Figure 3c), showing the partial reduction of structural Fe(III) to Fe(II).

Component	CS (mm/s)	QS (mm/s)	Area (%)
Fe^{III} 0 cycle	0.380 ± 0.003	1.440 ± 0.006	100
Fe^{III} 1 cycle	0.364 ± 0.003	1.501 ± 0.003	78.71 ± 3.48
Fe^{II} 1 cycles	1.512 ± 0.004	2.881± 0.008	21.29 ± 3.48
Fe^{III} 2 cycles	0.342 ± 0.006	1.482 ± 0.002	77.96 ± 3.08
Fe^{II} 2 cycles	1.448 ± 0.004	2.754 ± 0.008	22.04 ± 3.08
Fe^{III} 3 cycles	0.358 ± 0.005	1.491 ± 0.004	75.65 ± 1.37
Fe^{II} 3 cycles	1.428 ± 0.002	2.846 ± 0.003	24.05 ± 1.37
Fe^{III} 4 cycles	0.402 ± 0.003	1.511 ± 0.011	75.82 ± 3.29
Fe^{II} 4 cycles	1.508 ± 0.002	2.796 ± 0.011	24.18 ± 3.29

Table S2. Mößbauer hyperfine parameters of the C4VMT/$PNAT_{30}$ coatings after being suspended in the culture medium with *Geobacter metallireducens* with time differences.

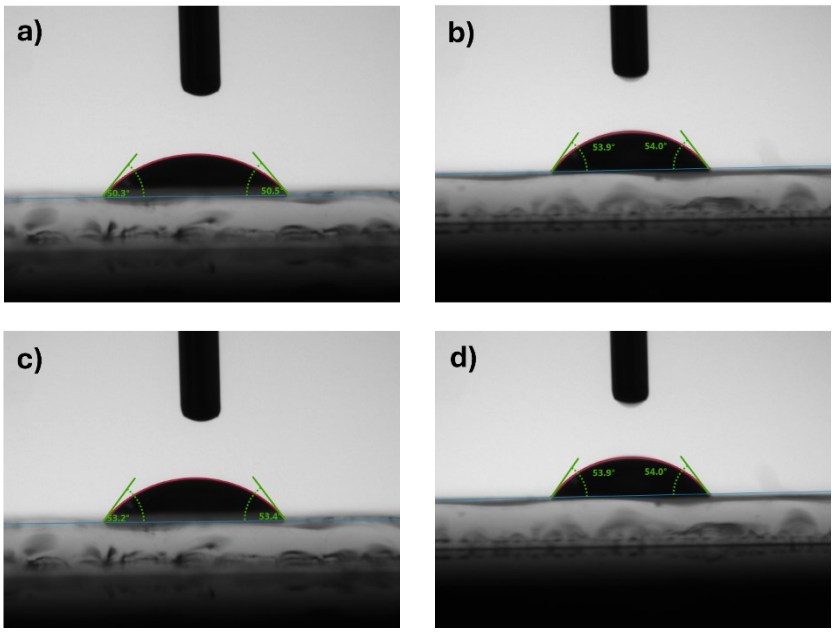

Figure S4. Contact angle of the pristine C4VMT/$PNAT_{30}$ coatings.

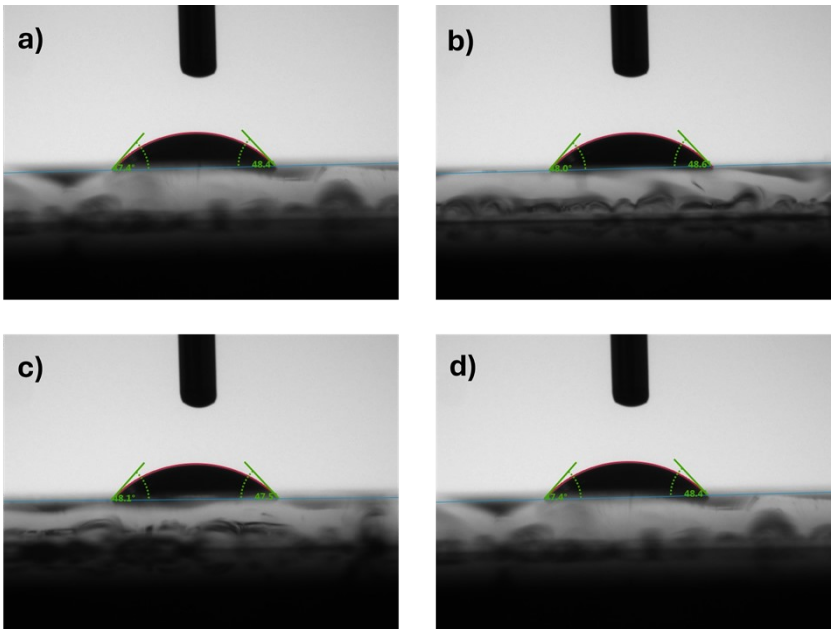

Figure S5. Contact angle of the C4VMT/PNAT$_{30}$ coatings after 1 redox cycle.

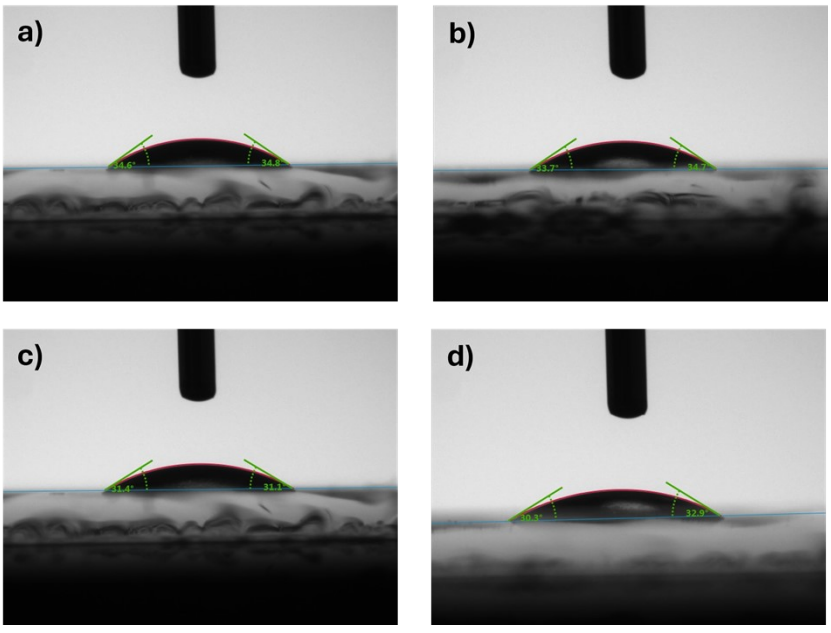

Figure S6. Contact angle of the C4VMT/PNAT$_{30}$ coatings after 2 redox cycles.

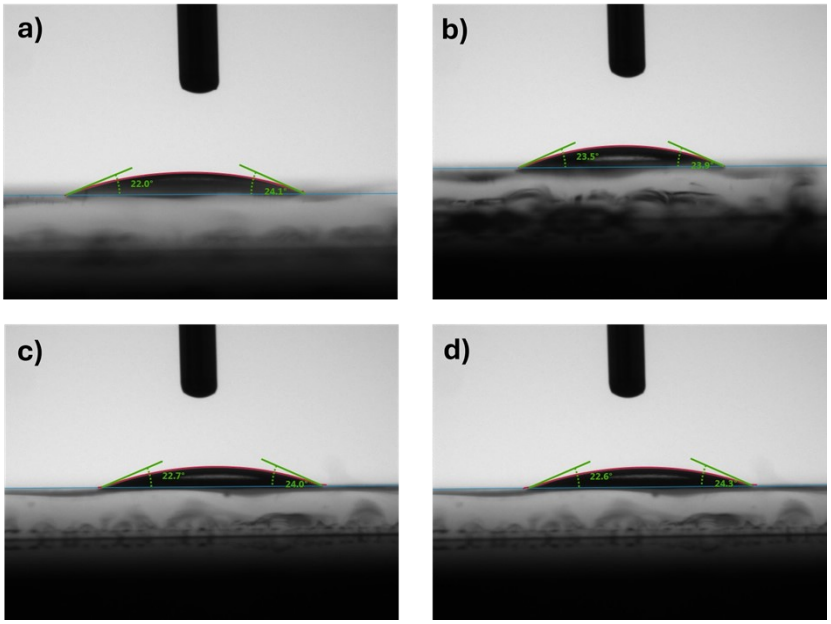

Figure S7. Contact angle of the C4VMT/PNAT$_{30}$ coatings after 4 redox cycles.

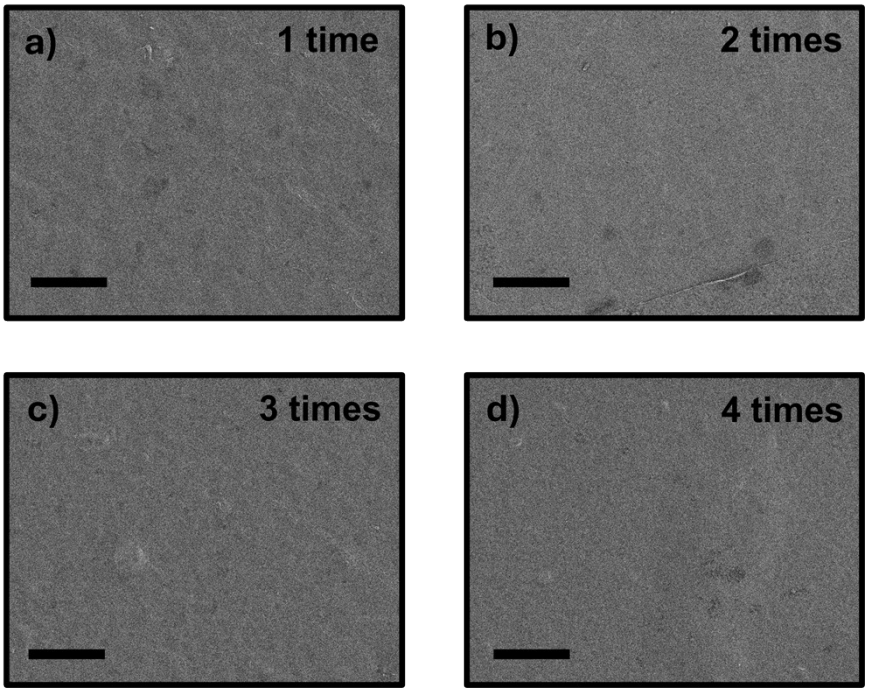

Figure S8. SEM images of the surface of the barrier coating and after a) 1, b) 2, c) 3, and d) 4 exposed times to the culture medium without inculation of *Geobacter metallireducens.* Scale bar: 100 µm.

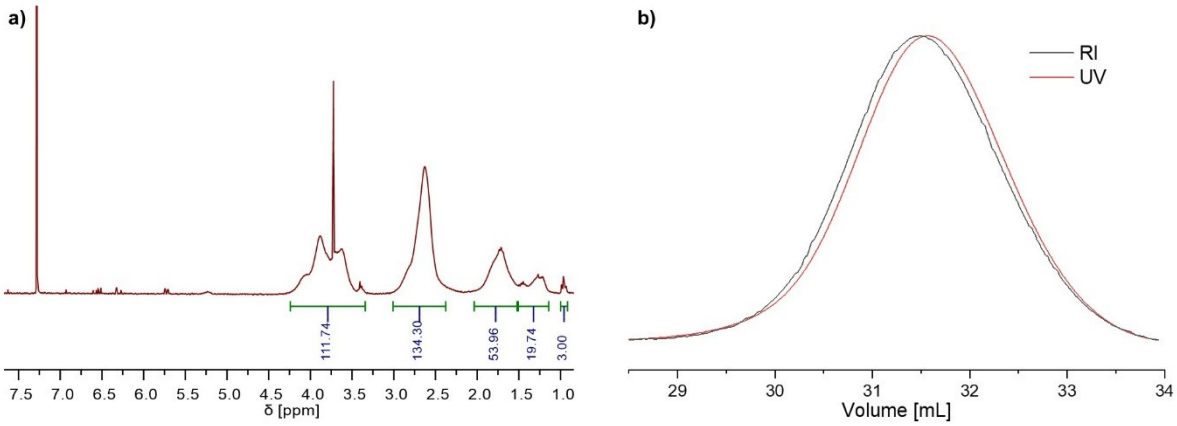

Figure S9. a) ^{1}H NMR spectrum and b) SEC trace (RI and UV detector) of $PNAT_{30}$ after drying.