Electronic Supplementary Material (ESI) for Soft Matter.


# Supporting Information

## Cathodic electrodeposition of organic nanocomposite coatings reinforced with cellulose nanocrystals

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**Table S1.** Mixtures used for dispersibility tests of CNC in polyacrylate.

| Sample ID | pH | σ (S/cm) | [PA] (%) | [CNC] (%) | Mass ratio CNC: PA | Size (nm) | PdI | Mobility (μmcm/Vs) |
|---|---|---|---|---|---|---|---|---|
| Pristine CNC (Batch 1) | 4.396 | 381.6 | - | 4.06 | - | 80.1 ± 0.2 | 0.619 | -5.138 ± 0.211 |
| Pure PA (Batch 1) | 4.654 | 712.2 | 10 | 0 | 0 | 130.2 ± 0.3 | 0.166 | 5.066 ± 0.121 |
| | 4.585 | 664.8 | 8.3 | 0 | 0 | 130.4 ± 0.7 | 0.175 | 5.078 ± 0.079 |
| PA-CNC-0.05 | 4.574 | 765.6 | 8.3 | 0.050 | 0.006 | 130.7 ± 0.8 | 0.173 | 5.149 ± 0.089 |
| PA-CNC-0.125 | 4.579 | 710.5 | 8.3 | 0.125 | 0.015 | 131.8 ± 0.6 | 0.187 | 5.142 ± 0.161 |
| PA-CNC-0.25 | 4.564 | 716.6 | 8.3 | 0.250 | 0.030 | 137.6 ± 0.6 | 0.176 | 4.739 ± 0.063 |
| PA-CNC-0.5 | 4.562 | 805.1 | 8.3 | 0.500 | 0.060 | 138.5 ± 1.0 | 0.213 | 3.875 ± 0.114 |

Size and mobility analysis was performed at 0.1% of mixture in DI $H_2O$.

PA-CNC mixtures sonicated at 2400 J/g.

**Table S2.** Mixtures used for dispersibility tests of DA monomer in PA-CNC system.

| Sample ID | pH | σ (S/cm) | [PA] (%) | [CNC] (%) | [DA] (mM) | Size (nm) | PdI | Mobility (μmcm/Vs) |
|---|---|---|---|---|---|---|---|---|
| Pristine CNC (Batch 2) | 5.374 | 168.9 | 0 | 2.84 | 0 | 126.0 ± 0.9 | 0.304 | -4.978 ± 0.300 |
| Pure PA (Batch 2) | 4.746 | 744.5 | 9.0 | 0 | 0 | 78.5 ± 0.4 | 0.179 | 4.164 ± 0.021 |
| | 4.784 | 719.0 | 8.3 | 0 | 0 | 79.1 ± 0.4 | 0.168 | 4.475 ± 0.018 |
| PA-CNC | 4.816 | 623.0 | 8.3 | 0.125 | 0 | 82.6 ± 0.5 | 0.192 | 4.875 ± 0.117 |
| PA-CNC-DA | 4.903 | 1216.9 | 8.3 | 0.125 | 5 | 81.5 ± 1.5 | 0.213 | 4.564 ± 0.135 |

Size and mobility analysis was performed at 0.1%of mixture in DI $H_2O$.

PA-CNC-DA mixtures sonicated at 2400 J/g.

**Table S3.** Characteristics of coating suspensions used for EPD on HSLA substrates.

| Sample[a,b] | [CNC](%) | [PA] (%) | pH | σ (S/cm) | Size (nm)[c] | PdI[(c)] | Mobility (μmcm/Vs) [c] | ζ potential (mV) [c] |
|---|---|---|---|---|---|---|---|---|
| CNC | | | | | | | | -59.4 ± 2.6 |
| PA-1 | 0 | 8.3 | 4.849 | 770.4 | 122.3 ± 0.2 | 0.162 | 5.051 ± 0.156 | 64.4 ± 2.0 |
| PA-2 | 0 | 8.3 | 4.901 | 724.2 | 61.1 ± 0.6 | 0.185 | 4.978 ± 0.085 | 63.5 ± 1.1 |
| PA-3 | 0 | 8.3 | 4.784 | 719.0 | 79.1 ± 0.4 | 0.168 | 4.475 ± 0.018 | 57.1 ± 0.2 |
| PA-CNC-1 | 0.125 | 8.3 | 5.061 | 769.7 | 78.0 ± 0.9 | 0.305 | 4.759 ± 0.155 | 60.7 ± 1.9 |
| PA-CNC-2 | 0.125 | 8.3 | 4.869 | 777.5 | 69.8 ± 0.3 | 0.252 | 3.616 ± 0.119 | 46.1 ± 1.5 |
| PA-CNC-3 | 0.125 | 8.3 | 5.299 | 672.8 | 90.9 ± 3.0 | 0.272 | 3.587 ± 0.049 | 45.8 ± 0.6 |
| PA-CNC-PDA-1 | 0.125 | 8.3 | 5.091 | 1075.1 | 79.1 ± 0.6 | 0.317 | 3.674 ± 0.090 | 46.9 ± 1.2 |
| PA-CNC-PDA-2 | 0.125 | 8.3 | 4.805 | 1111.9 | 91.4 ± 0.4 | 0.408 | 3.665 ± 0.189 | 46.7 ± 2.4 |
| PA-CNC-PDA-3 | 0.125 | 8.3 | 5.052 | 1057.0 | 88.35 ± 0.8 | 0.273 | 3.249 ± 0.123 | 41.5 ± 1.6 |

[a] Pristine CNC (Batch 2) was used for these sets of experiments.

[b] Dopamine concentration in the mixture was set at 5 mM and PA concentration at 8.3%.

[c] DLS and ELS analysis were performed at 0.1% of mixture in DI $H_2O$.

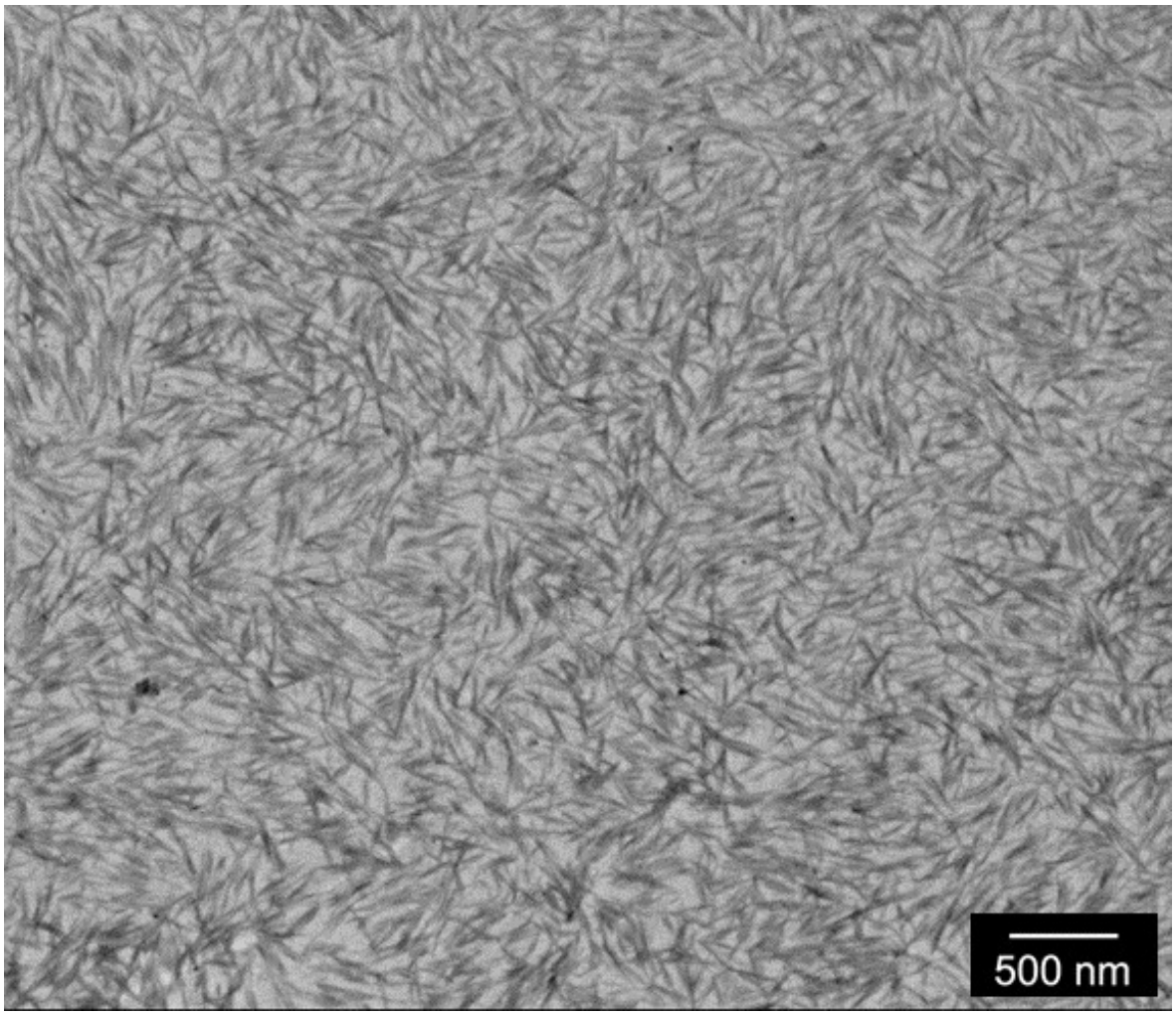


**Fig. S 1.** SEM image showing the good dispersion of sulfated CNC crystallites onto a glass slide.

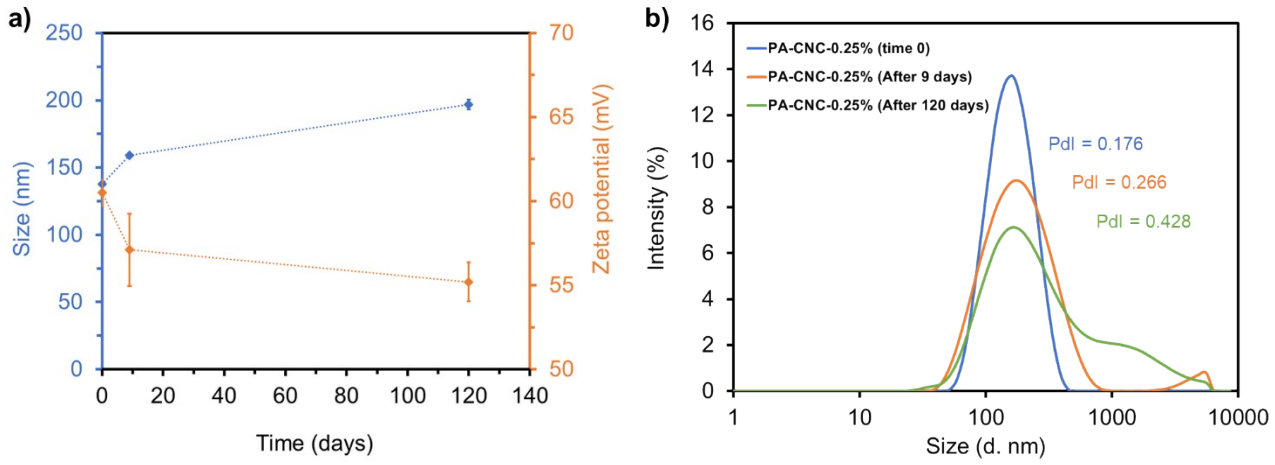


**Fig. S2.** Effect of storage time on ζ-potential (a) and particle size distribution (b) of PA-CNC suspension at 0.25% CNC concentration sonicated at 2400 J/g.

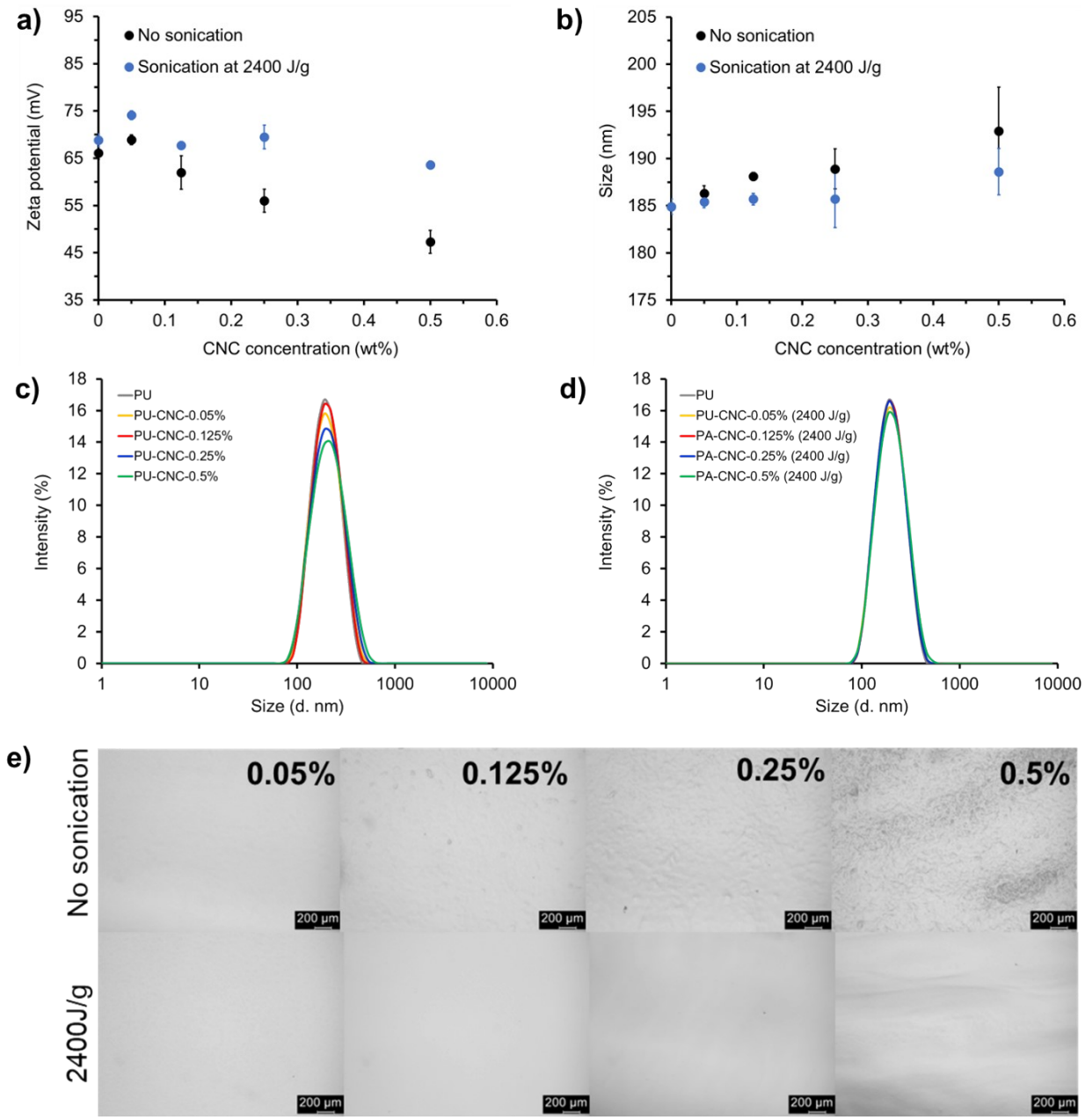


**Fig. S3.** ζ-potential (a) and particle size (b) of Polyurethane-CNC as a function of CNC concentration before and after sonication at 2400 J/g. Size distribution of PU-CNC suspensions at different CNC concentrations before (c) and after (d) sonication at 2400 J/g. (e) Optical microscopy images of the PU-CNC cast films on glass slides.

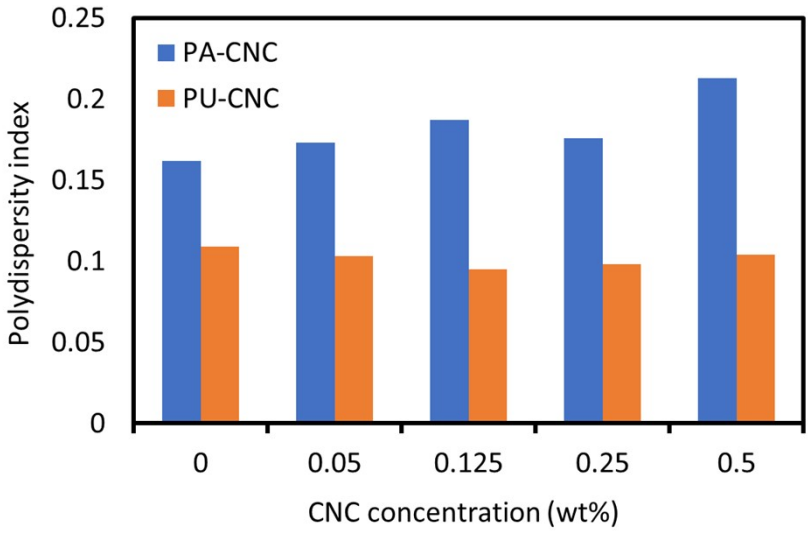


**Fig. S4.** Polydispersity index of PA-CNC and PU-CNC sonicated at 2400 J/g.

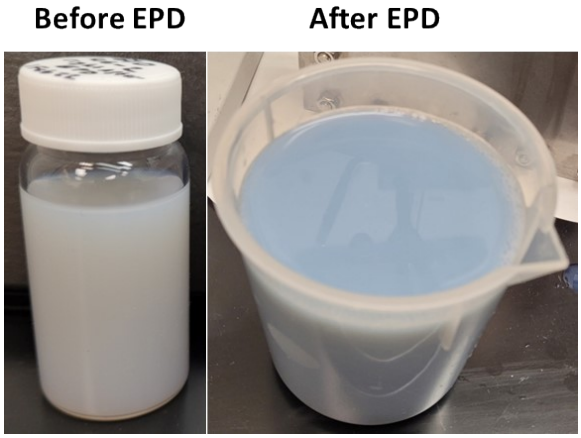


**Fig. S5.** Appearance of PA-CNC-PDA mixture before and right after the EPD process.

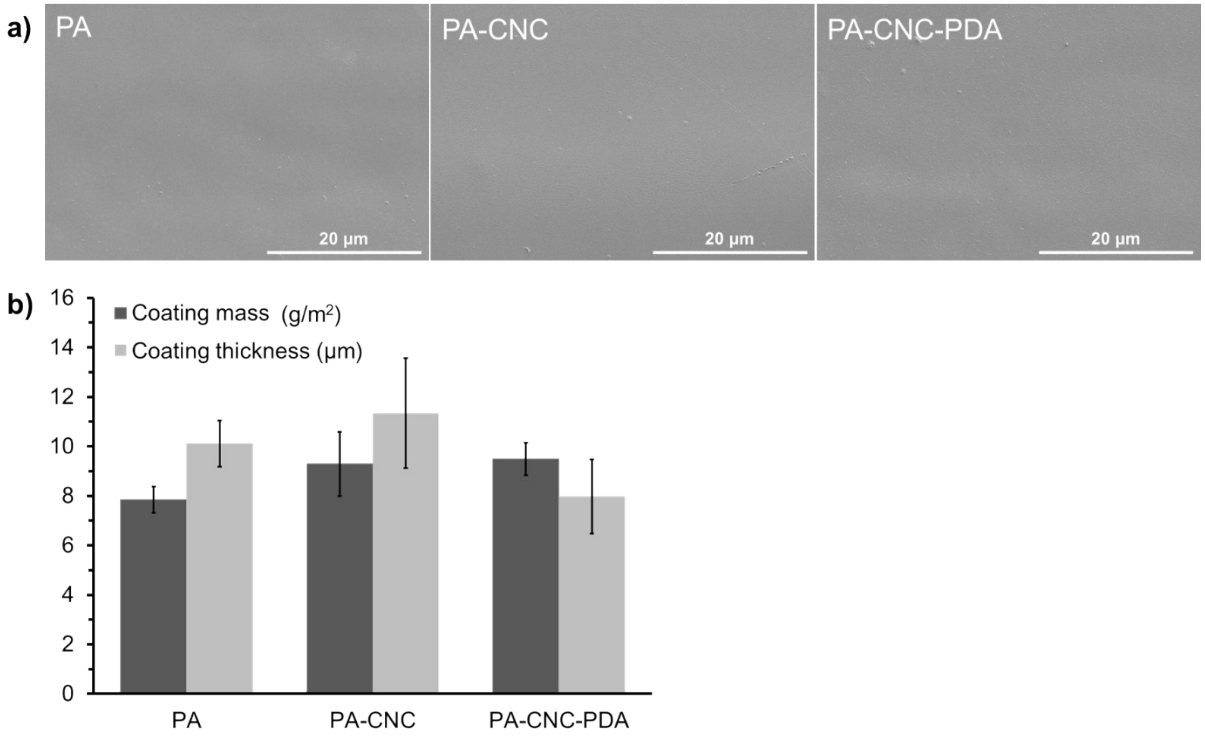


**Fig. S6.** (a) SEM topography images taken at low magnification of the surface of PA, PA-CNC and PA-CNC-PDA nanocomposite coatings, and (b) their deposit mass and thickness.

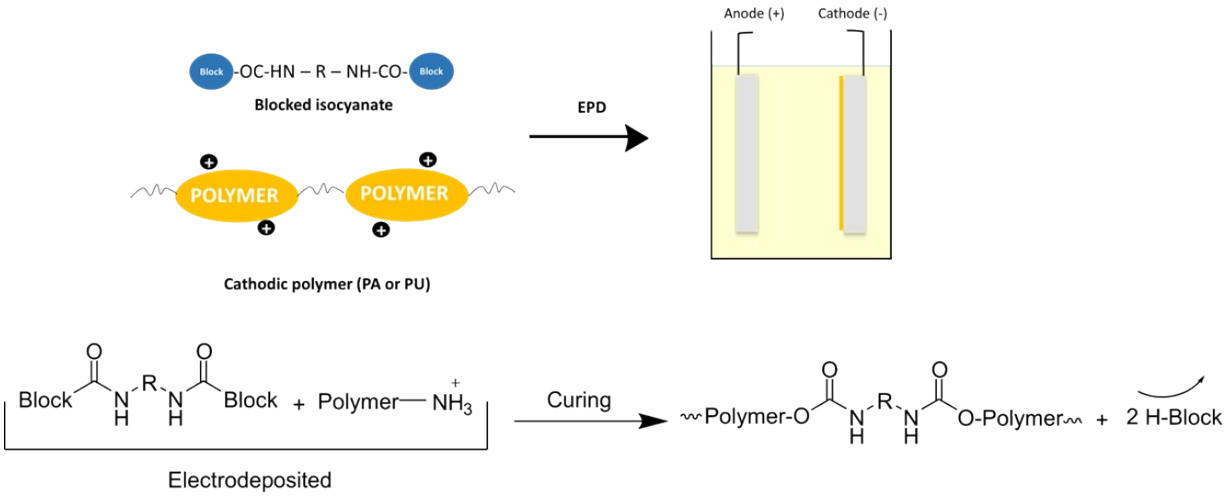


**Fig. S7.** Curing mechanism of the electrodeposited cathodic polymer and blocked isocyanate. When the blocked isocyanates are exposed to high temperature during curing, the blocking entities are released and the reactive NCO groups of the deblocked isocyanate will bond to OH or NH groups of the cathodic polymer.

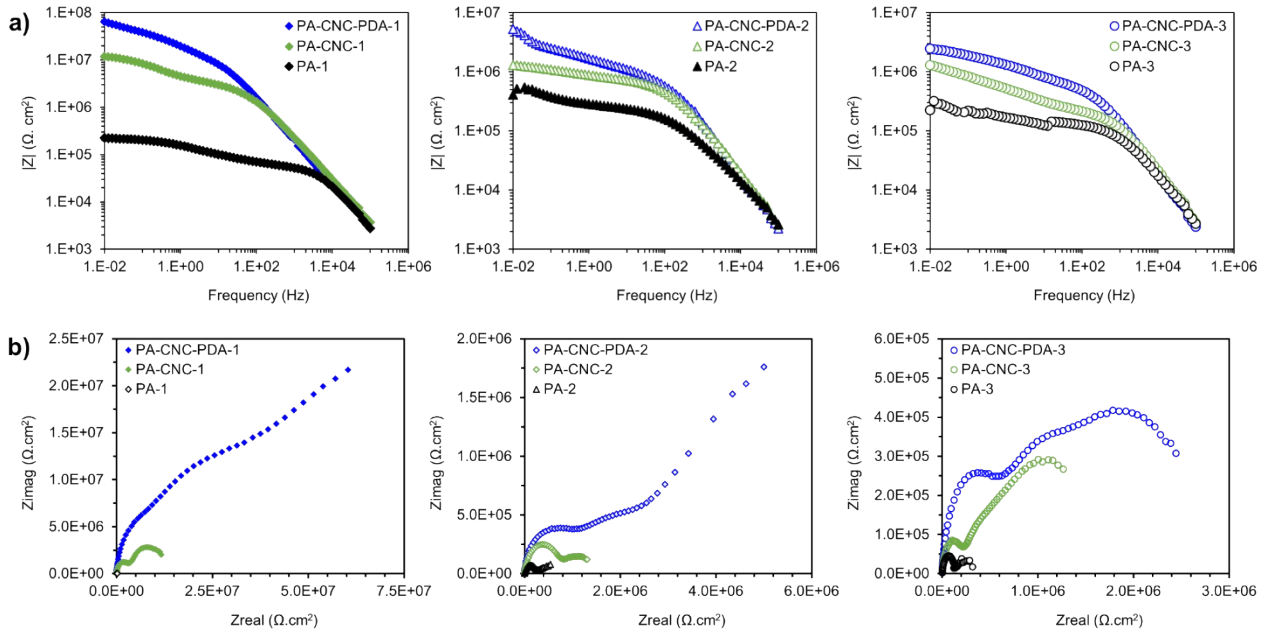


**Fig. S8**. (a) Bode and (b) Nyquist plots measured in 0.1M NaCl after 5 days of electrolyte exposure for PA, PA-CNC and PA-CNC-PDA coating replicates prepared using three different stock PA suspensions.