Supporting Information

Unveiling the Impact of Crosslinking Redox-Active Polymers on Their Electrochemical Behavior by 3D Imaging and Statistical Microstructure Analysis

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Figure S1. Synthesis scheme for yielding (crosslinked) PTMA with m = 0 for G-PTMA and $m \neq 0$ for crosslinked XG-PTMA.



Figure S2. (a) FTIR spectra of G-PTMA (blue), XG-PTMA (green) and the cross-linker, ethylene glycol dimethacrylate (EGDMA; black). (b) Comparison of the FTIR spectra of EGDMA (black) and poly-EGDMA (orange). (c) DSC data recorded for (X)G-PTMA, conducted with a scan rate of $5 \square \min^{-1}$ during the second heating cycle.



Figure S3. Example cutout of the hand-labeled training data of the sample with G-PTMA-CMK-8 on the left and the same cutout labeled by Ilastik on the right.



Figure S4. Distribution of the local surface area per unit volume of G-PTMA-CMK-8 (left) and XG-PTMA-CMK-8 (right)



Figure S5. (a) Galvanostatic cycling of XG-PTMA-SC65 (green) and G-PTMA-SC65 electrodes (blue) at a constant specific current of 50 mA g⁻¹. The dis-/charge profile of the 2nd cycle is displayed in panel (b) for XG-PTMA-SC65 and panel (c) for G-PTMA-SC65.



Figure S6. Basic physicochemical characterization of CMK-8 via (a,b) SEM at two different magnifications, (c) BET, and (d) BJH analysis.



Figure S7. (a,b) Charge and (c,d) discharge profiles of galvanostatically cycled (a,c) G-PTMA-CMK-8 and (b,d) XG-PTMA-CMK-8 electrodes at specific currents varying from 0.2 to 10 A g⁻¹. Plots of the average charge and discharge voltage vs. the applied specific current (logarithmic) for (e) G-PTMA-CMK-8 and (f) XG-PTMA-CMK-8; the inset shows the total dis-/charge hysteresis.