

## Supporting Information

### **Porous carbon-carbon composite electrodes synthesized by twin polymerization for vanadium redox flow batteries**

Maïke Schnucklake<sup>a</sup>, Lysann Kaßner<sup>b</sup>, Michael Mehring<sup>b</sup>, Christina Roth<sup>c</sup>

<sup>a</sup>*Institute of Chemistry and Biochemistry, Freie Universität Berlin, Arnimallee 22, D-14195 Berlin, Germany.*

<sup>b</sup>*Coordination Chemistry, Technische Universität Chemnitz, D-09107 Chemnitz, Germany.*

<sup>c</sup>*Electrochemical process engineering, Universität Bayreuth, Universitätsstraße 30, D-95447 Bayreuth, Germany.*

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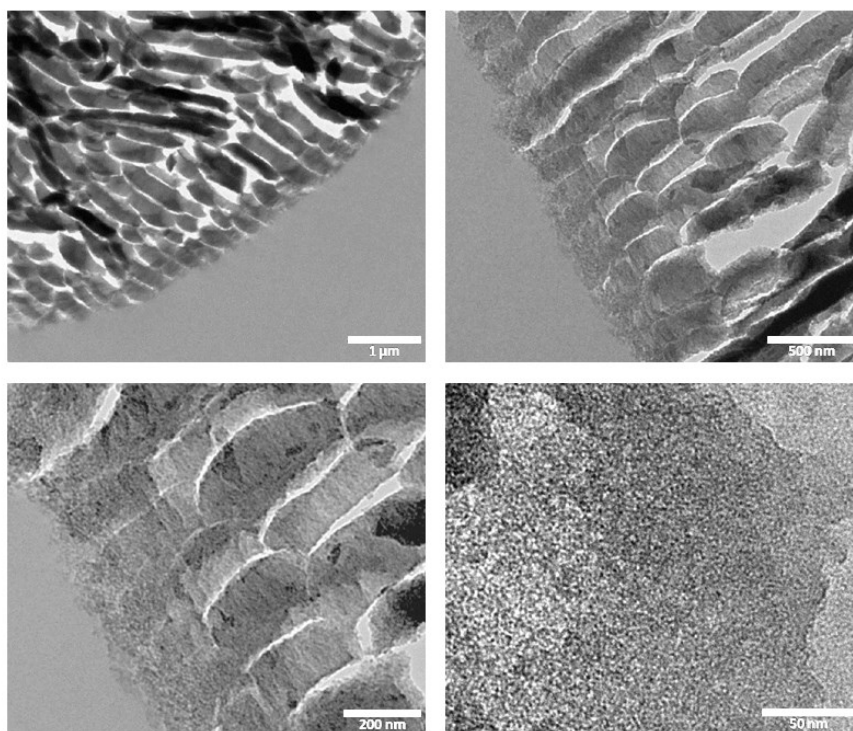
S7. Cyclic voltammetry

## S1. Experimental Details

**Table S1:** Overview of experiments with used and determined masses respectively volumes of solvents and catalysts.

	composite (MSA, 1h, E1)	composite (T, 1h, E1)	carbonized CF	composite (MSA, 1h, E1)
$m_{\text{felt}}$ (g)	0.166	0.1693	0.3477	0.7979
$m_{\text{TM}}$ (g)	0.0867	0.0943		0.4
ratio felt / twin monomer	1.9	1.8	-	2.0
$V_{\text{DCM}}$ (mL)	33	2	-	160
$m_{\text{MSA}}$ (mg)	40	-	-	192
$V_{\text{toluene}}$ (mL)	33	-	-	120
$V_{\text{toluene}}$ for twin monomer (mL)	8	-	-	40
$m_{\text{felt,polymer}}$ (g)	0.2123	-	-	1.0994
ratio felt / polymer	3.6	-	-	2.7
residue after carbonization (%)	47.5	45.8	51.2	45.1
etched silica (%)	26.5	85.1	-	20.8

## S2. TEM images



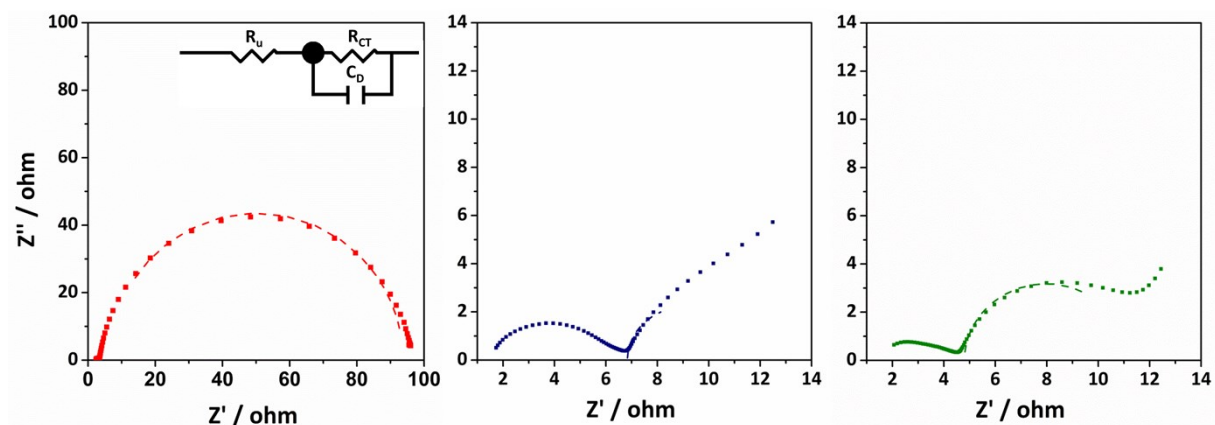
**Figure S1:** TEM images of the acidic catalyzed composite electrode in different magnification. The images show cross-sections of the macroporous carbon fibers. At the edges of the elongated scales, especially at high resolution, it seems like there is amorphous carbon material with small domain size present (which is a characteristic feature for the hybrid material synthesized via twin polymerization).

### S3. Electrochemical data

**Table S2:** Electrochemical data for the positive and negative redox reaction in VRFB.

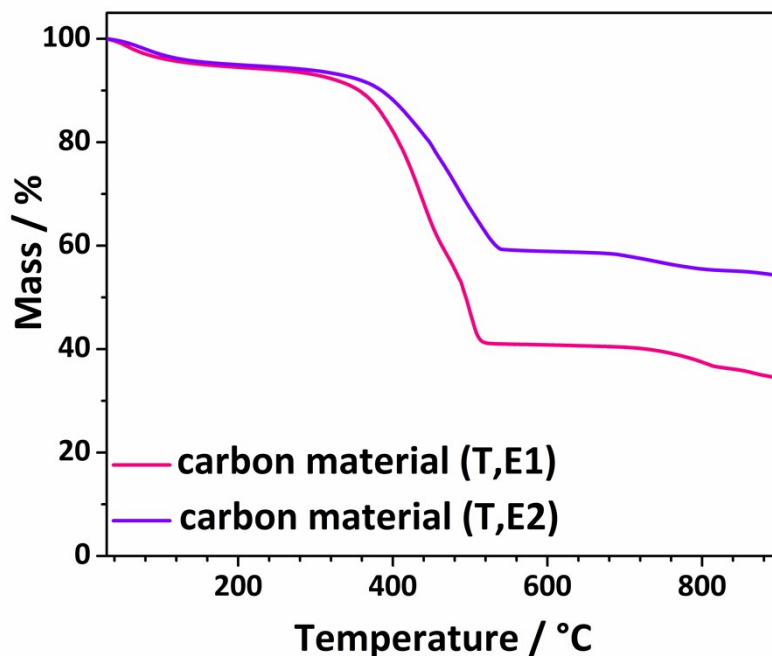
	redox couple	peak current $i_p$ / mA	peak position $E_p$ / mV	peak separation $\Delta E_p$ / mV	peak current ratio $I_A/I_C$
pristine	$VO^{2+}/VO_2^+$	-	-	-	-
		-	-		
carbonized CF	$VO^{2+}/VO_2^+$	9.1	965.1	229.0	1.69
		-5.4	736.1		
composite (T)	$VO^{2+}/VO_2^+$	7.5	911.2	149.8	1.47
		-5.1	761.4		
composite (MSA)	$VO^{2+}/VO_2^+$	9.1	898.5	128.4	1.28
		-7.1	770.1		
pristine	$V^{2+}/V^{3+}$	-	-	-	-
		-	-		
carbonized CF	$V^{2+}/V^{3+}$	0.1	-	-	-
		-	-		
composite (T)	$V^{2+}/V^{3+}$	3.7	-481.1	108.5	0.58
		-6.4	-589.6		
composite (MSA)	$V^{2+}/V^{3+}$	3.8	-475.5	126.8	0.53
		-7.2	-602.3		

## S4. Nyquist plots



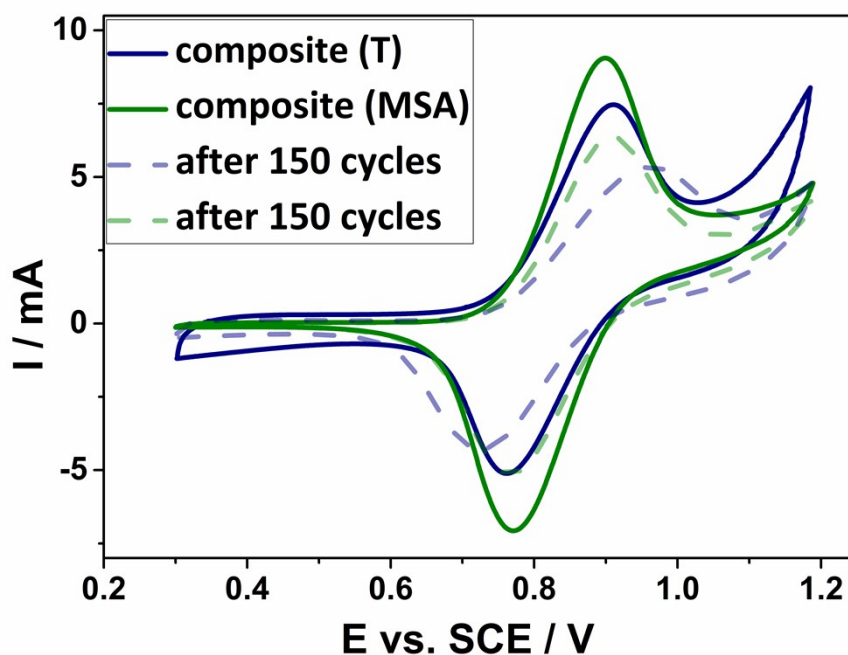
**Figure S2:** Nyquist plots showing the EIS data for the carbonized CF (left, red) as well as for the composite electrode catalyzed by acid (right, green) and for the electrode material received by thermally induced polymerization (middle, blue) in the positive electrolyte solution. The data were fitted to the circuit model displayed in the graph, where “ $R_u$ ” describes all ohmic resistances that arise from the experimental setup. All simulated curves are shown as dashed lines. It is worth mentioning here, that the order of magnitude of the x-axis of the carbonized CF is different.

## S4. TGA measurement



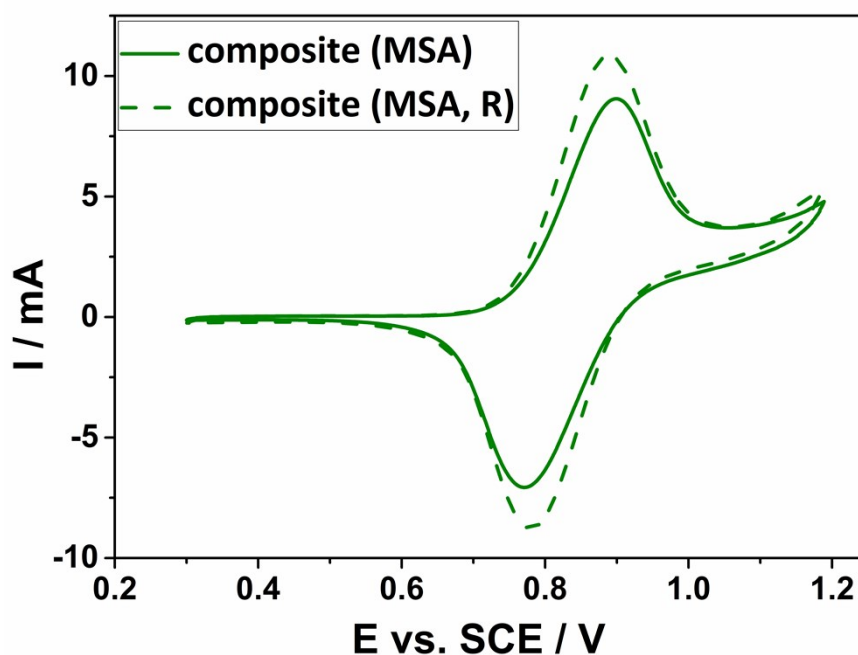
**Figure S3:** TGA measurement of the carbon material etched one time (purple) and carbon material etched for two times (pink).

### S5. Cyclic voltammetry - Accelerated stress test



**Figure S4:** Comparison between the cyclic voltammetry curves of the composite electrode obtained by thermal catalysis (blue) and composite, where the polymerization was catalyzed by acid (green) for the positive half-cell reaction compared to the same electrode materials measured again after 150 cycles (dashed lines).

### S6. Cyclic voltammetry



**Figure S5:** Comparison between the cyclic voltammetry curves of the composite electrode obtained by acidic catalysis (green) and the composite material synthesized for a second time (dashed lines, abbreviation 'R' is used for 'repeated synthesis').