Rapid wet-chemical oxidative activation of graphite felt electrodes for vanadium redox flow batteries

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Figure S1: a) SEM imaging of a MnO$_2$ decorated P-GF after step 1 in Figure 1 and b) SEM-EDX mapping of multiple fibers (Top left: Raw SEM image, Top middle: Oxygen, Top left: Phosphorus, Bottom left: Carbon, Bottom middle: Sulfur, Bottom right: Manganese).
Figure S2: XRF analysis of the P-GF, MnO\textsubscript{x} coated GF (denoted as Mn-GF) and the K-GF electrodes. Special attention must be paid to the y-axis scale bar.

Figure S3: Photographs depicting the physical condition of electrodes used in this study. a) P-GF before treatment, b) T-GF electrode after treatment, c) K-GF electrode after treatment, d) P-GF electrode bent with tweezers, e) T-GF electrode bent by tweezers and f) K-GF electrode bent by tweezers.
Table S1: Cyclic voltammetry results for the optimization of the KMnO4 treatment process as described in Table 1 and Figure 2. The best result for each testing group is marked with an *.

| Test Number | Test Type     | $E_{\text{peak ox}}$ V | $E_{\text{peak red}}$ V | $\Delta E_{\text{peak}}$ V | $I_{\text{peak ox}}$ A/g | $I_{\text{peak red}}$ A/g |
|-------------|---------------|-------------------------|--------------------------|-----------------------------|---------------------------|---------------------------|
| 1*          | KMnO4 Conc.   | 0.880                   | 0.724                    | 0.156                       | 2.69                      | -3.14                     |
| 2           |               | 0.893                   | 0.687                    | 0.206                       | 2.07                      | -2.80                     |
| 3           |               | 0.903                   | 0.677                    | 0.226                       | 1.84                      | -2.57                     |
| 4           |               | 0.908                   | 0.685                    | 0.223                       | 2.58                      | -3.01                     |
| 5*          | Time          | 0.873                   | 0.722                    | 0.151                       | 2.99                      | -3.03                     |
| 6           |               | 0.916                   | 0.682                    | 0.234                       | 2.44                      | -3.01                     |
| 7           |               | 0.926                   | 0.685                    | 0.241                       | 1.86                      | -2.85                     |
| 8           |               | 0.920                   | 0.667                    | 0.253                       | 1.78                      | -2.44                     |
| 9           | Temperature   | 1.010                   | 0.634                    | 0.376                       | 1.41                      | -1.54                     |
| 10          |               | 0.889                   | 0.708                    | 0.181                       | 2.11                      | -3.07                     |
| 11*         |               | 0.882                   | 0.717                    | 0.165                       | 2.75                      | -3.11                     |
| 12          |               | 0.907                   | 0.698                    | 0.209                       | 2.22                      | -2.95                     |
| 13*         | Acid          | 0.873                   | 0.714                    | 0.159                       | 2.51                      | -3.07                     |
| 14          |               | 0.892                   | 0.710                    | 0.182                       | 2.46                      | -2.95                     |
| 15          |               | 0.883                   | 0.714                    | 0.169                       | 2.40                      | -2.94                     |

Figure S4: Contact angle of DI water on the a) P-GF electrode, b) T-GF electrode and c) K-GF electrode. Sample sizes of 1 cm² were used and each water droplet volume is 5 µL.
Figure S5: XPS spectra of the P-GF electrode survey (a) and semicore states (b) and K-GF electrode survey (c) and semicore states (d).
Figure S6: Peak fitting of C1s (a, b) and O1s (c, d) of the P-GF and K-GF samples. Special attention must be given to the y-axis scale on (c) and (d).
Figure S7: EDLC measurement of the P-GF, T-GF and P-GF electrodes. CV was performed in a potential range of 0.4 to 0.9 V vs. SCE with different scan rates (200, 150, 100, 50, 20, 10, and 5 mVs⁻¹) in 2 M H₂SO₄. The peak currents of a) P-GF, c) T-GF and e) K-GF at 0.6 V were plotted against scan rates and fitted linearly in b), d) and f) respectively.
Figure S8: EIS curves of P-GF, T-GF and K-GF electrodes in the frequency range of $10^1$ to $10^4$ Hz. Inset: Focused view of the T-GF and K-GF electrodes. Real and imaginary ohmic values were multiplied by the active geometric cell electrode area (4 cm$^2$). Equivalent circuit fits are displayed as dashed green lines.

![Figure S8: EIS curves of P-GF, T-GF and K-GF electrodes in the frequency range of $10^1$ to $10^4$ Hz.](image)

Figure S9: Equivalent circuit used to fit T-GF and K-GF EIS data of Figure S8. $R_\Omega$ corresponds to the ohmic resistance of the cell and is equal to the HFR. $R_{ct}$ is the charge transfer resistance and $R_{Diff}$ is the finite diffusion resistance. CPE$_{dlc}$ and CPE$_{Diff}$ correspond to the double layer capacitance of the electrode/electrolyte interface and the diffusion capacitance of the vanadium ions into the electrode structure, respectively.

![Figure S9: Equivalent circuit used to fit T-GF and K-GF EIS data of Figure S8.](image)

Table S2: Obtained values from fitting Nyquist plots displayed in Figure S7 using the equivalent circuit shown in Figure S8.

| Electrode | $R_\Omega$ (Ω cm$^2$) | $R_{ct}$ (Ω cm$^2$) |
|-----------|-----------------|-----------------|
| P-GF      | 0.65            | 90.50           |
| T-GF      | 0.56            | 0.88            |
| K-GF      | 0.51            | 0.53            |