Supporting Information

Solid-state transformation of aqueous to organic electrolyte - Enhancing the operating voltage window of 'in situ electrolyte' supercapacitors

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Tab. S1: Elemental composition of Carb_{KTFSI} via elemental analysis.

| | C / wt.% | H / wt.% | N / wt.% | S / wt.% |
|-----------------------|----------|----------|----------|----------|
| Carb _{kTFSI} | 81.62 | 0.22 | 3.21 | n.d. |



Fig. S1 Operative voltage of the of the electrodes. At 1.2 V vs. Ag strong anodic dissolution occurs, while the negative electrode is very stable.



Fig. S2 Reference electrode with washed carbon (RefSC $_{\rm Wet}$) and 0.5 M KTFSI in ACN as electrolyte.



Fig. S3 EDX Spectrum of the cycled $C_{\mbox{\tiny KTFSI}}\mbox{-}Electrode.$



Fig. S4 Elemental distribution of the cycled C_{KTFSI} -electrode

Tab. S2: Elemental distribution of the cycled CKTFSI-electrode

| Element | Element | Element | Atomic | Error |
|---------|---------|-----------|---------------|-------|
| Number | Symbol | Name | Concentration | |
| 6 | С | Carbon | 63.9 | 0.8 |
| 9 | F | Fluorine | 15.3 | 0.1 |
| 8 | 0 | Oxygen | 18.0 | 0.4 |
| 19 | К | Potassium | 1.1 | 0.3 |
| 13 | AI | Aluminium | 1.1 | 0.0 |
| 16 | S | Sulfur | 0.7 | 0.2 |



 $\label{eq:Fig.S5} \textbf{Fig.S5} \ \textbf{Cyclic voltammogram of } \textbf{ISC}_{wet} \ \textbf{at different scan rates}.$

Tab. S3: Specific Energy and Power for $\mathsf{ISC}_{\mathsf{Wet}}.$

| Rate | Specific Capacitance | Specific Energy | Specific Power |
|---------------------|-------------------------|------------------------|-------------------------|
| 1 A g ⁻¹ | 38 F g ⁻¹ | 23 Wh kg ⁻¹ | 1000 W kg ⁻¹ |



Fig. S6 Nyquist plot of the dry-processed electrode with conductive additive and without.



Fig. S7 GCPL curves of ISC_{Dry} at specific currents.

Water content in the electrodes

In order to assess the soluble water content of the electrode, Karl-Fischer titration was applied to pure pre-dried ACN. The water content within the ACN was below 1 ppm water. Electrodes were then soaked into 150μ L of this reference ACN. The water content in the ACN was still below 1 ppm of water. There may be still traces of water in the electrodes, however if there are traces, these traces are not be soluble in the electrolyte and could potentially stay at the carbon surface.

BET-Surface Area of the electrode

The washed electrode including the aluminium current collector exhibits a surface area of 580 m²/g. The non-washed electrode shows a surface area below $1 \text{ m}^2/\text{g}$.

TGA analysis of water and KTFSI in the electrodes



Fig. S8 TGA/DTA of pure KTFSI. The salt melts at 200°C and decomposes at around 400°C.



Fig. S9 TGA/DTA of the composite electrodes containing KTFSI. The salt KTFSI melts at 200°C and decomposes at around 400°C. At 300°C there is a very small mass loss. However, the DTA signal shows an exothermic peak here, indicating decomposition of the carbon surface.

Additional electrode morphology measurements:

KTFSI containing sample



Fig. S10 SEM of the wet-processed electrodes containing KTFSI.



Fig. S11 SEM of the wet-processed electrodes containing KTFSI.



Fig. S12 Microscope image of a crystal growing out of a wet-processed electrode and building a crystalline surface on top of the electrode



Fig. S13 Microscope image of a crystal growing out of a wet-processed electrode and building a crystalline surface on top of the electrode, showing carbon particles underneath the outer crystal layer



Fig. S14 High Contrast, coloured microscope image of a crystal growing out of the wet-processed electrode and building a crystalline surface on top of the electrode, showing carbon particles underneath the outer crystal layer

Washed carbon sample:



Fig. S15 SEM image of the washed wet-processed carbon electrode showing a grove, where a crystal grew before.



Fig. S16 SEM image of the washed wet-processed carbon surface.



Fig. S17 SEM image of the washed wet-processed carbon surface with a residual KTFSI crystal.



Fig. S18 Microscope image of the washed wet-processed carbon surface.



Fig. S19 Microscopy profiling of the washed wet-processed carbon surface.



 $\label{eq:Fig.S20} \mbox{ S20 SD Visualization of the washed wet-processed carbon surface}.$