A novel insight into neutral medium as electrolyte for high-voltage supercapacitors

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SUPPORTING INFORMATION

1. Cyclability of capacitor operating in Li₂SO₄, Na₂SO₄ and K₂SO₄ (1 mol L⁻¹) solutions

To compare the stability of capacitors operating in different alkali metal sulphate solutions as electrolytes, cyclability tests were performed. Capacitor was charged/discharged during 5 000 cycles at the current density 1 A g⁻¹. The capacitance vs. cycle number dependence for these capacitors is placed below.

![Graph showing capacitance vs. cycle number for Li₂SO₄, Na₂SO₄, and K₂SO₄ solutions.]

Fig. 1 Cyclability of capacitor operating in Li₂SO₄, Na₂SO₄ and K₂SO₄ (1 mol L⁻¹) solutions

Capacitor operating in lithium sulphate solution as electrolyte was characterized by stable capacitance values (ca. 120 F g⁻¹) during 5 000 cycles, whereas for sodium sulphate solutions it was about 75 F g⁻¹ and for K₂SO₄ the capacitance was equal to ca. 50 F g⁻¹.
2. Solubility of Li$_2$SO$_4$ in water

Lithium sulphate reveals reversed solubility in water with temperature increase. However, it might aggravate capacitor behavior at higher temperatures, because the local concentration in pores can be higher than in electrolyte bulk and Li$_2$SO$_4$ can precipitate resulting in resistance increase. As it can be seen from the Fig. 2 the solution of 1 mol L$^{-1}$ is in the safe non-crystallization region, below meta-stable phase. Hence, the best electrochemical behavior can be observed for 1 mol L$^{-1}$ Li$_2$SO$_4$ solution as electrolyte.

![Fig. 2. Solubility of Li$_2$SO$_4$ in water vs temperature dependence](image.png)
3. Electrochemical impedance spectroscopy at different voltages

Electrochemical impedance spectroscopy was also performed for capacitor operating at 0.8, 1.6, 2.2 V and reveal small increase of resistance, which might suggest a slight oxidation of the electrodes. However, after cycling at 2.2 V no significant increase of resistance was observed.

Fig. 3. Nyquist plots recorded at different capacitor voltage for 1 mol L\(^{-1}\) Li\(_2\)SO\(_4\) as electrolyte
4. Electrochemical impedance spectroscopy before and after cycling

Impedance spectra recorded before and after cycling (5 000 cycles) of capacitor operating at 2.2 V with 1 mol L\(^{-1}\) Li\(_2\)SO\(_4\) aqueous solution are placed below.

Fig. 4. Nyquist plots recorded before and after cycling for 1 mol L\(^{-1}\) Li\(_2\)SO\(_4\) as electrolyte

As it can be observed, no significant increase of the resistance was observed which proves that there is no electrolyte decomposition during cycling.