Electronic Supplementary Material (ESI) for Materials Advances.

SUPPORTING INFORMATION

Effect of conductivity, viscosity, and density of water-in-salt electrolytes on the electrochemical behavior of supercapacitors: molecular dynamics simulations and *in-situ* characterization studies

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Table S1- The utilized atomic partial charges and Lennard-Jones parameters of the ClO_4^- and Na^+ models. [1]

	Cl	**O**	**Na^+**
σ/nm	0.347094	0.295992	0.333045
ε/kJ mol^{-1}	1.108760	0.878640	0.011598
qe	+1.0786	-0.51965	+1.0

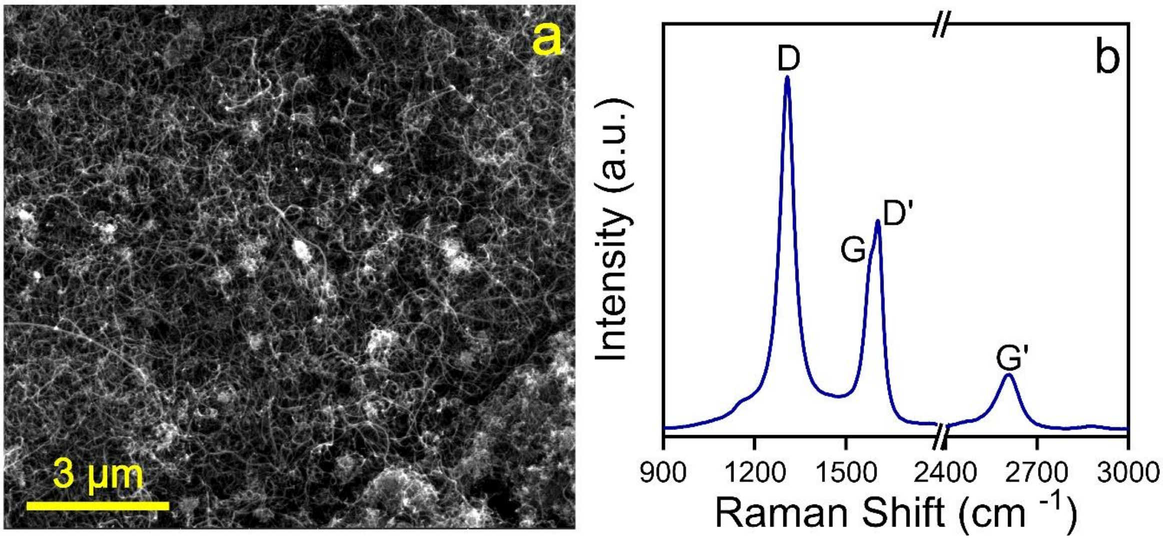

Figure S1: Electrode characterization. a) SEM and b) Raman spectrum of MWCNT.

Figure S1-a shows a micrograph of the electrodes and evidencing an arrangement of interlaced MWCNTs that form mesoporous structures. The high randomness of MWCNT guarantees that both the electrodes' electrical and mechanical properties are isotropically distributed. On the other hand, Figure S1-b shows the spectrum obtained from the electrodes, which was deconvolved to obtain the positions of the signals that compose it, obtaining bands in the regions of 1306.7 cm^{-1} (band D), 1578.3 cm^{-1} (G band), 1607.4 cm^{-1} (D' band), and 2606.4 cm^{-1} (G' band), which are characteristics of the MWCNT[2-3]. The most intense band present in this spectrum (1306.7 cm^{-1}) corresponding to the so-called D band together with the D' band (1607.4 cm^{-1}), are representative of the vibrational modes of graphitic materials with structural defects and involve, the first an optical transverse phonon activated by a double resonance inter-valley process, and the second an optical longitudinal phonon activated by an intra-valley process, according to double resonance theory. Both are due to a breathing mode of symmetry A_{1g} in which the carbon atoms move in phase radially to the center of the ring. The G' band (2606.4 cm^{-1}), also known as 2D, is an overtone of the D band and is characterized by being a process that involves two phonons. Finally, the G band, characteristic of carbonaceous materials, represents the vibrational stretching mode with E_{2g} symmetry from which an optical phonon arises due to the movement in opposite directions of the carbon atoms in the graphite ring.[1–2].

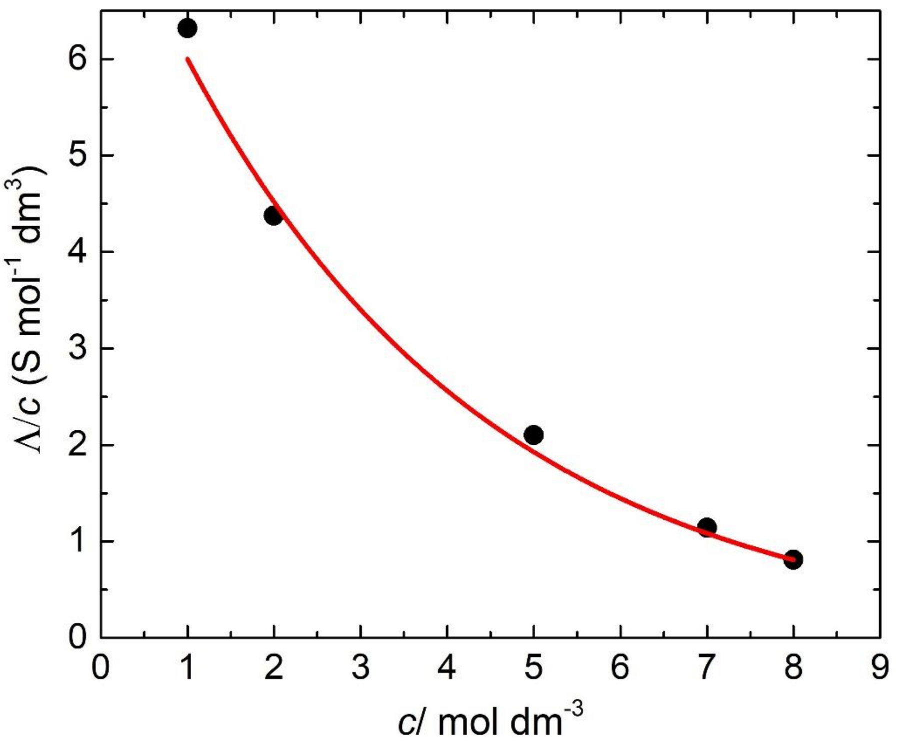

Figure S2: Molar conductivity of $NaClO_4$ aqueous electrolytes.

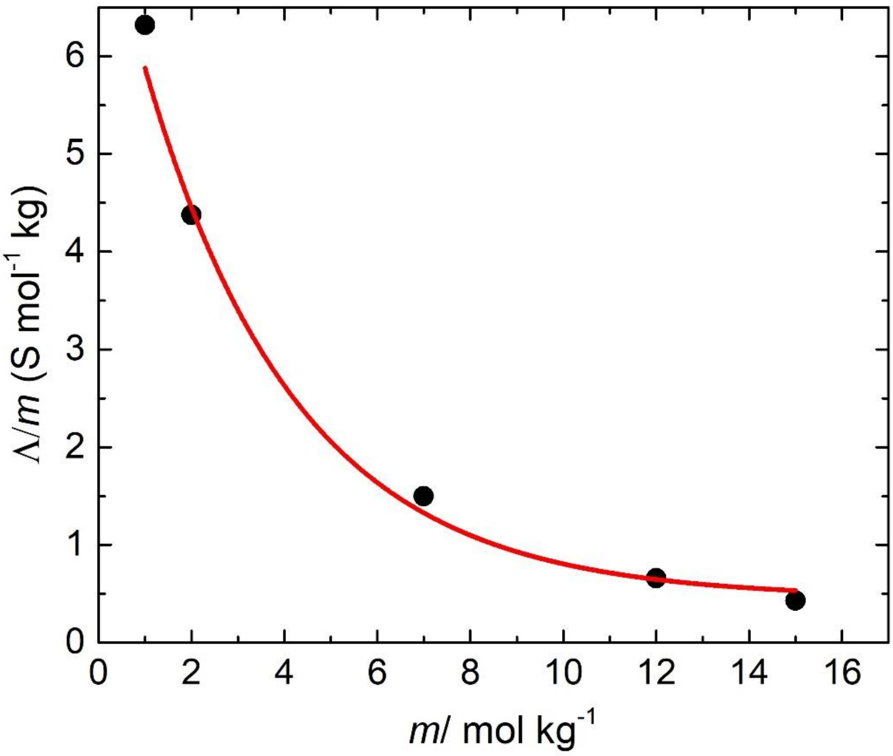

Figure S3: Molal conductivity of $NaClO_4$ aqueous electrolytes.

References

[1] K. Nieszporek, P. Podkościelny, J. Nieszporek, Phys. Chem. Chem. Phys. 18 (2016) 5957–5963.

[2] L. Bokobza, J.-L. Bruneel, M. Couzi, C 1 (2015) 77–94.

[3] Puech, Kandara, Paredes, Moulin, Weiss-Hortala, Kundu, Ratel-Ramond, Plewa, Pellenq, Monthioux, C — J. Carbon Res. 5 (2019) 69.