# SUPPORTING INFORMATION

## Contribution of Cations and Anions of Aqueous Electrolytes to the Charge Stored at the Electric Electrolyte/Electrode Interface of Carbon-Based Supercapacitors

by

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**Table S1. Potassium content deduced from EDS for several parts of a carbon fiber. The numbers 1 and 4 stand for the outer part and inner part of the fiber, respectively.**

| Part of the carbon fiber | K atomic content (%) |
|---|---|
| 1 | 1.7±0.2 |
| 2 | 1.7±0.2 |
| 3 | 1.7±0.2 |
| 4 | 1.7±0.2 |

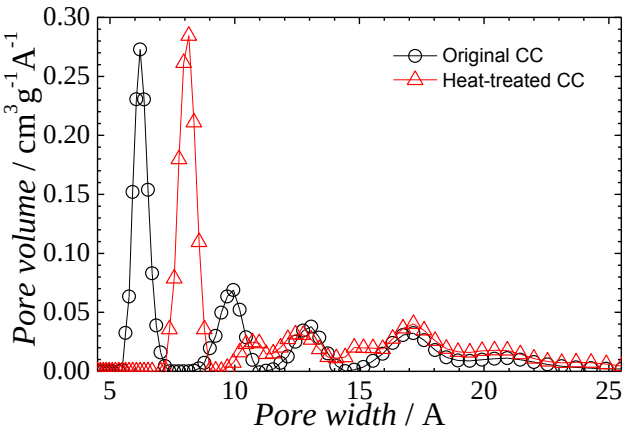


**Figure S1**. DFT pore size distribution for the original and heat-treated carbon cloth.

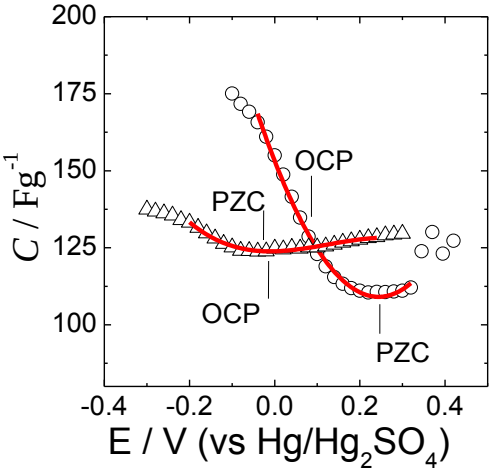


**Figure S2**. Dependence of the specific capacitance ($C$) as a function of potential ($E$). Fittings (red lines) of the experimental $C(E)$ data to the polynomic equation: $C = a+bE+cE^2+dE^3$, where a, b, c and d are the fitting parameters.
The electrolyte used was $H_2SO_4$. Circles and triangles stand for the original and heat-treated CC, respectively. The *PZC* was determined by solving the equation: $dC/dE = b + 2cE + 3dE^2 = 0$.
For comparison, the fitting parameters a, b, c and d, and the values of *PZC* and *OCP* are shown in Table S2.

**Table S2. Fitting parameters for determining the *PZC* values. The *OCP* values measured for the sulfuric acid electrolyte in presence of the two carbon cloths are also shown.**

| Electrode | a | b | c | d | *PZC* (V) | *OCP* (V) |
|---|---|---|---|---|---|---|
| Original CC | 141.6 ±0.8 | -386 ±20 | 1095 ±162 | -424 ±34 | 0.230 | 0.078 |
| Heat-treated CC | 123.9 ±0.2 | 3.8 ±1 | 167 ±7 | -445 ±52 | -0.027 | -0.010 |

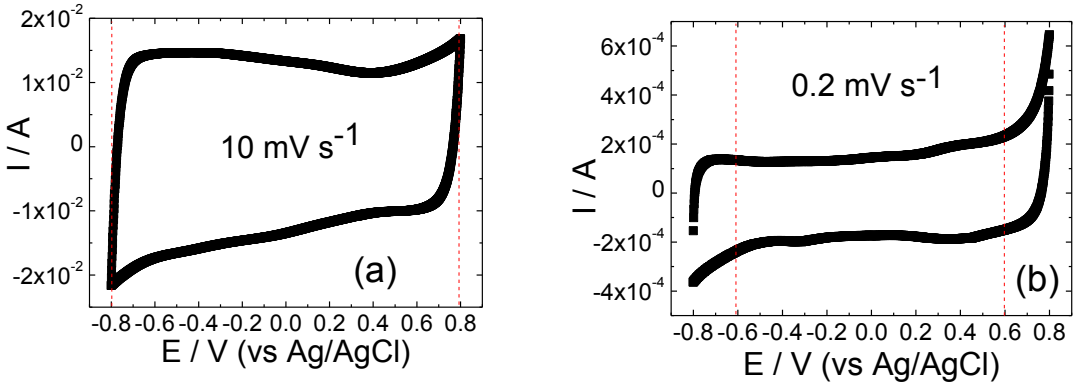


**Figure S3**. Ciclyc voltammetries recorded for the $Na_2SO_4$ electrolyte in presence of the original CC at two voltage scan rates: 10 mV $s^{-1}$ (a) and 0.2 mV $s^{-1}$ (b). While the total potential window is 1.6 V in the former case, that is 1.2 V in the latter.

**Table S3. Estimation of the pseudo capacitance, $C_+$(PS), and double layer capacitance, $C_+$(DL), for the cations $H_3O^+$ and $K^+$ from the $H_2SO_4$ and KOH electrolyte, respectively.**

| Electrolyte | Original CC | | | Heat-treated CC | | |
|---|---|---|---|---|---|---|
| | $C_+$ ($Fg^{-1}$) | $C_+$(PS) ($Fg^{-1}$) | $C_+$(DL) ($Fg^{-1}$) | $C_+$ ($Fg^{-1}$) | $C_+$(PS) ($Fg^{-1}$) | $C_+$(DL) ($Fg^{-1}$) |
| $H_2SO_4$ | 192 | 136 | 56 | 154 | 47 | 107 |
| KOH | 149 | 114 | 35 | 132 | 39 | 93 |

The pseudo capacitance was estimated according to the equations:
$C_{H3O+}$(PS)=0.042 F µmol$CO^{-1}$ · CO*content* (µmolCO $g^{-1}$) and
$C_{K+}$(PS)=0.035 F µmol$CO^{-1}$ · CO*content* (µmolCO $g^{-1}$)
where CO*content* is the content of oxygen groups evolved as CO in TPD measurements. The parameters 0.042 and 0.035 µmol$CO^{-1}$ were reported for the electrolytes $H_2SO_4$ and KOH, respectively, in reference 76.
The double layer capacitance was estimated for each cation according to the equation:
$C_+ = C_+(DL) + C_+(PS)$

where $C_+$ is the experimental capacitance measured, $C_+$(PS) is the pseudo capacitance estimated, and $C_+$(DL) is the double layer capacitance.

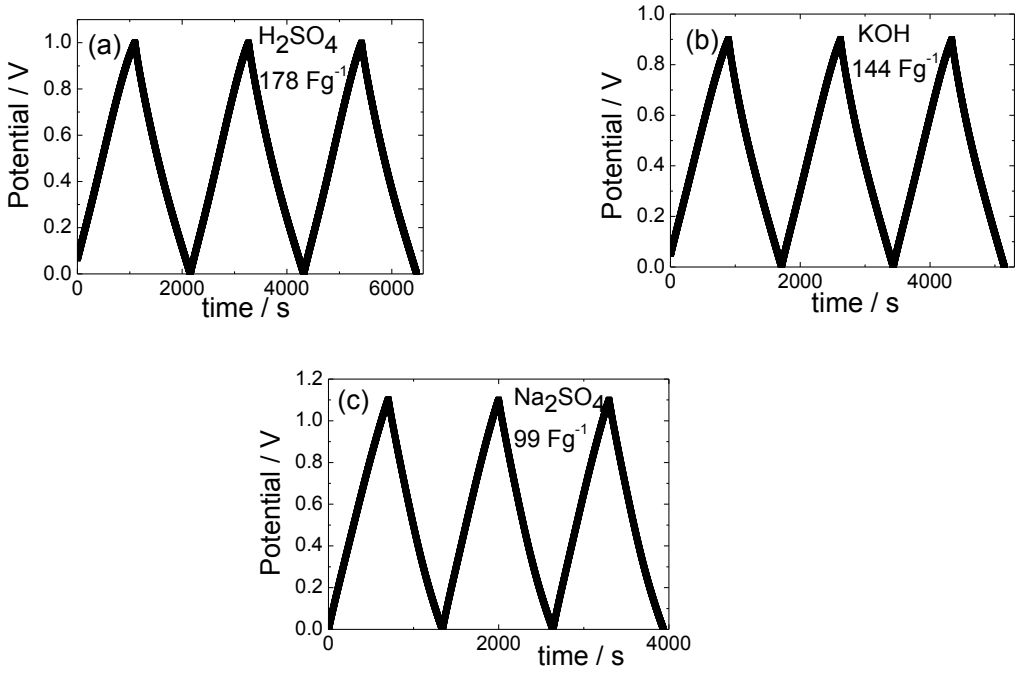


**Figure S4**. Charge/discharge galvanostatic plots recorded at 1 mA $cm^{-2}$ on symmetric two-electrode cells having the original CC as electrodes and three electrolytes: $H_2SO_4$ (a), KOH (b) and $Na_2SO_4$ (c). The specific capacitance referred to one electrode was determined according to $C_{2E}=2\cdot I\cdot t_d/\Delta V\cdot m$; where $I$ is the current applied, $t_d$ is the discharge time, $\Delta V$ is the voltage range excluding the voltage drop due to the internal resistance (*ESR*) and $m$ is the mass of one electrode only.