Supplementary information

Sustainable Conductive Ink for Printed Greener Supercapacitors

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Ink drying conditions

HCC ink was dried according to the data provided by the manufacturer of the ink: 90ºC, 30 minutes.

Ink drying conditions of the developed were studied by applying by Doctor Blade a 30 µm thick layer on a PET substrate, drying at 60ºC and measuring the mass loss at different periods of time. Data is collected on Figure S1.

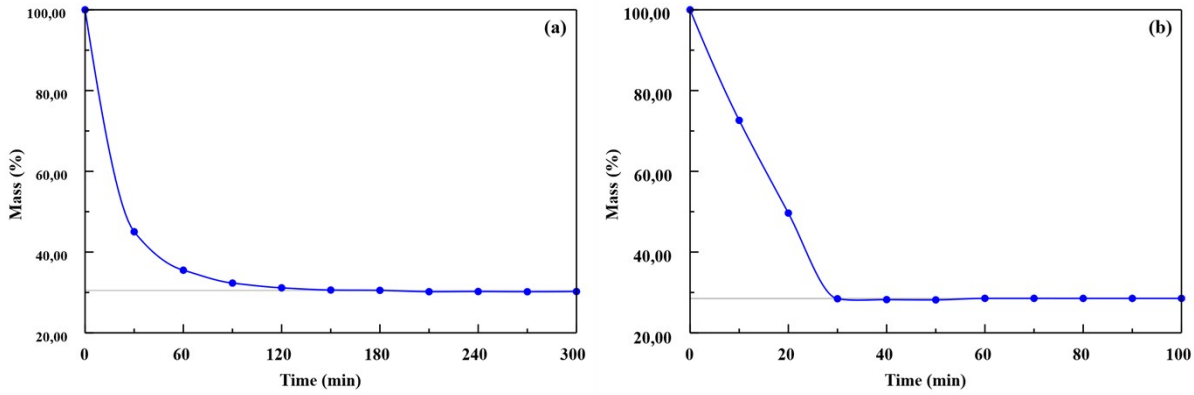

Figure S1. Mass loss at 60ºC of (a) CNC ink and (b) PLA ink

Analysing the data of Figure S1, the drying conditions were determined: 30 minutes at 60ºC for CNC inks and 150 minutes (2 hours and 30 minutes) for PLA inks, to ensure the proper evaporation of the solvents.

Optical microscope images of printed layers

After printing inks on PET substrate, the CNC_3 ink got cracked. Cracks are shown on Figure S2, with optical microscope images. PLA_3 ink and HCC ink images were also taken.

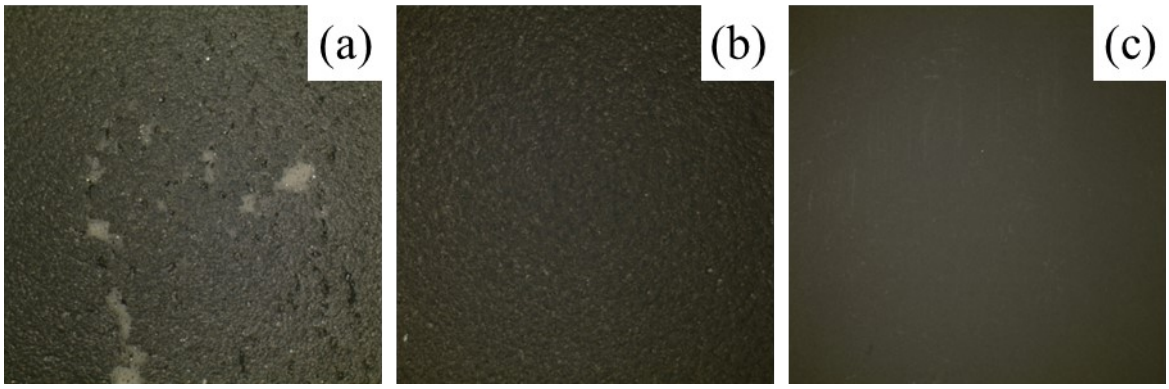

Figure S2. Optical microscope images of (a) CNC_3 ink, (b) PLA_3 ink and (c) HCC ink printed on PET substrate

Coulombic efficiency versus cycle number profiles

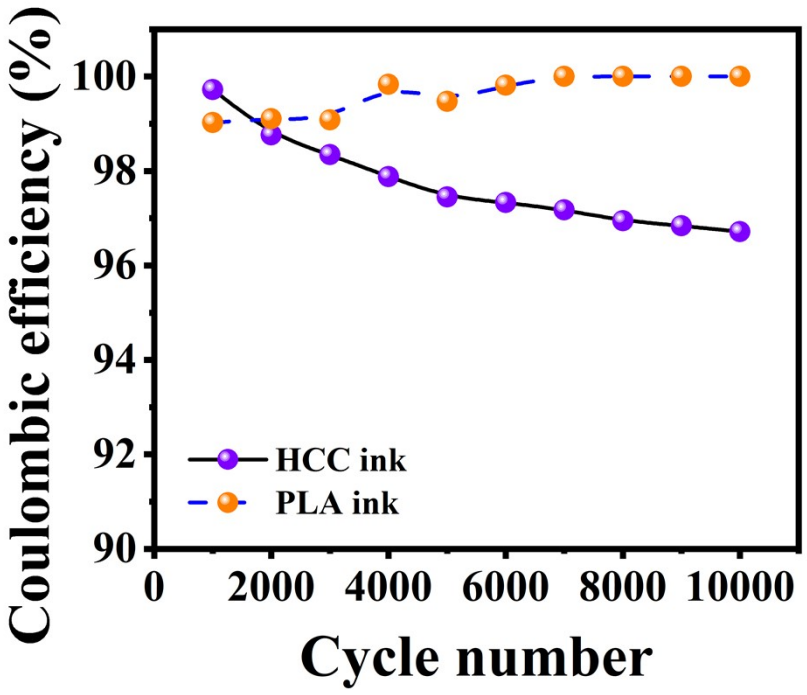

Figure S3. Coulombic efficiency versus cycle number profiles for HCC ink and PLA based SCs

Specific energy and power comparison

The specific energy and specific power were calculated using the following equations[1]. The achieved specific energy and power values demonstrate improved performance compared to previously reported printed activated carbon-based symmetric SCs, as summarized in Table S1.

$$S_E\,(W\,h\,kg^{-1}) = \frac{C_{device} \times \Delta V^2}{2 \times 3600} \quad (1)$$

$$S_P\,(W\,kg^{-1}) = \frac{S_E}{\Delta t} \quad (2)$$

Table S1. Comparative electrochemical performance of printed symmetric SCs employing activated carbon-based electrode materials.

Electrolyte	**Voltage window (V)**	**Specific energy** (W h kg^{1})	**Specific power (kW kg^{-1})**	**Capacitance retention rate**	**Ref.**
DES	1.8 V	22	0.4	90% after 10,000	This work
NaCl	1.2 V	5.4	0.056	85% after 10,000	2
DES	1.8 V	7.2	1.6	84% after 10,000	3
DES	1.8 V	7.2	1.5	67% after 10,000	1
DES	1.8 V	14.8	6.7	85% after 10,000	4
NaCl	1.2 V	7.5	1.5	80% post 10,000	5
NaCl	1.2 V	4	0.058	90% post 10,000	6
CNC/ glycerol/ NaCl	1.2 V	0.88	0.830	99% after 2000	7
DES	2.2 V	7	1	90% after 25,000	8
NaCl	1.2 V	1.92	0.536	95% after 10,000	9
H_2SO_4	1.0 V	7.59	0.5	-	10
NaCl	1.2 V	6.05	-	90.4% after 10,000	11,12
$K_xH_yPO_4$	1.2 V	6.55	-	90% after 10,000	

DES: Deep eutectic solvent, NaCl: Sodium chloride, CNC: cellulose-nanocrystals, H_2SO_4: Sulfuric acid and $K_xH_yPO_4$: Potassium Hydrogen Phosphate

LED lighting

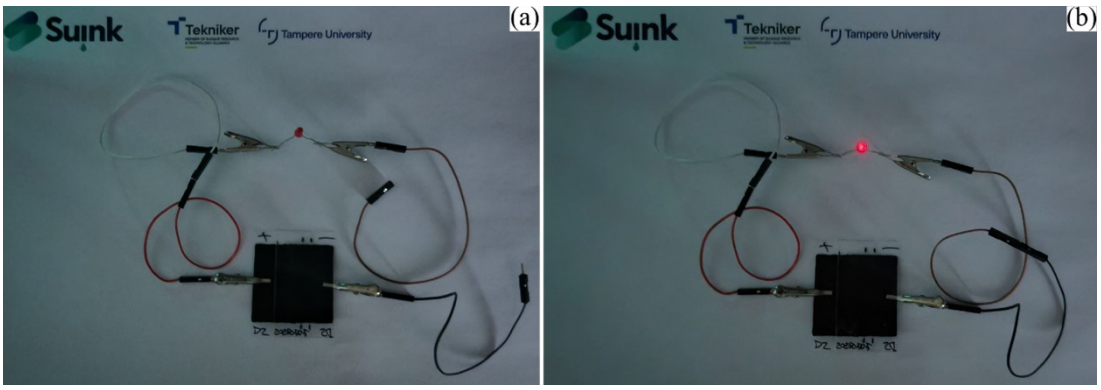

Figure S4. SC demonstration using red light emitting diode, open circuit (a) and closed circuit (b).

Material recovery

PLA ink recovery trials were made, by introducing a 30 mm x 30 mm printed on PET sample in 100 mL of Cyrene. The mix was stirred magnetically for 1 hour at 110ºC. Afterwards, the sample was taken out of the Cyrene, showing the detachment of the ink from PET substrate, as shows Figure S5.

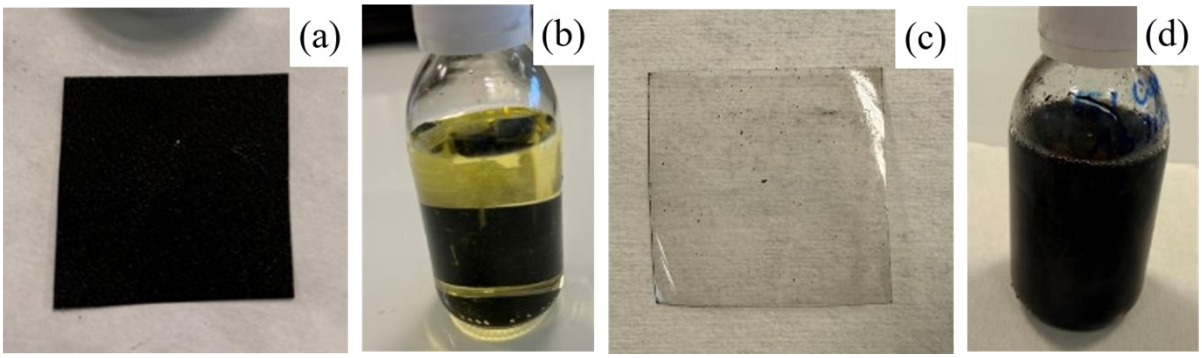

Figure S5. Ink recovery: (a) PLA_3 sample; (b) PLA_3 sample in Cyrene; (c) PET after 1 hour of stirring at 110 °C and (d) Cyrene after 1 hour of stirring at 110 °C.

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