Boosting the Capacity of All-Organic Paper Supercapacitors Using Wood Derivatives

Supplementary Information

Four-wire resistance:

The conductivity of the paper samples was measured using the four-wire resistance method. Four gold contacts were evaporated onto glass slides. Strips of the paper samples were then glued to the metal contacts using a carbon ink to ensure good electrical contact. Figure SI1 shows a picture of one of the samples used for resistance measurements.



Figure SI1: Picture showing one of the samples used for conductivity measurements.

Solvent content:

The thickness of the nanopaper films increases with the amount of LS in the samples (Figure 2b). This can be explained by the combination of adding more LS and the accompanied increase in the amount of solvents (DMSO, glycerol and water) which stay in the samples. The solvent content of the samples was estimated by weighing the samples after drying and comparing the mass to the known amount of solid material (PEDOT:PSS, CNF and LS).



Figure SI2: Solvent content (DMSO, glycerol and water) of paper samples with different amount of LS after drying.



Figure SI3. Areal capacitance for samples of different loading of LS measured at different current densities.

Specific capacitance:

In a three-electrode electrochemical measurement, the (gravimetric) specific capacitance is the capacitance of an electrode normalized by its weight. When the electrode consists of a composite material with both electronically active and inactive materials, there are different ways of performing the normalization which will show different aspects of the system. On the other hand, when a full device (two-electrode measurement) is characterized, the full mass of the device (including metal collectors, separators, electrolyte, etc.) is usually included in the calculations. The specific capacitance calculated in a three-electrode measurement is ~4 times higher than the value obtained from a two-electrode measurement (assuming that both electrodes have equal mass and capacitance). This comes from the fact that only half of the voltage and half the mass is used in the calculations in a three-electrode measurement compared to a two-electrode measurement.

In the CNF-PEDOT:PSS-LS composite, the electronically active materials are PEDOT and LS. The material also contains the solvents DMSO and glycerol, but these materials are assumed to leak out into the electrolyte during the measurements. Table SI1 present the specific capacitance of Figure 2c calculated in different ways: normalized to the mass of PEDOT+LS, PEDOT+PSS+LS or PEDOT+PSS+LS+CNF, respectively. The mass ratios of PEDOT:PSS, CNF, glycerol and DMSO (excluding LS), is as follows: 13.2/6.1/9.5/68.2 (wt%) respectively. The mass ratio of PEDOT and PSS is 1:2.5. The mass of PEDOT:PSS in a sample (4 mm diameter) is 650 µg.

Mass ratio	Specific capacitance	Specific capacitance	Specific capacitance (F/g)
(m_{LS}/m_{PEDOT})	(F/g)	(F/g)	(PEDOT+PSS+LS+CNF)
	(PEDOT+LS)	(PEDOT+PSS+LS)	
0	109	31	21
0.5	167	63	45
1	233	103	76
2	228	124	96

Table SI1. Specific capacitance calculated using different components of the total electrode mass.

Surface roughness:

The surface roughness of samples with (P:L_1:2) and without (P:L_1:0) LS was measured using a Dektak3ST surface profilometer (Veeco). Both the top and bottom side of the samples (with respect to how they were dried) were characterized. Figure SI4 shows the profile of two samples over a distance of 1000 μ m. The sample without LS had a smaller surface roughness (~3 μ m for the top and ~0.8 μ m for the bottom) compared to the sample with LS (~8 μ m for both the top and bottom).



Figure SI4. Surface profile of the top and bottom of samples with or without lignosulfonate. (a) P:L_1:0 top surface, (b) P:L_1:0 bottom surface, (c) P:L_1:2 top surface, and (d) P:L_1:2 bottom surface.



Figure SI5. Galvanostatic charge-discharge curves of samples with different mass ratios of PEDOT and LS (PEDOT:LS).