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Scalable lignin/graphite electrodes formed by mechanochemistry

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Fig. S1. IR transmittance spectra of LS, graphite, LS/graphite (2/1, 4/1, 10/1 w/w) pellets. The spectra have been shifted to allow visualization.



Fig. S2. UV-vis absorbance of LS solution in water and LS/graphite (5/1, w/w) supernatant.



Fig. S3. Raman spectra of LS/graphite hybrid material electrodes with different primary stoichiometry, with 2.33 eV (532 nm) laser excitation energy.



Fig. S4. Mass change of LS/graphite (1/1, 2/1, 4/1, 5/1, 7/1, 10/1 w/w) mixture vs. temperature in TGA curves. The measurements were obtained at a heating rate of 10 °C min⁻¹ under Ar atmosphere.



Fig. S5 Dependence of the redox peak currents on scan rate of the LS/graphite (4/1, w/w) hybrid material electrodes in 0.1 M HClO₄.



Fig. S6. Discharge capacity and coloumbic efficiency of LS/graphite (4/1, w/w) hybrid material electrodes after reversible charge-discharge cycles at 4 Ag^{-1} in 0.1 M HClO₄.



Fig. S7. The UV-vis absorption of the leaked LS from the LS/graphite (4/1, w/w) hybrid material electrodes into the electrolyte over time under different concentrations: (a) 0.01 M HClO₄, (b) 0.1 M HClO₄ and (c) 1 M HClO₄.



Fig. S8. The mass of the leaked LS from the LS/graphite (4/1, w/w) hybrid material electrodes into the electrolyte over time under different concentrations: (a) 0.01 M HClO₄, (b) 0.1 M HClO₄ and (c) 1 M HClO₄.



Fig. S9. (a) The UV-vis absorption and the (b) mass of the leaking LS from the LS/graphite (4/1, w/w) hybrid material electrodes into the electrolyte over time under 0.1 M HClO₄.



Fig. S10. (a) Discharge capacity after reversible charge-discharge cycles at 4 Ag⁻¹ and (b) galvanostatic discharge curves at the current density of 0.2 A g⁻¹, 0.5 A g⁻¹, 1 A g⁻¹, 2 A g⁻¹, 4 A g⁻¹, 8 A g⁻¹ and 16 A g⁻¹, of LS/graphite (4/1, w/w) hybrid material electrodes in 0.1 M HClO₄.



Fig. S11. Self-discharge profiles of LS/graphite (4/1, w/w) hybrid material electrodes in three electrode system at room temperature, in 0.1 M HClO₄.

SI 12 Stoichiometry calculation based on TGA results

There is a mixture of lignosulfonate (LS) and graphite in the LS/graphite hybrid material electrodes. We assume the final residual mass of LS and graphite in the LS/graphite hybrid material electrodes are constant with the pure LS and graphite after heating. For example, the residual mass fraction of LS solid after heating is 47.2%, and the final residual mass fraction of LS in the hybrid materials is also 47.2% after heating. It is the same with graphite. The total mass loss of LS/graphite hybrid electrodes after heating is equal to the mass loss of LS plus the mass loss of graphite in the LS/graphite hybrid electrodes, respectively. Accordingly, LS/graphite (4/1, w/w) hybrid material electrodes shows a residual mass fraction of 72.8%. The residual mass fraction of LS solid and graphite are 47.2% and 98.0%, respectively. In the LS/graphite hybrid electrodes, we set the amount of LS is "x" and the amount of graphite is "y". Below we have this equation:

$$x \cdot (1 - 0.472) + y \cdot (1 - 0.980) = (x + y) \cdot (1 - 0.728)$$
(S1)

We can calculate the ratio of x and y: $\frac{x}{y} = \frac{(0.98 - 0.728)}{(0.728 - 0.472)} = 0.99.$

Then we can calculate the stoichiometry of the other LS/graphite (x/1, w/w, x = 1, 2, 5, 7, 10) hybrid material electrodes and LS/graphite (x/1, w/w, x = 1, 2, 4, 5, 7, 10) mixture.