Polymer batteries: Towards the recycling of batteries

Context and objective:

The goal of this project was to develop electrodes for energy storage devices that can be recycled. To achieve this goal, we prepared electrodes based on redox-active polymers that can be processed from solution and are designed to function as single-phase electrodes in safe electrolytes (e.g. water-based electrolytes). The redox-active polymers are functionalized with polar side chains to achieve a good solubility in the solvents chosen for the processing (e.g. N,N-Dimethylformamide (DMF) or chloroform (CHCl₃)) while being insoluble in the solvents used for the electrolyte. The polar side chains also enable rapid transport of ions to enable fast charging of the electrodes with thicknesses > 1 μm.

Findings:

Polymer electrodes are prepared by drop-casting polymer solutions from low boiling point solvents onto conductive paper substrates to achieve mass loadings of 1.0 mg/cm² and 2.0 mg /cm² for cathode and anode, respectively. We assembled the battery with the anode material (~1 mg, 23 mAh/g) as the positive electrode and P-75 (~2 mg, 40 mAh/g) as the negative electrode. Since the cathode material has approximately twice the gravimetric capacity of P(3T2), twice the mass loading for the positive electrode is required to achieve cell potentials of up to 1.2 V (Figure 2 (a)). Finally, we evaluated the electrochemical stability of the battery (Figure 2 (a)) after 500 galvanostatic cycles. Encouragingly, high electrochemical stability is observed for both individual electrode mater The Ragone plot in Figure 2 (b) benchmarks the battery’s performance with other common electrochemical energy storage devices. The fabricated device achieves an energy density of 4.5 Wh/kg at power density of 3000 W/Kg. In comparison with its counterparts in the class of aqueous-based electrochemical capacitors, our work demonstrates higher energy density than the average reported values in the literature.
Following solvent extraction recycling process, the polymer on the paper electrodes are redissolved and then redeposited on new current collectors, followed by monitoring changes in the performance of the recycled battery. Encouragingly, we retain >85% of the capacity of the individual electrodes post-recycling. Figure 2 (c) shows the CV measurements after completing the stress testing protocol of 500 charge-discharge cycles and a remarkable 76% retention of charge capacity after recycling the battery twice and after a total of 1500 charging/discharging cycles.

**Conclusion**

We developed recyclable redox-active polymer electrodes that operate in aqueous-based electrolytes and maintain high specific capacities and electrochemical stability after several recycling steps. We take advantage of the outstanding electronic and ionic charge transport properties of conjugated polymers functionalized with hydrophilic side chains to employ them as single-phase electrodes. The side chains are chosen to enable device operation in environmentally friendly electrolytes such as salty water. Additionally, these polar side chains enable their high solubility in organic solvents for ease of processing the redox-active compounds from solution, paving the way for low-cost fabrication of devices and effective extraction of active phases at EOL. By combining the single-phase approach with the high solubility of polymers in processing solvents, we show that high recovery efficiency of the redox-active compounds can be achieved in simple one-step extraction processes, avoiding additional separation and purification steps commonly required for other energy storage architectures. Thus, we present an energy storage device that can be recycled with low energy consumption and waste production.