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Mechanism of aqueous electrolyte uptake by the hydrophilic lignin gel and its application as electrolyte for supercapacitor

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Lignin is one of the main components of plant cells and the most abundant aromatic polymer in nature. A distinct type of lignin, i.e., lignosulfonate (LS), is a by-product in the paper industries. Thus, using lignin-based materials to prepare gel electrolytes is environmentally beneficial and should lower the overall cost of the process. Recently, it was reported that lignin-based gels have high hydrophilic properties and thus make better combinations with selected electrolytes[1]. However, the aqueous electrolytes face several problems, i.e., enhanced corrosion, the limited operating voltage of the system (1.23V), and the possibility of leakage. Therefore, a new subgroup of aqueous electrolytes called water-in-salt (WIS) is proposed. Owing to their high concentration, WIS electrolytes effectively mitigate the problem of thermodynamic limitation of water and have a wide voltage range (2-3V)[2]. Hence, our research is dedicated to the development of lignin-based gel electrolytes with hydrophilic characteristics and combination with WIS electrolytes.

For that purpose, two types of lignosulfonates, i.e., LSp, LSpd, and a crosslinker i.e., poly(ethylene glycol) diglycidyl ether (PEO), were used for gel preparation. The obtained solution was cast into a thin layer ~100µm. Then, the prepared gel was soaked in selected acetate salts with different concentrations. Various physicochemical and electrochemical techniques were utilized for the characterization of prepared gels.

Cryo SEM analysis revealed porous and interconnecting hydrogel networks, which are crucial for high water-in-salt solution uptake and high conductivity. LSp gel has an exceptional ability to uptake a significant amount of water (water uptake=34.8 w/w) thus, the combination with various concentrations of acetate electrolytes was much better than in the case of LSpd gel (water uptake=8.8 w/w). The conductivity of prepared lignin-based gels was in the order of 10-3 S/cm at RT, which already represents a very good value for gels utilized in electrical double layer capacitors (EDLCs). Such gels combined with acetate electrolytes were assembled in Swagelok cells and tested using several electrochemical techniques (CV, GCD, EIS). The cyclic performance of such systems did not reveal any redox activity, which proved the stability and mechanical strength of the lignin-based gel electrolyte, which served as well as a separator between the carbon electrodes. In conclusion, the lignin-based gel electrolytes and WIS electrolytes (two-in-one) enhance the safety of the device and environmental sustainability over conventional designs based on fossil-based separators soaked with aqueous electrolytes. Importantly, the gel formula minimizes the possibility of leakage, which highlights the tremendous capability of lignin-based gel electrolytes for application in energy storage devices. Additionally, the effect of salt type and concentration on the uptake of aqueous solution and EDLC performance was clarified.

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