Viable morphology control strategy enabled by EG solvothermal treatment for high capacitance PEDOT:PSS films

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Abstract
Thick and highly conductive PEDOT:PSS films with ideal morphologies, are desirable as electrodes for supercapacitors. However, building uniform micro-morphology without templates or composite strategies is a formidable challenge, primarily caused by the inherent softness of dominant PSS. Herein, we successfully realized morphology control, transitioning from a layer-by-layer architecture to a porous structure in thick PEDOT:PSS films by employing solvothermal method with ethylene glycol (EG) as the solvent. The combined effect of high pressure and temperature effectively drove EG to construct the microstructure of thick PEDOT:PSS films by detaching insulating PSS chains and enhancing PEDOT crystallinity, and simultaneously facilitated the formation of a porous network through EG molecular tailoring. The achieved porous thick PEDOT:PSS films delivered a high conductivity of 1644 S cm\textsuperscript{-1} and a champion specific capacitance of 270 F cm\textsuperscript{-3}, significantly surpassing previously reports. The flexible all-solid-state supercapacitor assembled based on the films displayed an excellent specific capacitance of 97.8 F cm\textsuperscript{-3} and an energy density of 8.7 mWh cm\textsuperscript{-3}, representing the highest values for pure PEDOT:PSS-based supercapacitors. This research provides an effective novel method for conducting polymer morphology control and promotes the applications of PEDOT:PSS in the field of energy storage.

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