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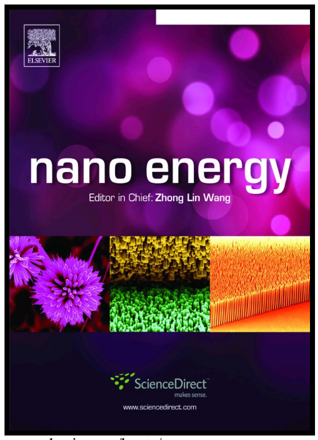

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Mechanochemical assembly of 3D mesoporous conducting-polymer aerogels for high performance hybrid electrochemical energy storage

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Abstract: Functional and structural tailoring of three-dimensional (3D) conducting polymer nanoarchitectures is a promising route but remains challenging to develop high-performance electrodes for electrochemical energy storage. Herein, we design poly(3, 4-ethylenedioxythiophene):polystyrene sulfonate (PEDOT:PSS) 3D mesoporous aerogel electrodes through a simple and original one-pot mechanochemical processing route. The hierarchical structure of neat PEDOT:PSS aerogels displays a highly interpenetrated porous conductive network with a record high active surface area of 470 m^2 g^{-1} amongst the class of conducting polymer architectures. Robust structural and electrochemical performances are achieved with high gravimetric, areal and volumetric capacitance metrics of 120 F g^{-1}, 2.5 F cm^{-2}, and 124F cm^{-3}, respectively, as the result of a mixed hybrid faradaic - capacitive charge storage mechanism. Moreover, these performances are attained in organic based electrolytes, reported so far to be incompatible or hinder the electrochemical activity of PEDOT:PSS. We undermine the fundamentals of the electrochemical operation in these electrodes and show that the electrolyte chemistry and the aerogel morphology particularly impact the charge storage performances demonstrating the superiority of the mesoporous architecture for charge transfer and ion exchange. Asymmetric cells are built having a specific capacitance of 40 F g^{-1} based on the total mass of electrodes and a good cyclic stability with 90 % capacitance retention after 1,000 charge and discharge cycles. This study offers a new route to enhance the electrochemical properties of conducting polymers and provides suggestive insights for developing high-performance polymer electrode materials for electrochemical energy storage.

Keywords: Mechanochemistry; PEDOT:PSS; aerogel; hybrid; electrochemical energy storage.