Biomimetic Strategies for 4.0 V All-Solid-State Flexible Supercapacitor: Moving toward Eco-friendly, Safe, Aesthetic, and High-Performance Devices

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To meet future demands for cutting-edge wearable electronics, flexible supercapacitors must possess many features, such as eco-friendly processing, aesthetic appeal and no health hazards, in addition to have lightweight, robust and excellent cycling stability. We proposed a biomimetic and scalable method to fabricate an all-solid-state flexible supercapacitor (assFSC) using bioinspired clay/polymer nanocomposites and electroplated manganese oxide as electrode materials and a gel electrolyte. Experimental results from X-ray techniques (tomography, small-angel x-ray scattering and diffraction) showed that the electrode's structure features a 3D ant-nest-like framework composed of 2D nacre-like clay nanosheets, i.e. hierarchical layers-*within*-networks structure, which is formed *via* water-evaporation induced self-organization. The shapeable electrodes made by a molding process could, therefore, be used to meet the demands for fashionable, wearable electronics. Accordingly, the structural electrodes exhibit high tensile strength of 62 MPa, Young's modulus of 4.4 GPa, and torsional strength of 165 MPa. Under a large operating potential of 4.0 V, the assFSC exhibited ultrahigh energy density (233.3 W h kg^{-1} at 2 kW kg^{-1}), ultrahigh power density (125 kW kg^{-1} at 55.5 W h kg^{-1}), and outstanding static cyclability (less than 10% loss after 5,000 cycles). We also performed a cycle-life test under dynamic deformation and demonstrated that the assFSC had charging and discharging abilities during motion, according to particle applications of wearable electronics. Thus stable and superior electrochemical performance can be attributed to the biomimetic layers-*within*-networks structure, which not only provided robust framework but also induced 3D conducting networks with increasing ion channels and shortening charge transports.

Keywords: Flexible supercapacitors; Biomimetics; Self-organization; Hierarchical structure; Shapeable electrode; High energy density