



INDIAN INSTITUTE OF TECHNOLOGY GUWAHATI
SHORT ABSTRACT OF THESIS

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SHORT ABSTRACT

Development of functional macrostructures from two-dimensional materials while addressing the key fundamental challenges in supercapacitive energy storage systems remains a monumental task in the field of energy storage. Meanwhile MXene, a new family of two-dimensional materials has emerged as a highly attractive material in supercapacitive applications due to their high conductivity, hydrophilicity and pseudocapacitive nature. This thesis aims towards development of functional macrostructures from MXene and related nanomaterials in the form of hydrogels while ensuring scalability and low-cost of synthesis. MXene hydrogels are an interconnected network of these two-dimensional materials which offers highly restacking controlled porous structure that enables facile accessibility of the bulk of the electrodes to the electrolyte ions. Such restacking controlled assembly enables high utilization of active mass ensuring high specific capacity and rate performance. This thesis introduces a room-temperature self-assembling strategy with the help of a small amount of graphene to induce gelation in a system of MXene and graphene for hybrid hydrogel development. It is shown that such room temperature induced assembly not only protects the intrinsic properties of MXene by preventing synthesis induced oxidation, but also enables state-of-the-art performance even in commercial scale mass loading electrodes. Furthermore, development of hydrogels of pristine MXene has been a great challenge due to the relative small sheet size and intrinsic stiffness of MXene. Here, in this thesis, for the first time a critical-density induced gelation strategy is introduced which enables the development of self-supporting pristine MXene hydrogels. It is established that liquid crystallinity induced ordering in MXene dispersion can lead to higher crosslinking of MXenes into the hydrogels which leads to better stability in such systems. The as developed hydrogels were used as supercapacitive electrodes having mass loading as high as $\sim 15 \text{ mg cm}^{-2}$, which simultaneously deliver excellent gravimetric capacity of 337 F g^{-1} and a very high areal capacitance of 5042 mF cm^{-2} . Further, in order to mitigate the high concentration requirement with critical-density controlled self-assembly, an electric-field guided forced assembly of MXene is also introduced which enables large scale assembly of MXene into hydrogels in seconds and also allows facile controllability over the MXene sheet orientation in 2D sheet like and 3D hydrogel monoliths. Such orientational controllability of MXene sheets in hydrogel enables their use in a plethora of applications which is not

otherwise easily achievable with conventional assembling strategies. The electric-field guided assembly is also demonstrated to enable co-assembly of nanomaterials with MXene. As an example, co-assembly of cellulose nanofiber (CNF) and MXene is performed to develop flexible electrodes that are used in asymmetric devices as well as wearables. The MXene-CNF hybrids were paired with a reduced graphene-carbon nanotube-polyaniline electrode to develop asymmetric supercapacitor which delivers a high energy density of 23 Wh kg⁻¹ at a power density of 501 W kg⁻¹. Furthermore, the possibility of developing wearable devices with the help of electric-field guided assembly is also demonstrated which shows excellent stability after 5000 bending cycles at extreme 90° and 180° and retains over 80% of their initial capacity after the bending tests. The high scalability, low-cost and a highly optimized electrode structure in all these works makes the MXene based electrodes, as developed in this thesis, ready for practical adoptability and commercialization.

