Department of Materials Science and Engineering

PhD Thesis Defense

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Charge Storage in Conductive Two-Dimensional Transition Metal Carbides (MXenes)

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There is increasing interest in researching materials with nanoconfined electrolytes for electrochemical energy storage applications due to the interesting affects that confinement can have on ion transport, ion desolvation and insertion, or solvent co-intercalation into host materials. Since their discovery in 2011, the family of two-dimensional (2D) transition metal carbides, nitrides, and carbonitrides known as MXenes have received significant attention for energy storage applications due to their tunable properties. The reactivity of their transition metal surfaces enables MXenes to store higher amounts of energy than all reported traditional double layer capacitor materials, and due to their unique chemical composition and layered structures MXenes can intercalate a wide variety of organic and inorganic cations. However, there are still improvements to be made in the charge storage capabilities of MXenes.

This dissertation broadly aims to understand how the interactions between MXenes and the electrolytes confined within their 2D sheets affects electrochemical energy storage in MXenes. My work has shown that ion transport in the MXene Ti_3C_2 can be significantly improved, independent of electrode thickness, by orientational control of the flakes in a MXene electrode. Furthermore, it was found that electrolyte composition plays a major role in the storage of charge in Ti_3C_2 in organic electrolytes. By simply changing electrolyte solvents. the capacity of Ti_3C_2 was doubled. Finally, a new type of electrochemical process was discovered when Ti_3C_2 was tested in highly concentrated aqueous electrolytes. What was at first deemed "battery-like" faradaic charge storage, was found to be an electrochemical mechanism more akin to pseudocapactive intercalation *via* combined *operando* and *in situ* measurements. The results presented in this thesis offer unique insight into the use of MXenes as electrode materials to achieve simultaneous high-power, high-energy electrochemical energy storage systems