Anomalous pseudocapacitive charge storage of vanadium carbide MXene in Ca-ion electrolyte

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Calcium-based rechargeable batteries hold great potential for sustainable energy storage, but their development is hindered by the irreversible electrochemical behavior of calcium metal anodes. Due to the formation of nonporous solid electrolyte interphase (SEI) that blocks the transportation of calcium ions. Alternatively, the advancement of calcium-ion batteries requires the development of robust charge storage hosts such as layered and open channel materials to enable reversible and rapid diffusion of high charge density calcium ions. The two-dimensional (2D) transition metal carbides, nitrides, and carbonitrides, commonly known as MXenes, have gained significant attention as a conductive redox charge storage host for multivalent metal ions (Zn, Mg, and Al). We report on the preliminary experimental efforts on vanadium carbide MXene (V_2CT_x) as a 2D charge host for the reversible (de)intercalation of calcium ions. Typically, hydrated metal ions form electrical double layers within the interlayer spaces of MXenes, leading to intercalation capacitance and undergo irreversible anodic oxidation beyond 0.2 V vs Ag/AgCl in aqueous electrolyte. However, employing water-in-salt electrolyte (WiSE) expands the anodic potential window of operation (0.6 V vs Ag/AgCl) by suppressing the kinetics of oxygen evolution reaction and electrode decomposition. The extended potential window unlocks an additional V^{3+}/V^{4+} redox couple and was confirmed through ex situ X-ray photoelectron spectroscopy. V_2CT_x MXene exhibited a reversible calciation capacity of 88 mAh g^{-1} (current density of 0.5 A g^{-1}) in WiSE, which is 4 times higher than that of conventional salt in water electrolyte. Furthermore, a symmetric V_2CT_x // V_2CT_x full cell operates within a voltage window of 1.4 V in WIS electrolyte, exhibiting good cycling stability over 10,000 charge-discharge cycles. This study opens up new avenues for exploring MXene electrodes in water-in-salt electrolytes and advances the development of multivalent metal-ion batteries.