Department of Materials Science and Engineering,

PhD Defense

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Polysulfide Adsorption by MXenes for Enhancing Performance of Metal-Sulfur Batteries

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The global shift towards sustainable energy has sparked significant interest in metal-sulfur batteries, which offer advantages over traditional lithium-ion batteries, such as lower cost, non-toxicity, and abundant material availability. Lithium-sulfur (Li-S) batteries show great promise for electric vehicles, while sodium-sulfur (Na-S) batteries are well-suited for grid-scale energy storage, supporting renewable energy integration. However, their commercialization is limited by challenges like sulfur loss to the electrolyte, the polysulfide shuttle effect, and low sulfur loading due to its insulating nature. MXenes, a class of two-dimensional (2D) nanomaterials, have emerged as a potential solution due to their high surface area, outstanding conductivity, and catalytic properties. When employed as passive components (cathode host, interlayers, additives) in Li-and Na-S batteries, $Ti_3C_2T_x$, Ti_3CNT_x, Ti_2CT_x, V_2CT_x, Nb_2CT_x, and Mo_2CT_x MXenes have shown significant improvements in cell-level performance by immobilizing polysulfides and accelerating sulfur redox reactions. Despite their potential, the impact of MXene composition and structure on polysulfide adsorption and sulfur conversion remains understudied, which is foundational to their application in metal-sulfur batteries. This dissertation investigates the influence of MXene composition and structure on polysulfide adsorption and sulfur conversion kinetics. It addresses a gap in current understanding crucial to their application in metal-sulfur batteries. Seven MXenes, Ti_2CT_x, V_2CT_x, Nb_2CT_x, $Ti_3C_2T_x$, Ti_3CNT_x, $Mo_2TiC_2T_x$, and $Nb_4C_3T_x$/$Ti_3C_{2-y}O_yT_x$, covering 3 M_nX_{n-1} structural groups, 4 M-elements and multiple compositional variations were considered to establish a relationship between MXene chemistry and adsorption properties through experimental (qualitative and quantitative spectroscopic methods) and theoretical (density functional theory (DFT) calculations) analyses. We demonstrated that all MXenes adsorb polysulfides and reduce high-order polysulfides to low-order polysulfides, but to different extents. Selected MXenes were tested as conductive cathode hosts and showed both fast electron/ion transport and conversion of soluble lithium polysulfide species. Besides, $Ti_3C_2T_x$, with outstanding electronic conductivity (~10,000 S/cm), proved to be an effective conductive additive, minimizing sulfur loss and enhancing capacity retention. The findings of this dissertation offer guidance for selecting MXenes tailored to the functional requirements of specific Li-S battery components and advance sustainable energy storage technologies.