

## ABSTRACT

## 2D MXenes and their composites for high-rate energy storage

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MXenes, a family of 2D transition metal carbides/nitrides, exhibit exceptional pseudocapacitor performance due to their ionophilicity, metallic conductivity, and highly reactive surfaces. While MXenes show strong proton-coupled redox activity in acidic electrolytes, their surface redox activity diminishes in neutral electrolytes due to weaker ion interactions, as revealed by in-situ UV-Vis spectroscopy.[1] To enhance MXene's performance in non-acidic electrolytes, we explored how optimizing MXene electrode structures and electrolytes influences capacitance and rate capability. For example, a novel desolvation-free intercalation of Li<sup>+</sup> ions into MXene layers was observed in water-in-salt (WIS) electrolytes, significantly enhancing the capacitance of MXene in neutral aqueous electrolytes.[2] Furthermore, partially oxidizing MXene in WIS electrolytes activates additional surface redox reactions below -0.2 V, increasing pseudocapacitance by shifting Ti oxidation states. [3]

We recently developed a superlattice-like MXene-n-type conjugated polyelectrolyte (CPE) heterostructure via self-assembly, facilitating rapid ammonium ion storage. [4] The superlattice-like structure persists as the CPE ratio increases, leading to a linear expansion of the interlayer spacing in MXene flakes and enhanced overlap of CPE units. Remarkably, the redox activity per unit of CPE intensifies as well, a phenomenon attributed to reinforced desolvation of ammonium ions due to the increased volume of 3 Å-sized pores. Additionally, we employed a PEG-based molecular crowding electrolyte (MCE) to improve MXene's electrochemical performance.[5] Introducing PEG into dilute LiTFSI electrolyte increased capacitance by 25% (100.8 F g<sup>-1</sup>), likely due to the co-insertion of PEG molecules with fully solvated Li<sup>+</sup> ions.

- [1] Zhang D. et al., Nat Energy, 8, 567 (2023).
- [2] Wang X. et al., ACS Nano, 15, 15274 (2021).
- [3] Wang X. et al., ACS Energy Lett., 7, 30 (2022).
- [4] Chen C. et al., Adv. Energy Mater., 202402715 (2024).
- [5] Chen C. et al., 2D Mater., 11(1), 015001 (2023).