

# ***In-operando* tracking ion intercalation into 2D MXenes in aqueous electrolytes**

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In the past decade, supercapacitors have drawn considerable attention for portable electronics, grid and transportation systems due to their rapid power delivery and almost unlimited cycle life. To enhance the energy stored, the intensive efforts have been devoted to exploring new electrode materials, new electrolytes, and novel cell configurations. In search for new electrode materials, 2D transition metal carbides-MXenes, are of particular interest owing to their excellent electrical conductivity and high volumetric capacitance[1][2]. Various aqueous cations can be electrochemically intercalated into  $Ti_3C_2$ , resulting in very high volumetric capacitance outperforming a variety of carbons[1]. The mechanism for high capacitance was essentially described as intercalation pseudo-capacitance arising from redox reactions of the Ti atoms. Similar to graphite or other electrode materials, MXene also show a significant change in volume during intercalation. This electro-chemo-mechanical coupling can be used to get unprecedented insight into ion intercalation pathways with lateral resolution of 10's of nm using Scanning Probe Microscopy (SPM) techniques. Herein, we introduce contact resonance (CR) SPM to extract mechanical properties and its changes under electrochemical control [3][4]. It is of great importance to explore the different intercalation contributions with varying aqueous cations to the mechanical property's variations of material itself, which is required to evaluate the electrochemical long-term stability of electrode materials. Of special interest to boost energy storage is the intercalation of multivalent ions suffering from sluggish intercalation and transport kinetics due to its ion size. By combining electrochemical dilatometry and CR SPM, the synergetic effects of smaller ion size cation and larger ion size cation are demonstrated to improve charge storage to maximize the utilization of electrode volume, as well as tune mechanical and actuation properties of  $Ti_3C_2$  MXene[4][5]. Our results have important implications for quantitatively understanding the charge storage processes in intercalation compounds and provide a new path for studying the mechanical evolution for electroactive materials.

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