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Two-Dimensional Unilamellar Cation-Deficient Metal Oxide Nanosheet Superlattices for High-Rate Sodium-Ion Energy Storage

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Abstract

Cation-deficient two-dimensional (2D) materials, especially atomically thin nanosheets are highly promising electrode materials for electrochemical energy storage that undergo metal ion-insertion reactions, yet have rarely been achieved thus far. Here, we report a Ti-deficient 2D unilamellar lepidocrocite-type titanium oxide ($Ti_{0.87}O_2$) nanosheet superlattice for sodium storage. The superlattice composed of alternately restacked defective $Ti_{0.87}O_2$ and nitrogen-doped graphene monolayers exhibits an outstanding capacity of ~490 mA h g−1 at 0.1 A g−1, an ultralong cycle life of more than 10,000 cycles with ~0.00058% capacity decay per cycle, and especially superior low-temperature performance (100 mA h g−1 at 12.8 A g−1 and −5 °C), presenting the best reported performance to date. A reversible Na^+ ion intercalation mechanism without phase and structural change is verified by first-principles calculations and kinetics analysis. These results herald a promising strategy to utilize defective 2D materials for advanced energy storage applications.