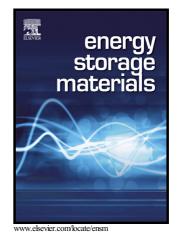
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Bilayered Vanadium Oxides by Chemical Pre-Intercalation of Alkali and Alkali-Earth Ions as Battery Electrodes

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ABSTRACT

We report the use of the chemical pre-intercalation synthesis technique to insert alkali (Li⁺, Na⁺, K⁺) and, for the first time, alkali-earth (Mg²⁺ and Ca²⁺) ions into the structure of vanadium oxide leading to the formation of the bilayered δ -M_xV₂O₅ (M = Li, Na, K, Mg, Ca) phase with expanded interlayer spacing, enabling a large number of insertion sites for and faster diffusion of charge-carrying ions. By altering the nature of the chemically preintercalated ion, interlayer spacing of the synthesized δ -M_xV₂O₅ materials was varied between 9.62 and 13.40 Å. We for the first time show that the interlayer spacing increases with the increase of the hydrated ion radius. The ion $(Na^+, K^+, Mg^{2+}, Ca^{2+})$ stabilization effect was investigated in Li-ion cells, with Li-preintercalated phase, δ -Li_xV₂O₅, serving as a reference material. Our analyses indicate that cyclability and rate performance of the δ - $M_x V_2 O_5$ improves with increasing interlayer spacing. The highest initial capacity (198 mAh g⁻¹), greatest capacity retention (81.8% after 50 cycles at 20 mA g⁻¹), and highest capacity retention at higher current rates (74.5% when current rate was changed from C/15 to 1C) were exhibited by Mg-stabilized δ -V₂O₅ with the largest interlayer spacing (13.40 Å). This research demonstrates the efficacy of a facile chemical pre-intercalation strategy to synthesize ion-stabilized layered electrode materials with improved electrochemical stability. Ion-stabilized layered materials with large interlayer spacing are attractive for applications that involve electrochemically driven movement of ions through two-dimensional diffusion channels, ranging from beyond Li-ion energy storage and electrochromics to actuation and water treatment.

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