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Sodium-pillared vanadium oxide decorated with carbon particles as electrode material for more sustainable energy storage of the future

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An increased utilization of intermittent renewable energy (wind, solar...) requires the expansion of energy storage/conversion systems to a large scale, which can be integrated with renewable source devices into electrical grid. The state-of-the-art Li-ion batteries present the good choice in terms of energy and power density. However, their massive applications are strongly limited by restricted lithium resources and safety issues. In that context, the research on different alternative battery systems, relying on Earth abundant elements such as Na, Ca, Mg, Al, etc, have been launched, where advanced materials play the central role as electrodes in their operation [1,2].

Due to adaptable interlayer distance and multivalent stage of vanadium, layered vanadium oxides are interesting electrode materials for multivalent-ion charge storage devices. Herein, the multiphase Napillared vanadium oxide was hydrothermally synthesized in the form of the composite with carbon (SVM/C) to serve as a platform for understanding mono/multivalent ion chemistry and to be examined as an electrode for different aqueous rechargeable batteries. The carbon (Vulcan XC72) presence in the precursor solution during the synthesis of targeted Na₂V₆O₁₆, was found to change the reaction path, leading to a phase mixture Na₂V₆O₁₆/NaV₆O₁₅ (SVM/C) instead of the expected single Na₂V₆O₁₆ phase. Both Scanning electron microscopy (SEM) and Transmission Electron microscopy (TEM) revealed nanowire nanosphere arrays of SVM/C (Fig1). Some of nanowires are stacked into larger parallel bundles, 2-3 μ m in diameter (Fig.1, left), while the spherical-shape particles (\approx 100 nm in diameter) are grouped into smaller or larger clusters, with the tendency to form chains (Fig.1, right). The amount of carbon in the composite, determined by thermogravimetric and differential thermal analysis (TG/DTA), was found to be \approx 8 wt.%.

The applicability of the synthesized composite as electrode material was first checked in Li-ion aqueous battery. As anode of SVM/C//LiNO₃//LiFe_{0.95}V_{0.05}PO₄/C full cell, multiphase SVM/C composite shows better performance than those for reported NaV₆O₁₅ and Na₂V₆O₁₆ anodes in the form of either Li or Na rechargeable aqueous batteries. Furthermore, the high insertion capability of the synthesized multiphase oxide was shown not only in Li but also in Mg-, Al- and Ca-containing aqueous electrolytes. The main novelty we found here is that Ca redox processes of sodium vanadate in an aqueous solution are more stable than Li and Mg redox processes. Significantly higher Ca²⁺ vs. Li⁺ capacity retention (87 vs. 45 wt%) is shown during prolonged potentiodynamic cycling and that was explained by coinserted water/hydroxyl groups. Hydroxyl groups introduced by coinsertion, as evidenced by Fourier-transform infrared spectroscopy (FTIR), may take over the charge fraction and make vanadium ions less strained, thus producing better cyclic stability. Their redox process during charging/discharging of vanadium oxide is proposed. The influence of different parameters (pH, the type of anion and electrolyte concentration) to the stability of Ca redox processes is examined. The synthesized SVM/C is combined with the carbon anode to get the aqueous multivalent hybrid cell, whose performance will be presented.

With these results, we would like to put in motion the interest for Ca interfacial reactions of vanadate structures, in the aqueous-based electrolytes, as new directions towards development of a new generation of more sustainable energy storage devices.

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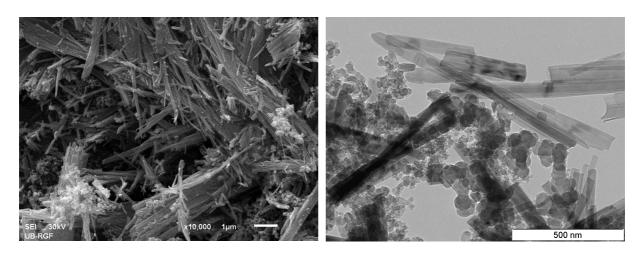


Figure 1. SEM (left) and TEM (right) micrographs of SVM/C.

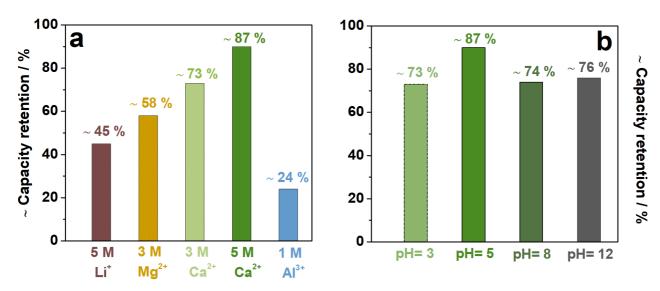


Figure 2. The capacity retention of SVM/C after 20 cycles in a) different M-containing aqueous electrolytes (M=Li, Mg, Ca and Al) and b) pH-different Ca-containing electrolytes.

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