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Phosphate-Based Mixed Polyanion Compounds as Promising Electrode Materials for Post-Lithium Ion Batteries

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The investigations on next generation of electrode materials that should meet the increasing requirements for high power, safety, low cost and environmental compatibility are a hot research topic in the field of energy storage. Iron containing phosphate-based electrode materials offer a great promise for large scale application due to high structural and thermal stability, good cycling stability and low cost.

One effective way to achieve high energy density is to increase the $M^{(n+1)+}/M^{n+}$ redox potentials though introducing more anion units with different electronegativity in the chemical composition (cooperative inductive effect). In this regard, we have focused on compounds which composition contains two kinds of anionic units: $NaFeV(PO_4)(SO_4)_2$ and $Na_4Fe_3(PO_4)_2P_2O_7$ (labeled NFVPS and NFPP, respectively). They are prepared by a precursor method using freeze-drying of solutions containing the needed components. To overcome the low electronic conductivities of NFPP and NFVPS two types of carbon-based composites have been prepared as cathode materials *via* ball-milling: with rGO(reduced graphene oxide) and carbon black (15 % each). The composites have been characterized by different methods. Their electrochemical performance have been studied in Lihalf-cells using $LiPF_6$ in EC:DMC as electrolyte in galvanostatic and potentiostatic regimes at 20 and 40 °C. It is established that rGO is more effective additive than carbon black in the achievement of higher specific capacity for the two compounds. After 100 cycles at C/2 rate very good values for the discharged capacities have been obtained: 115 mAh/g and 82 mAh/g for NFVPS/rGO at 40 and 20 °C, respectively, and 105 mAh/g for NFPP/rGO (20 °C). The presence of rGO additive promotes the capacitive behavior in addition to the intercalation reactions owing to Fe^{3+}/Fe^{2+} and V^{3+}/V^{4+} redox reactions which results in higher specific capacities.The present study demonstrates the effectiveness of carbon-based composites with tunnel-type mixed polyanion compounds for achievement of promising electrochemical performance in hybrid metal-ion batteries.

Keywords: Li/Na-ion batteries, Electrode materials, Mixed polyanion compounds