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# New Iron-Based Mixed-Polyanion Cathodes for Lithium and Sodium Rechargeable Batteries: Combined First Principles Calculations and Experimental Study

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**ABSTRACT:** New iron-based mixed-polyanion compounds $Li_xNa_{4-x}Fe_3(PO_4)_2(P_2O_7)$ ($x = 0-3$) were synthesized, and their crystal structures were determined. The new compounds contained three-dimensional (3D)-sodium/lithium paths supported by $P_2O_7$ pillars in the crystal. First principles calculations identified the complex 3D paths with their activation barriers and revealed them as fast ionic conductors. The reversible electrode operation was found in both Li and Na cells with capacities of one-electron reaction per Fe atom, 140 and 129 mAh $g^{-1}$, respectively. The redox potential of each phase was ~3.4 V (vs Li) for the Li-ion cell and ~3.2 V (vs Na) for the Na-ion cell. The properties of high power, small volume change, and high thermal stability were also recognized, presenting this new compound as a potential competitor to other iron-based electrodes such as $Li_2FeP_2O_7$, $Li_2FePO_4F$, and $LiFePO_4$.