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Solvothermal synthesis and electrochemical properties of Na_2CoSiO_4 and Na_2CoSiO_4/carbon nanotube cathode materials for sodium-ion batteries

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Abstract:

The commercialization of large-scale energy storage systems has renewed the scientific interest on electrode materials for sodium-ion batteries (SIBs). Polyanionic sodium orthosilicates [$Na_2M(SiO_4)$] are an interesting group of SIB cathode materials with high redox potentials *via* a two-electron redox process, and their three-dimensional framework. However, studies on these materials are limited due to difficulties in synthesizing them in stable and electrochemically desirable phase. Exhibiting rich polymorphism, orthosilicates undergo irreversible structural changes during processing and/or cycling. Here, we report a solvothermal process, which yields *in situ* carbon-coated Na_2CoSiO_4 with monoclinic structure. Besides physical characterization, we discuss its electrochemical performance in coin-cell configuration *vs.* Na anode. Na_2CoSiO_4 exhibits significantly lower polarization (0.15 V) than Na_2FeSiO_4 and Na_2MnSiO_4. As polyanionic compounds are poor electronic conductors with sluggish redox kinetics, we observe that these limitations could be overcome in Na_2CoSiO_4 by incorporating functionalized multi-walled carbon nanotubes (MWCNTs). The Na_2CoSiO_4/MWCNT composite cathode reversibly