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Na^+ self-diffusion in Co-substituted $Na_2Ni_{2-x}Co_xTeO_6$ Na-ion battery cathode material

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$Na_2Ni_2TeO_6$ honeycomb layered oxide has suitable properties for use as a Na-ion battery cathode material. The substitution of Ni with Co has been shown to have a detrimental effect on the energy density of $Na_2Ni_{2-x}Co_xTeO_6$, whereas the plateau potential vs Na^+/Na increases. Thus, to ascertain the cause of the electrochemical properties change upon substituting Ni with Co Na-ion self-diffusion properties are investigated with the use of zero field and longitudinal field $^+$SR methods in $Na_2Ni_{2-x}Co_xTeO_6$ with x of 0.0, 1.0 and, 1.5. Na-ion site occupancies and crystal structure was determined from neutron powder diffraction measurements and used for the determination of Na-ion jump paths. All measurements were performed in a temperature range from 50 K to 550 K. Three distinct Na-ion sites are determined from the neutron powder diffraction measurements. In addition, two distinct temperature regions for Na-ion self-diffusion, with different Na-ion diffusion pathways, are determined and analysed. The Na-ion diffusional pathway dependence on the substitution of Ni with Co is shown and discussed. Based on the obtained results we propose a cause for the decrease in the capacity, with the simultaneous increase in plateau potential vs Na^+/Na, with the increased substitution of Ni with Co. Based on the results, a roadmap on how to further improve $Na_2Ni_{2-x}Co_xTeO_6$-based Na-ion battery cathode materials is given.

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