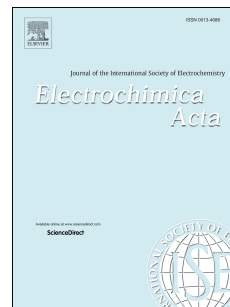


# Accepted Manuscript

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## Revisiting the Alluaudite $\text{NaMnFe}_2(\text{PO}_4)_3$ Sodium Insertion Material: Structural, Diffusional and Electrochemical Insights

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### Abstract

Among the gamut of sodium battery insertion materials,  $\text{NaMnFe}_2(\text{PO}_4)_3$  was reported as the first alluaudite framework compound albeit with poor electrochemical activity [Chem. Mater. 22 (2010) 5554]. We hereby report auto-combustion synthesis of carbon coated  $\text{NaMnFe}_2(\text{PO}_4)_3$  alluaudite and its  $\text{Na}^+$  diffusion, ionic conductivity and electrochemical activity synergizing experiments with bond valence site energy (BVSE) modelling. It registered a 2.8 V redox activity with a reversible capacity of  $\sim 60 \text{ mAh g}^{-1}$  with good cycling stability. BVSE calculations revealed an exceptionally low one-dimensional migration barrier of 0.31 eV for Na-ion diffusion. It was in sync with the low activation energy barrier of 0.162 eV derived from ac impedance spectroscopy.  $\text{NaMnFe}_2(\text{PO}_4)_3$  alluaudite cathode was found to have conductivity value of  $0.5 \times 10^{-6} \text{ S cm}^{-1}$  at room temperature. Among the  $\text{PO}_4$ -based sodium battery insertion materials,  $\text{NaMnFe}_2(\text{PO}_4)_3$  alluaudite shows excellent ionic conductivity with very low  $\text{Na}^+$  migration barrier. It can lead to the realization of superior reversible capacity in this alluaudite cathode comparable to  $\text{LiFePO}_4$ .

**Keywords:** Sodium-ion battery, alluaudite, combustion, bond valence site energy, conductivity.

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