ABSTRACT BOOK

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Effect of Li Substitution on the Magnetic Properties of $Na_{1-x}Li_xFeSO_4F$ by Mössbauer spectroscopy

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Iron based fluorosulfate, MFeSO$_4$F (M=Na, Li) compounds has been known as theoretical positive electrode material with high operating voltage [1]. The addition of Li ions engenders a charge difference and modification to the dimensionality of the lattice along with an redox potential, thus offering the new architectures and electrochemical behavior [2]. In this study, the crystal and magnetic structure of $Na_{1-x}Li_xFeSO_4F$ fluorosulfates has been investigated with the XRD, VSM and Mössbauer analysis. The $Na_{1-x}Li_xFeSO_4F$ (x=0.01, 0.05, 0.1) samples were synthesized by ionothermal method due to the poor hydrolytic stability. The prepared $Na_{1-x}Li_xFeSO_4F$ samples have monoclinic strucutres with space group of $P2_1/c$. The lattice constants and volume of the $Na_{1-x}Li_xFeSO_4F$ samples decreased with increasing Li concentrations. According to the temperature dependent magnetic susceptibility of $Na_{1-x}Li_xFeSO_4F$ samples showed abnormal antiferromagnetic behaviors with decreasing Néel temperture (T_N) due to Li substitution. For $Na_{1-x}Li_xFeSO_4F$, the T_N ordering temperatures, determined to be 33, 32.5, and 32 K for x=0.01, 0.05, and 0.1, respectively. In order to investigate the magnetic structure in terms of Fe nucleus, temperature dependent Mössbauer spectrum recorded at various temperatures ranging from 4.2 to 295 K. The Mössbauer spectrum of $Na_{1-x}Li_xFeSO_4F$ at room temperature consists of two doublets, indicating the existence of Fe^{2+} sites. Each spectrum of $Na_{1-x}Li_xFeSO_4F$ in the T< T_N was fitted to two set(A, B-site) of asymmetrical 8 lines, considering the magnetic hyperfine field(H_{hf}) and electric quadrupole splitting(ΔE_Q) interaction. The H_{hf} and ΔE_Q values of $Na_{0.95}Li_{0.05}FeSO_4F$ at 4.2 K were determined to be $H_{hf,A}$ = 245 kOe, $\Delta E_{Q,A}$ = 2.76 mm/s and $H_{hf,B}$ = 312 kOe, $\Delta E_{Q,B}$ = 2.77 mm/s. From these results, these chagnes in $Na_{1-x}Li_xFeSO_4F$ are originated from the strong electric crystalline field which can be enhanced the asymmetric Mössbauer spectra below T_N.

Keywords: Antiferromagnetism, Cathode, Mössbauer spectroscopy

[1] TRIPATHI, R., RAMESH, T. N., ELLIS, B. L., NAZAR. L. F. 2010. Scalable Synthesis of Tavorite LiFeSO$_4$F and NaFeSO$_4$F Cathode Materials. *Angew. Chem. Int. Ed*., 49, 8738-8742.

[2] XIE, Y., YU, H. T., YI, T. F., WANG, Q., SONG, Q. S., LOU, M., ZHU, Y. R. 2015. Thermodynamic stability and transport properties of tavorite LiFeSO$_4$F as a cathode material for lithium-ion batteries. *J. Mater. Chem. A*, 3, 19728-19737.

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