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Magnetic properties of NaFe$_{0.9}$Mn$_{0.1}$PO$_4$
by Mössbauer spectroscopy

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Maricite-type NaFePO$_4$ is promising for use as the cathode in Na-ion batteries because it is advantageous of environmental friendliness and low-cost. However, it has low electrochemical conductivity and poor performance than other materials. Mn-based phosphate has higher redox reduction potential than Fe-based phosphate. Therefore, NaMnPO$_4$ can obtain higher potential than NaFePO$_4$. In this paper, we have substituted other transition-metal ions such as Mn ions for Fe sites and investigated the hyperfine electromagnetic interaction of Fe ions. The crystal structure and magnetic properties of the as prepared materials were studied by X-ray diffraction (XRD), vibrating sample magnetometer (VSM), and Mössbauer spectroscopy. The NaFe$_{0.9}$Mn$_{0.1}$PO$_4$ sample was prepared using the ball mill method. Structure refinement of NaFe$_{0.9}$Mn$_{0.1}$PO$_4$ was analyzed using Fullprof program. The crystal structure of NaFe$_{0.9}$Mn$_{0.1}$PO$_4$ sample was found to be orthorhombic with space group of Pnmb. Lattice parameters of NaFe$_{0.9}$Mn$_{0.1}$PO$_4$ are as follows: $a_0 = 6.866$ Å, $b_0 = 8.988$ Å, $c_0 = 5.047$ Å, and $V = 311.544$ Å$^3$. The zero-field-cooled (ZFC) and field-cooled (FC) curves were examined by VSM at 100 Oe from 4.2 to 295 K. The magnetic susceptibility curves showed that antiferromagnetic behavior below Néel temperature ($T_N = 14$ K). We have investigated the magnetic hyperfine interaction using Mössbauer spectroscopy at various temperatures between 4.2 and 295 K. At 4.2 K, the magnetic hyperfine field ($H_{hf}$), the electric quadruple splitting ($\Delta E_Q$), and isomer shift ($\delta$) are found to be $H_{hf} = 166.09$ kOe, $\Delta E_Q = 2.18$ mm/s, and $\delta = 1.24$ mm/s. The room-temperature Mössbauer spectrum showed one-doublet with measured values of $\Delta E_Q = 2.20$ mm/s and $\delta = 1.08$ mm/s. We confirmed that $T_N$ of NaFe$_{0.9}$Mn$_{0.1}$PO$_4$ are lower than those of pure NaFePO$_4$ ($T_N = 15$ K). This is due to the Fe-O-Mn superexchange interaction being lower than that of the Fe-O-Fe link.

Fig. 1. Mössbauer spectra of NaFe$_{0.9}$Mn$_{0.1}$PO$_4$ at various temperature range from 4.2 to 295 K.