

Electrochemical sodium-ion intercalation at graphite electrode

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Summary

Sodium-ion battery (SIB) is widely focused on as one of the post lithium-ion batteries (LIBs) from the view of element strategy. When graphite, which is a typical negative electrode for LIBs was used for SIBs, negligible sodium-ion reversible capacity was observed. However, the reason why sodium-ion insertion at graphite electrode hardly occurs has not been clarified yet. In this study, sodium-ion intercalation behaviour at graphite electrode was investigated to understand the sodium-ion intercalation mechanism. X-ray diffraction pattern and Raman spectra indicated that Na⁺ intercalated into only the surface region of natural graphite. This result suggests that the diffusion of Na⁺ in graphite was very slow.

Experimental

Electrochemical measurements were carried out using a three-electrode cell. A working electrode was natural graphite composite electrode. A reference electrode was Ag/Ag⁺ electrode and a counter electrode was also natural graphite composite electrode. The electrolyte solution was 0.9 mol kg⁻¹ sodium bis(fluorosulfonyl)amide (NaFSA)/ethylene carbonate (EC) + dimethyl carbonate (DMC) (1:1 by vol.). Cyclic voltammetry and charge–discharge measurement were carried out. To characterize the reaction products, Raman spectroscopy and X-ray diffraction (XRD) measurement were conducted.

Results

In cyclic voltammograms, clear reversible sodium-ion intercalation behaviour was not indicated. In charge–discharge curves, reversible capacity was around 5 mA h g⁻¹. Hence, only a small amount of reversible sodium-ion intercalation into graphite was indicated. XRD patterns of graphite electrodes held at –3.03 V vs. Fc/Fc⁺ during from 1 to 7 days were almost the same with that of pristine graphite electrode. Figure 1 shows Raman spectra of the graphite electrode held at –3.03 V vs. Fc/Fc⁺. After 1 day, G-band around 1580 cm⁻¹ was split and a new band around 1600 cm⁻¹ derived from graphite intercalation compound (GIC) [1] appeared. The intensity of the new band increased with increasing holding time, and the formation of low stage Na-GIC was indicated. Therefore, the formation of low stage Na-GIC was limited on the surface region of graphite at –3.03 V vs. Fc/Fc⁺. It suggested that the diffusion of sodium-ion in graphite is quite slow.

References:

[1] M. S. Dresselhaus and G. Dresselhaus., *Adv. Phys.*, 30 (1981) 139.

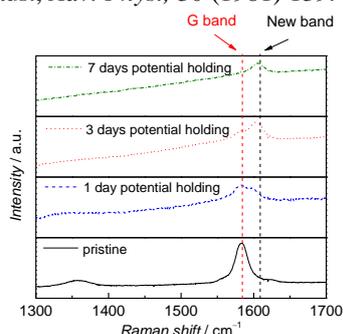


Fig. 1 Raman spectra of natural graphite electrodes after potential holding for various days.