Electric Double Layer Capacitance of Nitrogen-rich C/N Material

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1. Introduction

Several kinds of carbon/nitrogen (C/N) materials were prepared and their properties such as electrochemical capacitance were investigated [1]. Nitrogen-rich C/N materials could be prepared by chemical reactions using nitrogen-rich starting materials [2,3].

In the present study, new nitrogen-rich C/N materials, which was prepared by pyrolysis of 2,3,6,7-tetracyano-1,4,5,8-tetraazanaphtha lene (TCNA), have been applied for electrochemical capacitor.

2. Experimental

The starting material TCNA were heated at a temperature between 670K and 1270K for 1 hour under N_2 atmosphere. The obtained C/N materials were pulverized into fine powders under 45 μ m and mixed with PTFE and acetylene black (C/N materials : PTFE : acetylene black = 8 : 1 : 1 by weight). The mixed powders were pasted on two Pt plates to make electrodes (WE and CE) for the capacitor. The electrochemical measurements were performed by 3 electrode cell with a saturated calomel electrode as a reference in 1M-H₂SO₄ aqueous solution. The specific capacitances of the materials were calculated from the charge/discharge curves by galvanostatic method. Cyclic voltammograms were also measured.

3. Results and Discussion

The largest capacitance was observed for the C/N material heat-treated at 1070K, as is shown in Figure 1. The material treated at 1070K had the largest specific surface area ($990 \text{ m}^2/\text{g}$) among the materials. The largest capacitance, however, could not be explained only by the specific surface area, because of the nonlinear relationship between them. ESCA measurements suggested the existence of quaternary or pyridine-type nitrogen in the material heat-treated at 1070K.

Figure 2 shows galvanostatic charge/discharge curves of C/N material prepared at 1070K. Specific capacitance was calculated from this curve to be 185F/g (per single electrode of capacitor), which is larger than that of activated carbon with the same specific surface area.

Figure 3 shows a relation between specific capacitance and current density. Almost no change in the specific capacitance was observed by using different current densities for galvanostatic method. Cyclic voltammetry suggested a reversible pseudo-faradaic reaction was observed at lower potentials in addition to a typical faradaic reaction at higher potentials.

References

- [1] For example: M. Kodama, et al., Mat. Sci. Eng. B-Solid, **108**, 156 (2004).
- [2] M. Kawaguchi, and K. Nozaki, *Chem. Mater.*, **7**, 257(1995).
- [3] M.Kawaguchi, et.al., Chem.Lett., 1003(1997).

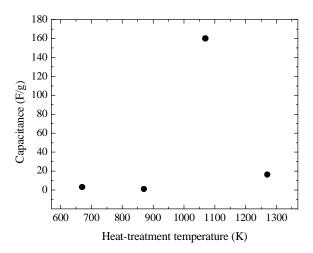


Figure 1 Relation between specific capacitance and heat-treatment temperature for the C/N materials.

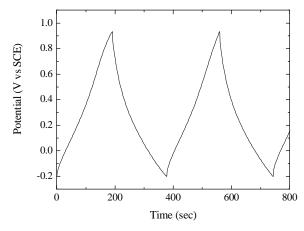


Figure 2 Galvanostatic charge/discharge curves of C/N material prepared by the heat-treatment of TCNA at 1070K. Current density: $20~\text{mA/cm}^2$.

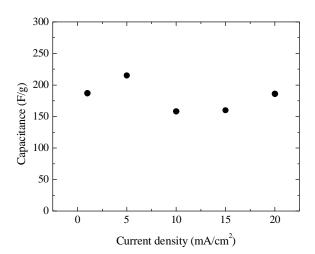


Figure 3 Relation between specific capacitance and current density on galvanostatic charge/discharge for the C/N materials prepared by the heat treatment of TCNA at 1070K.