Application of vertically aligned carbon nanotubes for electric double layer capacitors

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Electric double layer capacitors (EDLCs) are promising energy storage devices that exhibit high power density and long cycle life. EDLCs have been mainly used as memory back-up devices and are expected as power sources of electric vehicles in the future. However, the energy density of EDLCs is smaller than that of the secondary batteries. Accordingly, the development of EDLCs with both high power density and high energy density is necessary.

Activated carbons have been widely used as polarized electrodes of EDLCs. Although they have very high specific surface area, most of their pores are micropores, which do not contribute to capacitance. On the other hand, carbon nanotubes (CNTs) are attractive as electrodes of EDLCs. CNTs possess mesopores between the tubes and inside the tube, which are readily accessible to ions. However, the capacitance of EDLCs fablicated using CNTs electrodes is not enough. As for fabrication of CNTs electrodes, two kinds of methods have been reported to date. One is the assembling of CNTs on conducting substrates by using binder material [1], where CNTs formed by various methods are harvested. The other is the direct growth of CNTs on conducting substrates [2]. The latter preparation is particularly important for low internal resistance and high capacitance of EDLCs.

In this study, we have successfully synthesized vertically aligned CNTs on metal substrates by thermal chemical vapor deposition method. A gas mixture of C_2H_2 and He was introduced into a chamber as a source gas. The growth temperature was 700℃. Before the CNTs growth, Fe/Al thin films were deposited on Ta substrate by RF magnetron sputtering. Figure 1 shows a SEM image of the resultant CNTs. As a result of SEM and TEM observation, it was found that the diameter, length and density of the CNTs were about 20 nm, 80 μ m and 10^{10}cm^{-2}, respectively.

A test cell for the EDLC was fabricated with two CNTs electrodes separated by thin cellulose film in 1M $TEABF_4$/PC electrolyte. The test cell was charged to 2.5 V at a constant current, and then was discharged to 0 V at a constant current. Therefore, it was found that the EDLC using the CNTs electrodes exhibited stable charge/discharge characteristic, as shown in Figure 2. The specific capacitance exhibited 6.5 F·g^{-1}.

Furthermore, the CNTs electrodes were annealed at 550℃ in air for 10 minutes. The EDLC using the annealed electrodes exhibited the specific capacitance of 7.6 F·g^{-1}. According to TEM observation, it was found that many tips of CNTs were open. As shown in Figure 3, even after 10,000 cycles, the decrease of the specific capacitance didn't appeared.

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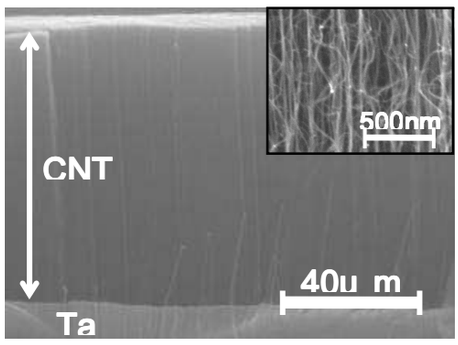

Fig. 1. SEM image of the resultant CNTs. The inset is a higher magnification image.

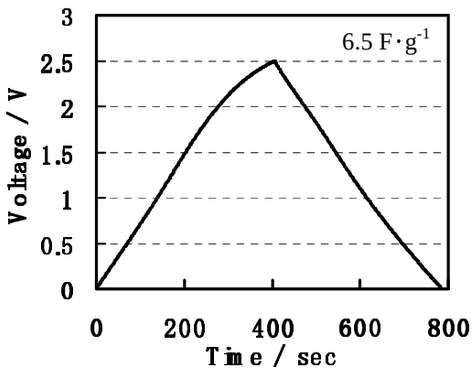

Fig. 2. Charge/discharge characteristic of an EDLC using the CNTs electrodes. Constant current : 0.05 mA.

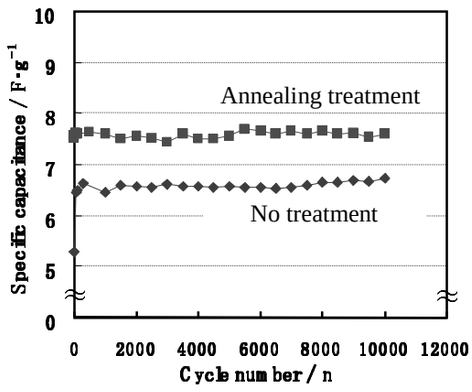

Fig. 3. Correlation between cycle number and specific capacitance. Annealing condition : 550℃ in air, 10 min.