PREPARATION AND CHARACTERIZATION OF SUPERCAPACITORS MANUFACTURED FROM NITROGEN-ENRICHED CARBON

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INTRODUCTION

The presence of heteroatoms and functional groups in a carbon matrix change the electron/donor characteristics of the carbon electrode materials resulting in the improvement of the capacitive behavior [1].
For the first time we have manufactured the capacitors from a carbon derived from melamine with the moderate amount of nitrogen embedded in the carbon matrix.

EXPERIMENTAL

Melamine/mica composite was prepared by the polymerization of melamine in the interlayer spaces of mica in formaldehyde in the ratio of melamine/ mica/ formaldehyde = 3/ 1/ 8 by weight at pH~ 9.1 and temperatures of 80 ºC. After drying at 60 ºC, samples were carbonized in an infrared furnace under the nitrogen atmosphere and temperatures of 650 ºC, 750 ºC, 800 ºC, 850 ºC, and 1000 ºC. Each sample was also stabilized in air at 250 ºC for 4h prior to the carbonization. Samples subjected to no-stabilization process are hereafter denoted as Me650, Me750, Me800, Me850, and Me1000; their stabilized counterparts with the‘s’ subscript, e.g. Me650s.

RESULTS AND DISCUSSION

Table 1 shows the results on the elemental compositions of each sample evaluated by CHN combustion method and XPS as well as specific surface areas calculated from α_s plots by subtracting pore effect (SPE) method. The nitrogen amounts of stabilized samples are higher compare to their non-stabilized counterparts while the porosities are less developed. N_{1s} core level peak analyses [2] of all samples revealed that the main nitrogen species are pyridinic (398.5± 0.2 eV), quaternary (401.2± 0.2 eV), and oxidized nitrogen (402.9± 0.2 eV) and that the stabilized samples contained high ratios of pyridinic nitrogen located at the periphery of the graphene sheets (Figure1).
The specific gravimetric capacitances (C_g) and specific capacitances per surface areas (C_{SA}) of capacitors built from each sample and evaluated from galvanostatic charge/discharge cycling measurements are summarized in Table 2. Concerning the C_g of non-stabilized samples, it is remarkable that the values are unexpectedly high with respect to the low porosities, e.g. 204.8 Fg^{-1} for Me750. C_g values of stabilized samples are close to the non-stabilized samples despite the remarkably lower surface areas. C_{SA} of the stabilized samples are however generally higher. We suggest that pyridinic nitrogen species affect the electron donor-acceptor characteristic of carbon materials and that the pseudocapacitive attraction between the protons of electrolyte and the carbon electrode materials occurred. This became more significant in the stabilized samples.

Sample	N/C_{CHN}	N/C_{XPS}	SA[m^2g^{-1}]
Me650	0.37	0.22	260
Me750	0.24	0.20	442
Me800	0.24	---	345
Me850	0.20	0.21	248
Me1000	0.08	0.11	86
Me650s	0.45	0.45	138
Me750s	0.43	0.34	256
Me800s	0.41	---	202
Me850s	0.32	0.25	120
Me1000s	0.13	0.14	17

Table 1. N/C ratios of non-stabilized and stabilized samples evaluated from CHN and XPS; and surface areas of all samples.

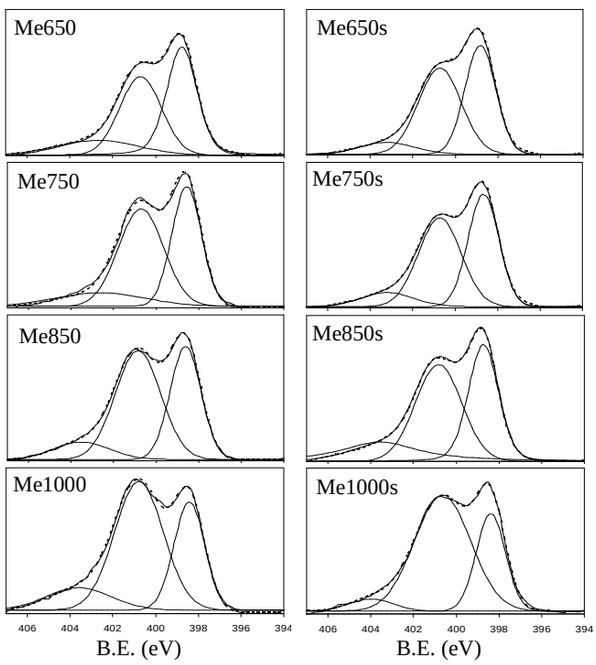

Figure 1. N_{1s} core level peak analyses of non-stabilized and stabilized samples. Pyridinic, quaternary, and oxidized nitrogen are detected.

Sample	C_g [Fg^{-1}]	C_{SA} [Fm^{-2}]
Me650	141.1	0.54
Me750	204.8	0.46
Me800	198.8	0.57
Me850	157.4	0.63
Me1000	47.92	0.55
Me650s	128.2	0.93
Me750s	200.1	0.78
Me800s	185.6	0.92
Me850s	195.9	1.63
Me1000s	62.24	3.66

Table 3. Specific gravimetric capacitances (C_g) and specific capacitances per surface area (C_{SA}) of all samples. Calculations are done from the discharge process of the third cycle in the potential range of 0.2-0.1V; surface areas are estimated by SPE method.

REFERENCES

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2. Raymundo-Pinero, E.; Cazorla-Amoros, D.; Linares, S.A.; Find, J.; Wild, U.; Schlogl, R. *Carbon* **2002**, *40*, 597