STRUCTURAL AND ELECTROCHEMICAL PROPERTIES OF NITROGEN-ENRICHED MESOPOROUS CARBON

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INTRODUCTION

Template carbonization method has been regarded as unique and versatile technique for providing well-designed nanostructure to the carbon body[1,2]. We have already reported nitrogen-enriched mica template carbon exhibits high double layer capacitance in H_2SO_4[3]. Because of non-porous characteristic of it, specific capacitance per unit surface area could reach ~$2.2F\cdot m^{-2}$, which is over ten times as high as the capacitance of commercially available activated carbons. Accordingly, we aim at obtaining the synergy effect of mesoporous structure and nitrogen doping on EDLC performance. The present study describes synthesis, structural characterization and EDLC performance of nitrogen-enriched mesoporous carbons prepared by a template method.

EXPERIMENTAL

Nitrogen-enriched mesoporous carbon has been prepared from quinoline polymerized pitch using laboratory made mesoporous silica as a template. Quinoline pitch was synthesized according to previously reported method by Mochida et al.[4]. The obtained pitch was impregnated at 573K into the pore of SBA-15. The resulted pitch/silica composite was heat-treated at 1023K in N_2 atmosphere and then washed by HF for removing silica. Samples for EDLC experiments were molded from a mixture of template carbon (80wt%), Teflon® powder (10wt%) and acetylene black (10wt%). Prepared electrodes were evaluated using three electrodes cell in 1M H_2SO_4.

RESULTS AND DISCUSSION

Two types of negative replica carbons were prepared using quinoline pitch (designated as MC-Q) and divinylbenzene (as MC-D). The nitrogen content of MC-Q and MC-D prepared at 1073K were ascertained to be 5.1 and 0wt%, respectively. Figure 1 shows nitrogen adsorption isotherms onto SBA-15 silica, MC-Q and MC-D. SBA-15 indicates typical isotherm for mesoporous materials having relatively large and uniform mesopores. The isotherm of MC-D displays developed micropores and disordered mesostructure. In case of the MC-Q, the isotherm shows presence of smaller mesopores. Pore size distribution of SBA-15 and MC-Q is also shown in the inset graph in Fig.1. Both of them indicate narrow size distribution, while the average pore size is markedly different. Figure 2 shows SEM images of MC-Q prepared at 1073K. An enlarged micrograph reveals the mesostructure of MC-Q to be an ordered hexagonal assembly of fibrous carbon having ca. 6~7nm in diameter. Consequently, inter-fiber gap can act as mesopores. The results chronopotentiometrically collected at $20mA\cdot g^{-1}$ demonstrated reproducible curve as shown in Fig.3. The gravimetric capacitance was found to be $170F\cdot g^{-1}$ (MC-D) and 289 $F\cdot g^{-1}$ (MC-Q). Thus, the capacitance of nitrogen-enriched carbon was about 1.7 times as high as that of ordinary carbon from divinylbenzene. Furthermore, being considered with lower surface area of MC-Q, the obtained capacitance is notably high ($0.40F\cdot m^{-2}$). Such a high capacitance appears to be due to the presence of residual nitrogen in carbon. Although the mechanism of implication of nitrogen for the capacitance is still left uncertain, owing to pseudo capacitive additional effect, the nitrogen-enriched mesoporous carbon can be presumed as a promising electrode material.

REFERENCES

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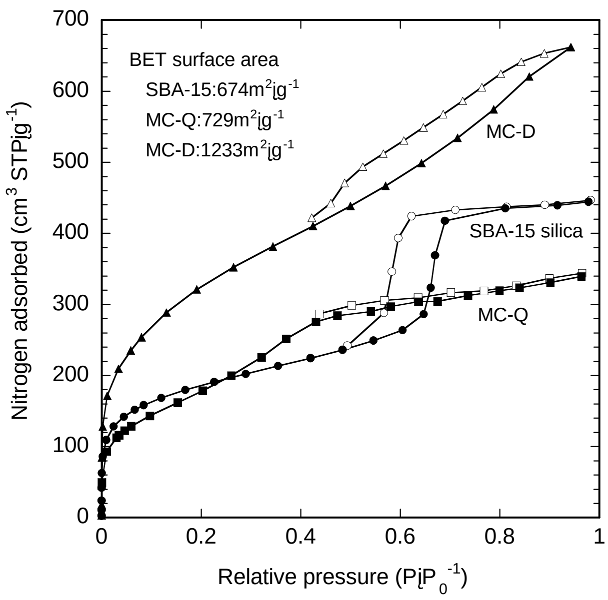

Figure 1. Adsorption isotherms of N_2 at 77K for SBA-15 silica and template carbons. The inset is pore size distribution plot calculated by DH-method.

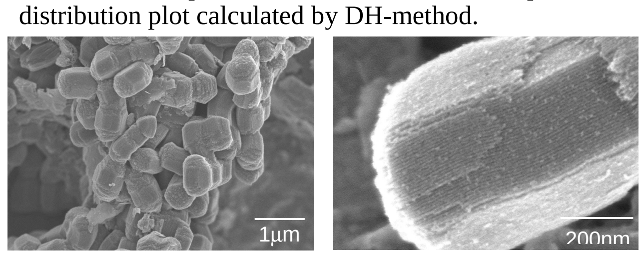

Figure 2. Scanning electron micrographs of MC-Q prepared from quinoline pitch in SBA-15 silica template.

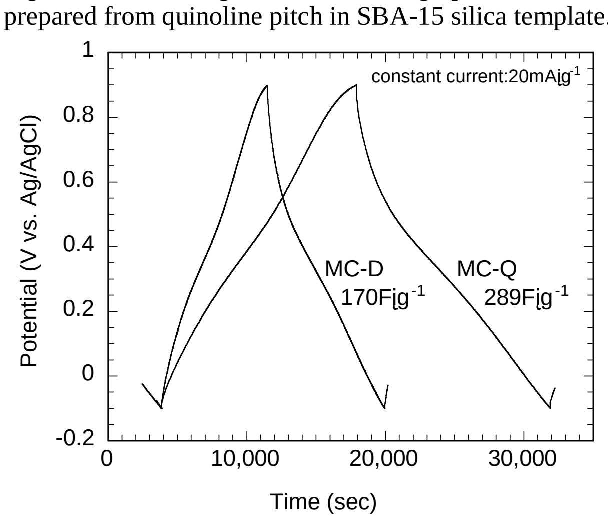

Figure 3. Galvanostatic charge/discharge curves of MC-D and MC-Q in 1M H_2SO_4 electrolyte.