Influence of Nitrogen Surface Chemistry in Electric Double Layer Capacitance of Nitrogen Doped Ordered Mesoporous Carbon

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Surface heteroatoms such as oxygen have been shown to have significant effect on electrical double-layer capacitance (EDLC) of carbon [1]. Oxygen groups can increase the wettability of the carbon and have pseudocapacitive effect due to formation of reversible quinone/hydroquinone complex [1]. However, surface oxygen groups can also promote carbon corrosion leading to decrease in capacitance overtime [2]. Nitrogen doped carbon has been shown to be highly resistant to corrosion [3]. Nitrogen functional groups in carbon are proposed to have pseudocapacitive effect [4], however, the influence of nitrogen surface groups on the fundamental behavior of the EDLC of carbon is not well understood.

Nitrogen doped ordered mesoporous carbon (NOMC) synthesized by template based method have uniform pore size distribution. When SBA-15 is used as a template, NOMC having well connected pore system can be synthesized [5]. Hence, in NOMC casted on SBA-15 surface area, pore size and pore interconnectivity could be well controlled so that effect of surface chemistry could be studied independently.

Here, SBA-15 was prepared first by self-assembly of triblock copolymer Pluronic® P123 ($E_{20}PO_{70}EO_{20}$, BASF) and tetraethyl orthosilicate (TEOS) in 2 M HCl. Pyrrole was then vacuum infiltrated into the template, polymerized to polypyrrole, and carbonized at high temperature for 3 h in N_2 atmosphere. Finally, the template was removed in hot 10 M KOH. The NOMC obtained was divided into four batches and heated to 800, 1000, 1200 and 1400 °C to remove oxygen groups formed during KOH treatment and modulate surface nitrogen content. The porosity and surface chemistry of NOMC were characterized using the N_2 isotherm and X-ray photoelectron spectroscopy. The EDLC was measured in a custom built three-electrode cell using cyclic voltammetry (CV).

In Figure 1, typical CV curves at different scan rates for NOMC are shown. The ideal rectangular shape demonstrates double layer charge storage/discharge mechanism. Hence, EDLC can be calculated using the equation;

Current (i) = scan rate (υ)×double layer capacitance (C_{DL})

One of the interesting findings was that the presence of nitrogen groups led to very high EDLC in NOMC (156 F/g) with respect to Vulcan carbon (11.6 F/g). The effect of nitrogen and oxygen heteroatoms on EDLC of NOMCs was studied and it was found that the EDLC was mostly dominated by the nitrogen groups rather than the oxygen groups. Hence, the role of nitrogen in EDLC of NOMC will be discussed and an enhancement mechanism will be proposed.

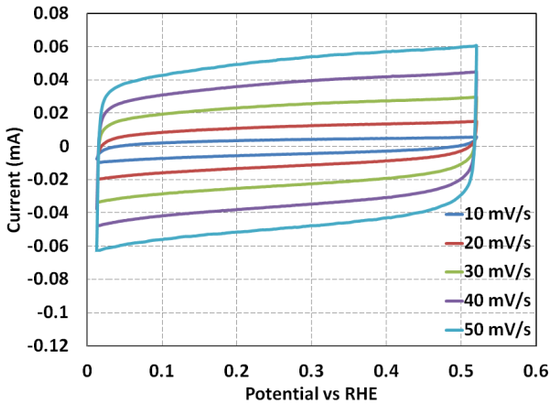

Figure 1. Typical CVs of nitrogen doped ordered mesoporous carbon (NOMC) taken in N_2 saturated 0.5 M H_2SO_4 at 25 °C with different scan rates.

References

(1) E. Frackowiak, *Phys. Chem. Chem. Phys.*, 9 (**2007**) 1774-1785.
(2) P. Simon and Y. Gogotsi, *Nature Mater*, **7** (**2008**) 845-854.
(3) S. Shrestha and W. E. Mustain, *J. Electrochem. Soc.* 157 (**2010**) B1665-B1672.
(4) F. Béguin, K. Szostak, G. Lota, and E. Frackowiak, *Adv. Mater.*, 17 (**2005**) 2380-2384.
(5) R. Ryoo, S. H. Joo, M. Kruk, and M. Jaroniec, *Adv. Mater.*, 13 (**2001**) 677-681.