Superior Adsorption Capacity of Mesoporous Carbon Nitride with Basic CN Framework for Phenol

Enamul Haque,
" Jong Won Jun," Siddulu Naidu Talapaneni," Ajayan Vinu" and Sung Hw
a Jhung $^{\ast^a}$

^a Department of Chemistry, Kyungpook National University, Daegu 702-701, Korea. Fax: 82-53-950-6330; E-mail: <u>sung@knu.ac.kr</u>

^b International Center for materials nanoarchitectonics (MANA), World Premier International (WIP) Research Center, National Institute for Materials Science, 1-1 Namaki, Tsukuba 305-0044, Japan.

Supplementary Information

Synthesis and characterization procedure of MCN-1:

Tetraethyl orthosilicate, ethylenediamine (EDA), carbon tetrachloride (CTC), and tri-block copolymer poly (ethylene glycol)-*block*-poly(propylene glycol)-*block*-poly(ethylene glycol) (Pluronic P123, molecular weight = 5800, EO₂₀PO₇₀EO₂₀ were obtained from Aldrich.

Mesoporous carbon nitride materials were prepared by using mesoporous silica SBA-15-100 as template. In a typical synthesis, 0.5 g of calcined SBA-15 was added to a mixture of EDA (1.35 g) and CTC (3 g). The resultant mixture was refluxed and stirred at 90 °C for 6 hours. Then, the obtained dark brown colored solid mixture was placed in a drying oven for 12 hours, and ground into fine powder. The template-carbon nitride polymer composites were then heat treated in a nitrogen flow of 50 ml per minute at 600 °C with the heating rate of 3.0 °C min⁻¹ and kept under these conditions for 5 h to carbonize the polymer. The mesoporous carbon nitride was recovered after dissolution of the silica framework in 5 wt % hydrofluoric acid, by filtration, washed several times with ethanol and dried at 100 °C.

The powder X-ray diffraction (XRD) pattern of mesoporous carbon nitride materials was collected on a Rigaku diffractometer using CuK α ($\lambda = 0.154$ nm) radiation. The diffractogram was recorded in the 2 θ range of 0.8 to 10 ° with a 2 θ step size of 0.01 and a step time of 1 s. Nitrogen adsorption and desorption isotherms was measured at -196 °C on a Quantachrome Autosorb 1 sorption analyzer. All samples were outgassed at 250 °C for 3 h prior to the nitrogen adsorption measurements. The specific surface area was calculated using the Brunauer-Emmett-Teller (BET) method. The pore size was obtained from the adsorption branch of the nitrogen isotherms by Barrett-Joyner-Halenda method.

Regeneration procedure of spent MCN-1:

The used MCN-1 adsorbent was activated with deionized water under ultrasound. MCN-1 (0.01 g) was taken in a glass vial and mixed with 25ml deionized water. Then ultrasound (35% of maximum power 750 W) was irradiated for 60 min to that mixture with an ultrasonic generator (VCX750, Sonics & materials). After separation by centrifugation, the adsorbent was dried (100 °C) overnight and reused for the next adsorption.

Adsorbent	Items	Fresh	1 st Reuse	2 nd Reuse
MCN-1	q _e (mg/g)	583	542	495
	k_2 (g/mg·h)	2.03	1.32	0.55

Supplementary Table S1. The equilibrium adsorbed amount (q_e) and pseudo-second-order kinetic constant (k_2) of fresh and reused MCN-1 (Temperature: 25 °C, C_i : 400 ppm).



Supplementary Fig. S1. Effect of contact time and initial phenol concentrations on the adsorption of phenol over the three adsorbents: (a) $C_i=200$ ppm; (b) $C_i=300$ ppm; (a) $C_i=400$ ppm.



Supplementary Fig. S2. The plots of pseudo-second-order kinetics for phenol adsorption over the three adsorbents at several initial phenol concentrations: (a) 200 ppm; (b) 300 ppm; and (c) 400 ppm.



Supplementary Fig. S3. (a) Effect of contact time on the phenol adsorption and (b) Pseudo-second-order plots to show the re-usability of a spent MCN-1.