Supplementary Material

Modelling of borrowing hydrogen amination reactions of alcohols and amines in NaOH- or KOH-containing media over metal-free ordered mesoporous nitrogenous carbon catalyst

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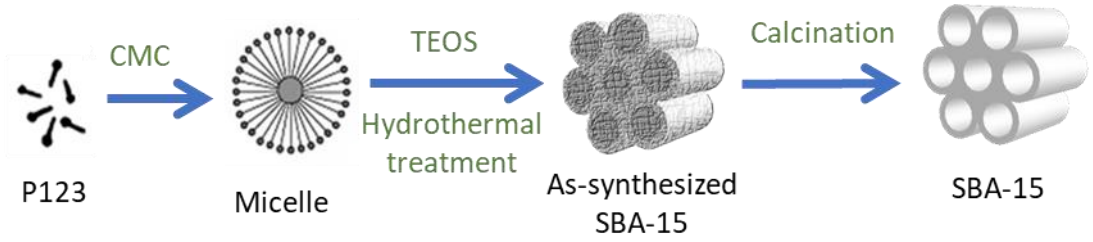
EXPERIMENTAL PROCEDURE

Synthesis of SBA-15

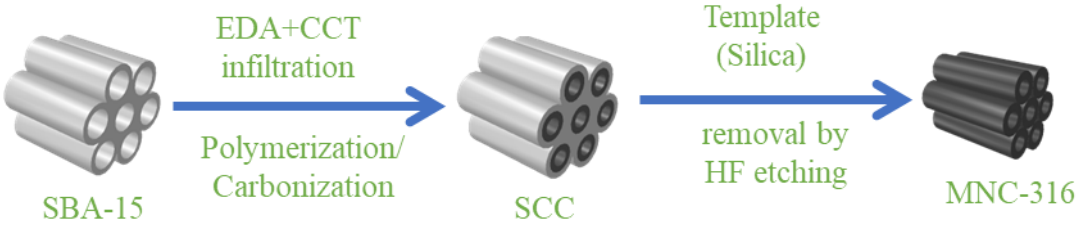
The graphical depiction of the synthesis of template SBA-15 is shown in scheme S1. In a typical synthesis of SBA-15, 1.0 *g* of P123 triblock copolymer was dissolved in 2.0 *mL* of HCl, and 20.0 *mL* water, and was stirred for 1 *h* at 40 ℃. To this, 2.5 *mL* of TEOS was added and the solution was stirred for 1 day at 35 ℃. The resultant gel solution was subjected to hydrothermal treatment for another 24 *h* at 100 ℃. The obtained precipitate was filtered and dried at 60 ℃. The as-synthesized sample was then subjected to calcination at 550 ℃ for 6 *h* in the air to remove the template.



Scheme S1. A generalized representation of the formation of ordered mesoporous Silica (SBA-15) by liquid crystal templating method.

Synthesis of MNC-316

MNC-31 was synthesized by mixing ordered mesoporous silica SBA-15 (1.0 *g*) with 4.4 *g* ethylene diamine and 10.8 *g* carbon tetrachloride (Scheme S2). This mixture was subjected to reflux at 90 ℃ for 6 *h* to obtain a dark brown solid mixture. This solid mixture was grounded to a fine powder after drying and then pyrolyzed at 600 °C, for 6 *h* in an argon atmosphere with a heating rate of 5 °C min^{-1}. The silica template was etched by stirring in HF solution for 24 *h*, followed by washing with copious distilled water and ethanol, then dried to obtain MNC-316.



Scheme S2. A generalized representation of the formation of ordered mesoporous nitrogenous carbon (MNC-316) by nano-casting method.

Catalyst Characterization

All the samples were systematically characterized using various analytical, and microscopy techniques. The Powder X-ray diffraction (XRD); Rigaku Miniflex II X-ray diffractometer with a Cu Kα radiation (λ = 1.5406 *Å*) with an operating voltage of 30 *kV* and a current of 15 *mA, and* a scanning speed of 0.5°min^{-1}. Nitrogen adsorption-desorption isotherms were measured at 77 *K* on a Micrometrics ASAP 2020 apparatus. The specific surface area of the samples was calculated using the Brunauer-Emmet-Teller (BET) method, and pore size distribution curves were obtained from the analysis of the desorption branch of nitrogen adsorption isotherms by Barrett-Joyner-Halenda (BJH) method. High-resolution transmission microscopy (TEM) images were carried out on a 2100 JEOL microscope operated at 200 *kV.* The samples for TEM studies are prepared by coating the carbon on a 200-mesh lacey foam-coated copper grid. Figures S1-S3 present the XRD, BET, and TEM figures, respectively.

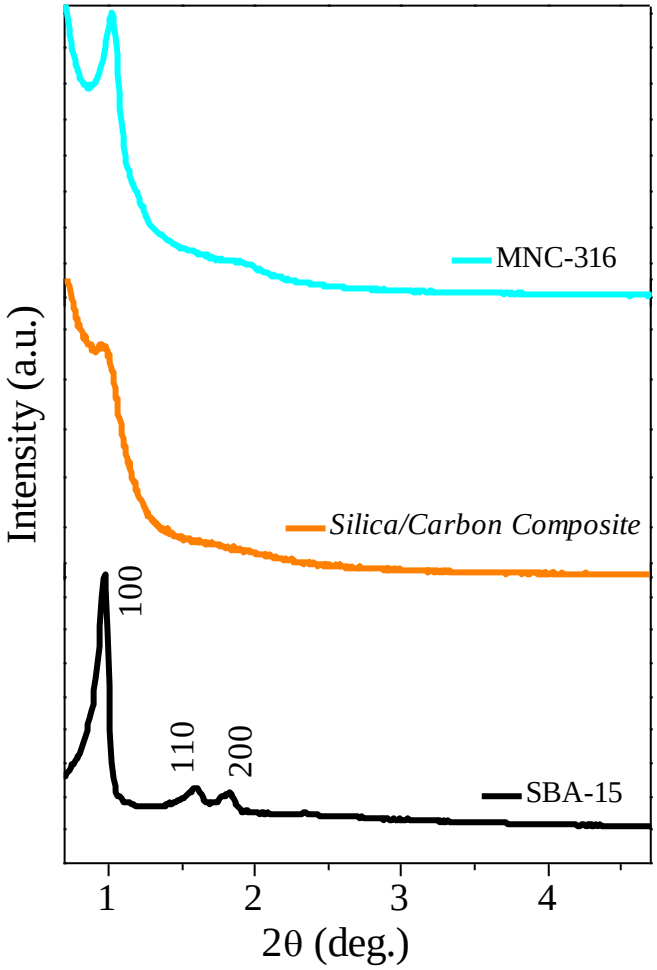


Figure S1. Low-angle XRD of SBA-15, Silica-Carbon composite and MNC-316.

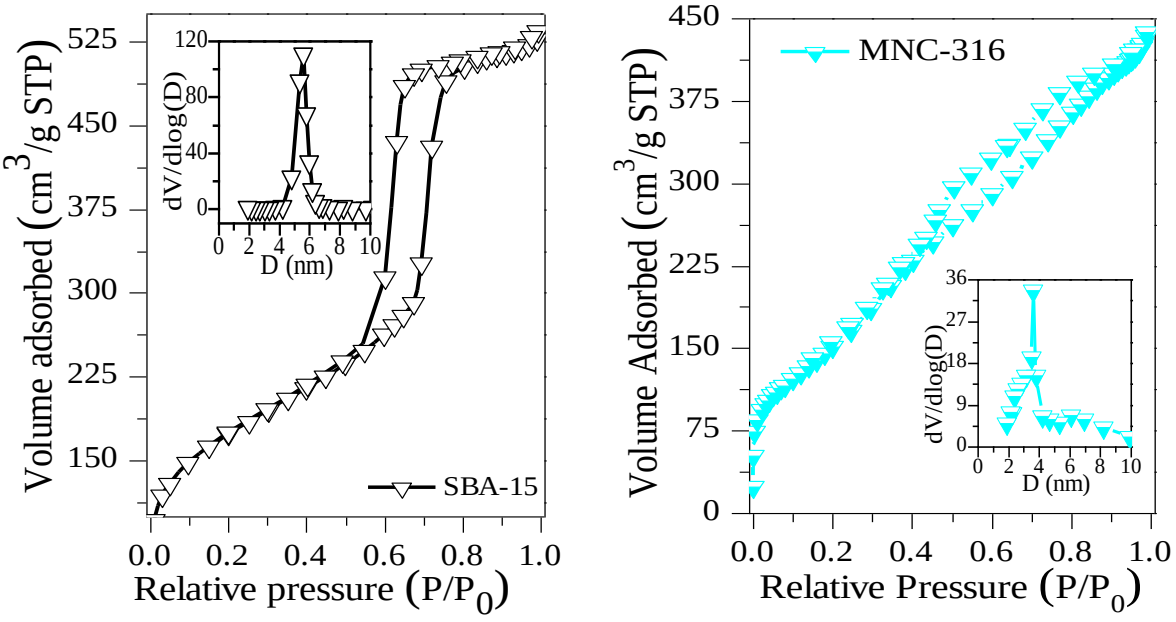


Figure S2. N_2 sorption isotherms and corresponding pore size distributions of SBA-15 and MNC-316.

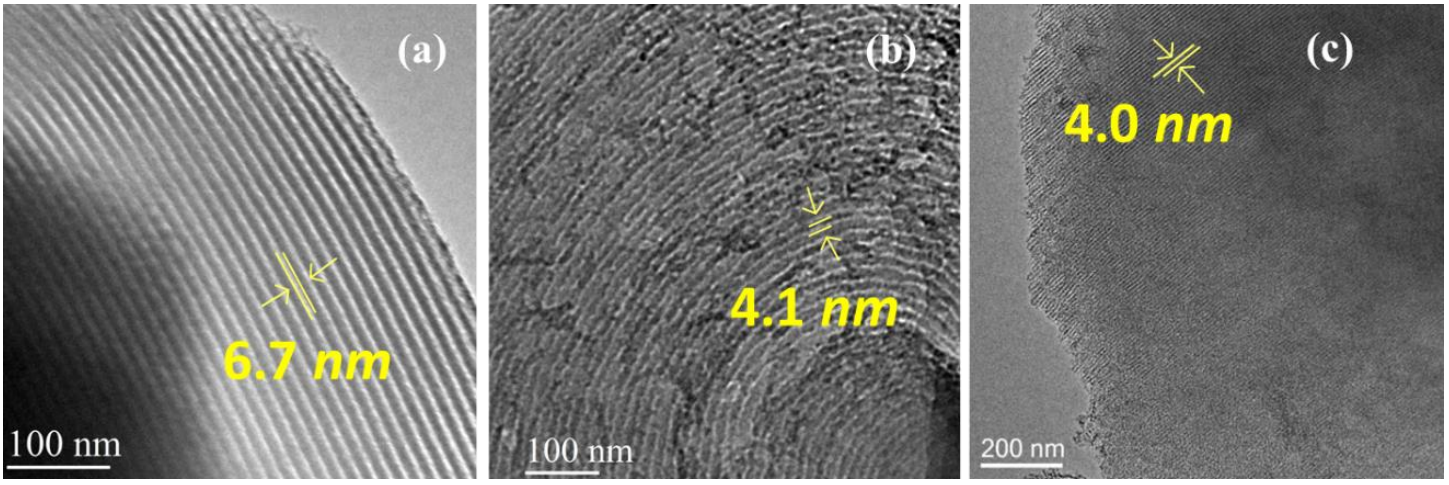


Figure S3. TEM images of (a) SBA-15, (b) MNC-316, (c) MNC-316 (recycled).

COMPUTATIONAL DETAILS

The raw data for the DFT calculations is provided in the zip file Raw_Data_DFT.zip, which can be requested from the corresponding author (using the email address on the cover page of this article). This file contains the Excel files, Adsorption_Energy.xls and Transition_State.xls, which contain the energy of reactants, transition states, and products used to calculate the adsorption energies and energy barriers shown in Figure 2. These energies are extracted from the CP2K output files cp2k_job.out, which are stored in the folders provided within the zip file.

