Supporting Information

Metal organic framework derived mesoporous carbon nitrides with a high specific surface area and chromium oxide nanoparticles for CO₂ and hydrogen adsorption

Stalin Joseph,^a Devaraju M. Kempaiah,^{*a} Mercy Benzigar,^a Arun V. Baskar,^a Siddulu N. Talapaneni,^{*a} Sung Hwa Jhung,^b Dae-Hwan Park,^a and Ajayan Vinu^{*a}

^aFuture Industries Institute (FII), University of South Australia, Mawson Lakes Campus, Mawson Lakes, SA 5095, Australia.

^bDepartment of Chemistry, Green-Nano Materials Research Center, Kyungpook National University, Daegu 41566, Republic of Korea

*To whom the correspondence should be addressed

Email: <u>Ajayan.Vinu@unisa.edu.au</u>

Ph: +61-8-83025384, Fax: +61-8-830-25639

Materials characterization

The powder X-ray diffraction (XRD) measurements were carried out on a PANalytical Empyrean platform diffractometer using Bragg-Brentano geometry. The measurements were collected using Cu K α radiation from a sealed tube source operating at 40 kV and 40 mA, a fixed divergence slit of 0.1 ° and a PIXcel3D detector. The scan rate used was 0.01⁰ sec⁻¹. The structural morphology and elemental analysis of Cr-MCN-10 material was observed by scanning electron microscopy (SEM) (JEOL FEM 7001) with energy dispersive X-ray spectroscopy (EDS). Nitrogen adsorption and desorption isotherms were measured at -196 °C on a Micromeritics ASAP 2420 surface area and porosity analyser. All the samples were degassed for 8 h at 200 °C under a vacuum ($p < 1 \times 10^{-5}$ pa) in the degas port of the adsorption analyser. The specific surface areas of samples was calculated using the BET (Brunner-Emmett-Teller) model in the pressure range where the term $V(1 - P/P_0)$ continuously increases with P/P_0 in the Rouquerol plot and the NLDFT model was used for the pore size distribution analysis. Fourier transform infrared spectra was recorded on a Nicolet 5700 FTIR spectrometer fitted with a diamond attenuated total reflection (ATR) accessory by averaging 200 scans with a resolution of 2 cm⁻¹ measured in absorbance mode. X-ray photoelectron spectroscopy (XPS) was carried out using a Kratos Axis ULTRA X-ray photoelectron spectrometer incorporating a 165 mm hemispherical electron energy analyser. Survey and multi region spectra were recorded at C1s, N1s and Cr2p photoelectron peaks. Each spectra region of photoelectron of interest was scanned several times to obtain good signal to noise ratios. The incident radiation was Monochromatic Al Kα X-rays (1486.6eV) at 225 W (15 kV, 15 ma). Survey (wide) scans were taken at analyser pass energy of 160 eV and multiplex (narrow) high resolution scans at 20 eV. Survey scans were carried out over 1200-0 eV binding energy range with 1.0 eV steps and a dwell time of 100 ms. Narrow high-resolution scans were run with 0.05 eV steps and 250 ms dwell time. Base pressure in the analysis chamber was 1.0 x 10^{-9} torr and reduced to 1.0 x 10^{-8} torr during sample analysis. Atomic concentrations were calculated using the CasaXPS version 2.3.14 software and a Shirley baseline with Kratos library Relative Sensitivity Factors (RSFs). Peak fitting of the high-resolution data was also carried out using the Casa XPS software.

S2



Fig. 1S SEM image of Template MIL-100(Cr)





Fig. 2S Elemental mapping of Cr-MCN-10-1.5





Fig. 3S Elemental mapping of Cr-MCN-10-2.0





Fig. 4S Elemental mapping of Cr-MCN-10-2.5



Fig. 5S TEM images of Cr-MCN-10-2.0



Fig. 6S TEM images of Cr-MCN-10-2.5



Fig. 7S (a) Calculated Rouquerol plot for Cr-MCN-10-1.5, the Rouquerol Plot with the pressure ranges used for the BET surface area calculations(inset) and (b) BET surface area report with a value of BET constant.



Fig. 8S (a) Calculated Rouquerol plot for Cr-MCN-10-2.0, the Rouquerol Plot with the pressure ranges used for the BET surface area calculations(inset) and (b) BET surface area report with a value of BET constant.



Fig. 9S (a) Calculated Rouquerol plot for Cr-MCN-10-2.5, the Rouquerol Plot with the pressure ranges used for the BET surface area calculations(inset) and (b) BET surface area report with a value of BET constant.



Scheme S1. Illustration for the proposed structure of MCN-10