Supplementary Material for PCCP

Supporting Information

One-pot Generation of Mesoporous Carbon Supported Nanocrystalline Calcium Oxides Capable of Efficient CO_2 Capture at a Wide Range of Temperature

*Zhangxiong Wu [1, 2], Na Hao [1], Gongkui Xiao [1], Liying Liu [1], Paul A. Webley [1 *], and Dongyuan Zhao [1, 2 *]*

1. Department of Chemical Engineering, Monash University, Clayton, VIC 3800, Australia.

2. Department of Chemistry and Laboratory of Advanced Materials, Fudan University, Shanghai 200433, P. R. China.

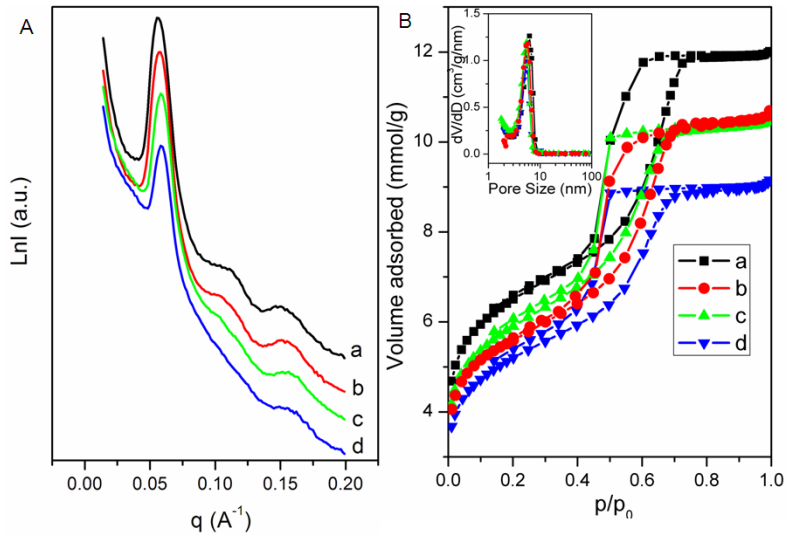

Fig. S1 SAXS patterns (left), N_2 sorption isotherms and the corresponding pore size distribution curves (right) of the mesoporous polymer composites: a) CaO/C-1-350; b) CaO/C-2-350; c) CaO/C-3-350 and d) CaO/C-4-350. The cell parameters and pore sizes of the composites are calculated to be around 12.8 and 6.0 nm, respectively. While about 12.0 and 5.1 nm for the calcium-free polymer sample, indicating that introduction of calcium could probably impose small swellings of the micelles and thus increases of the cell parameters and the pore size.

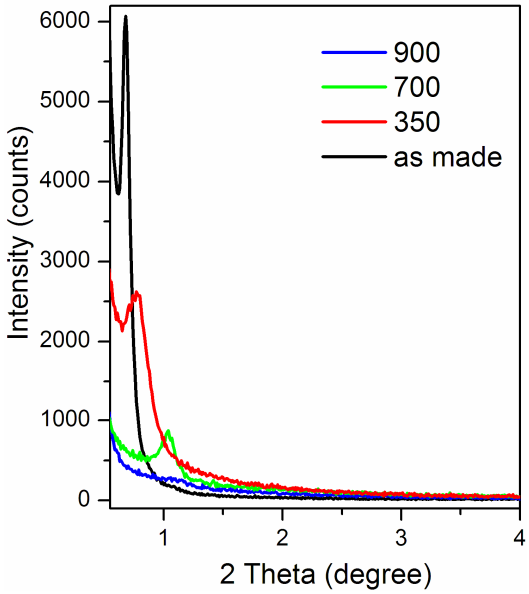

Fig. S2 Small-angle XRD patterns of the as-made mesoporous composites CaO/C-5 and the composites CaO/C-5-T obtained after being pyrolyzed at 350, 700 and 900 °C, respectively. The contents of calcium oxide are up to 28.3 and 34.6 wt % for the samples obtained at 700 and 900 °C, respectively. It is observed that the regularity of mesostructure is well ordered for the as-made sample, but deteriorates significantly at 350 ~ 700 °C and almost collapses at 900 °C, indicating severe oxidation or/and activation of the carbon supports by calcium oxide at higher temperature.

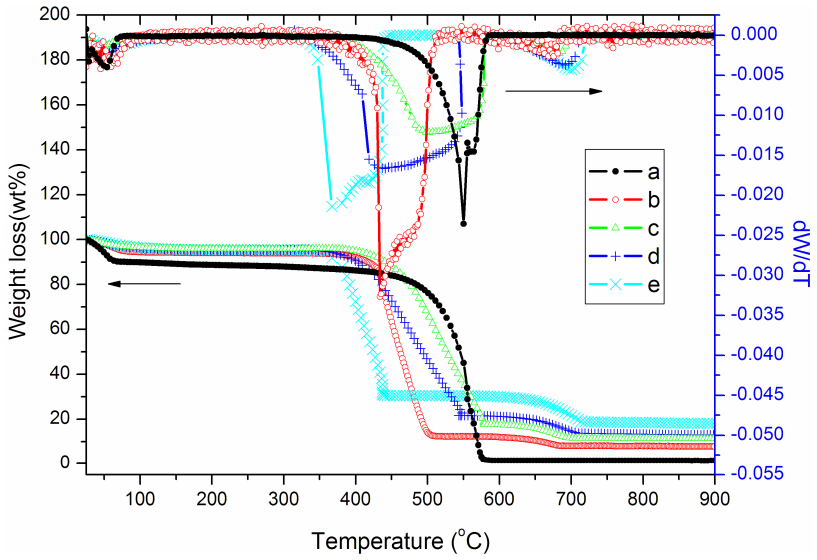

Fig. S3 TG and DTG curves (conducted under O_2) of a) the calcium-free mesoporous carbon sample and the mesoporous calcium oxide/carbon composites with the CaO content of b) 8.0, c) 11.5, d) 13.5 and e) 18.5 wt %. The combustion temperature lowers down considerably from ~ 580 °C for the pure carbon sample to ~ 420 °C for the composites with 18.5 wt % of CaO. During the combustion, calcium carbonate is formed first and then fully decomposed at ~ 680 °C.

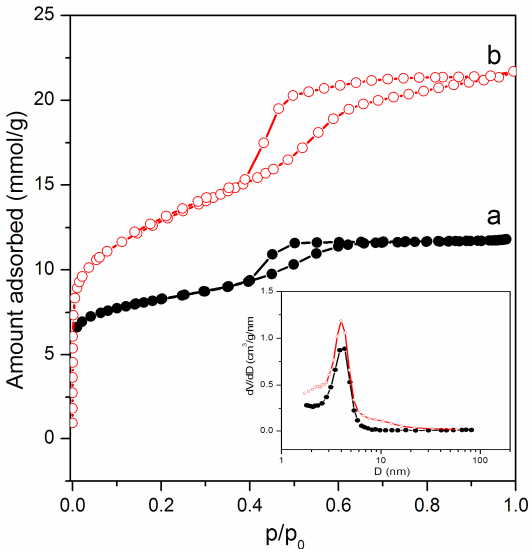

Fig. S4 N_2 sorption isotherms of a) the mesoporous CaO/C composites obtained at 700 °C with CaO content of ~ 14 wt % and b) the corresponding mesoporous carbon material after washing away the calcium component by 2 M HCl solution at 50 °C overnight. High surface area and pore volumes are obtained after the treatment, indicating a good *in-situ* activation effect of the carbon support by calcium oxide at high temperatures.

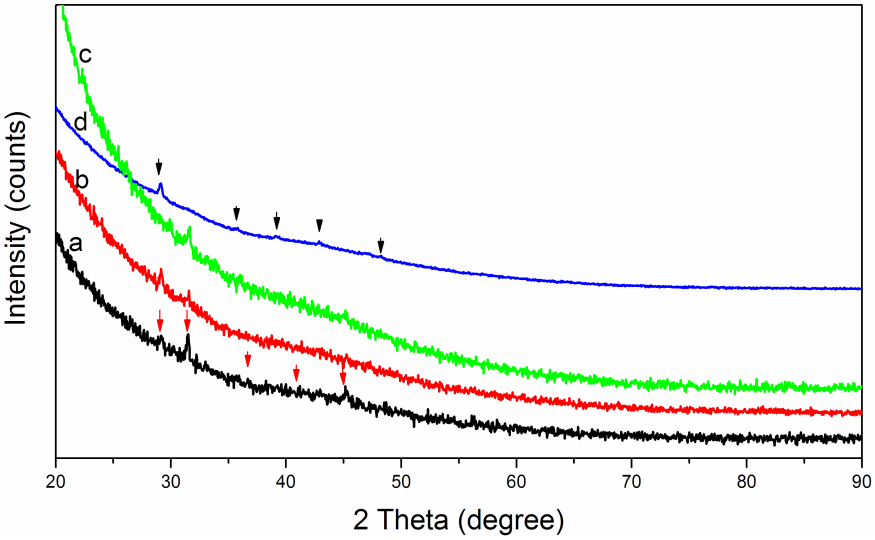

Fig. S5 Wide-angle XRD patterns of the mesoporous composites obtained by pyrolyzing the as-made CaO/C-3 composite at a) 350, b) 400, c) 500 and d) 600 °C under Ar. The marked red arrows are supposed to ascribed to $Ca(NO_2)_2$ (JCPDS 280-232) while the black ones to $CaCO_3$ (JCPDS 862343).

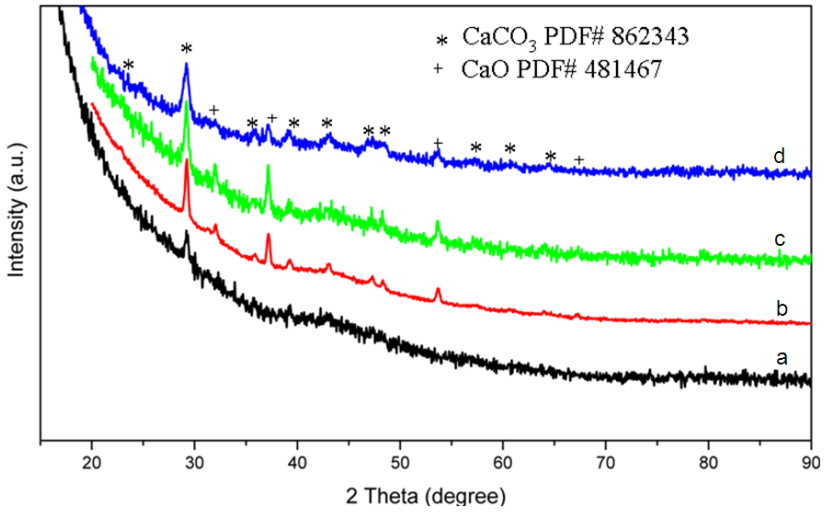

Fig. S6 Wide-angle XRD patterns of the mesoporous composites CaO/C-x-700 obtained by pyrolysis at 700 °C under a gas atmosphere of 2.4 % O_2 in N_2.

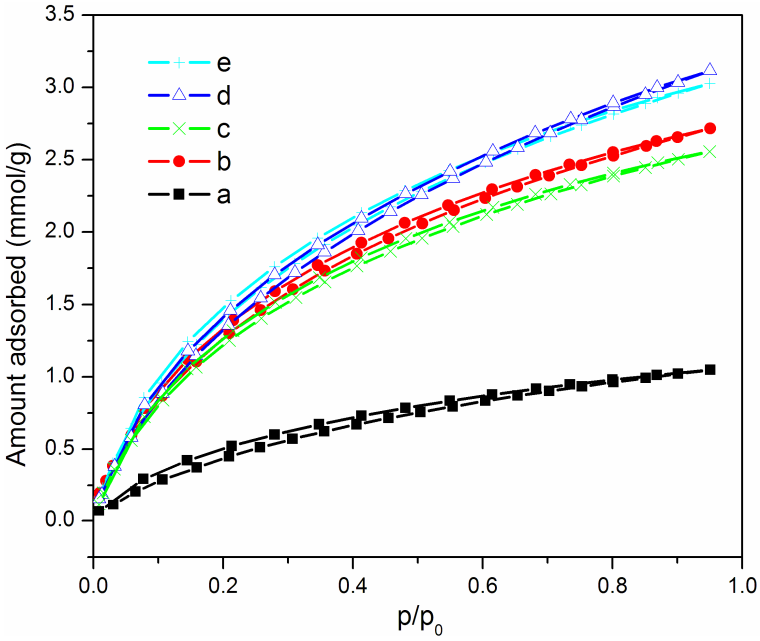

Fig. S7 CO_2 adsorption isotherms at 25 °C of the ordered mesoporous CaO/C composites obtained by heating the as-made CaO/C-3 sample under Ar at a temperature of a) 350, b) 500, c) 600, d) 700 and e) 900 °C, respectively.

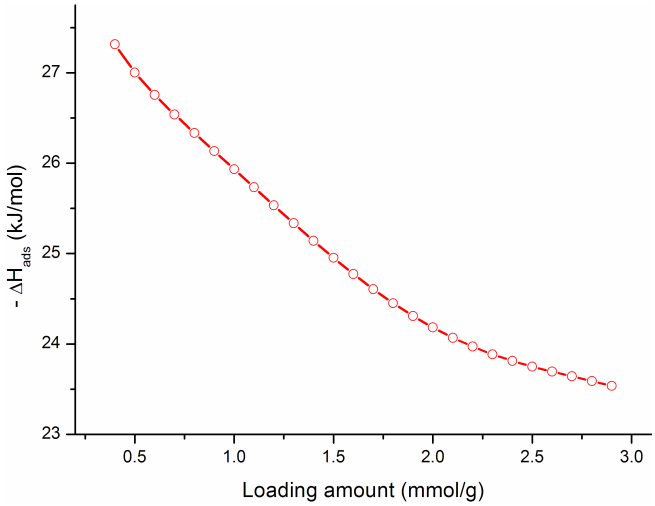

Fig. S8 Dependence of the isosteric heats of adsorption as function with loadings for adsorption CO_2 on the ordered mesoporous composites CaO/C-3-900 with a CaO content of 15.8 wt %.

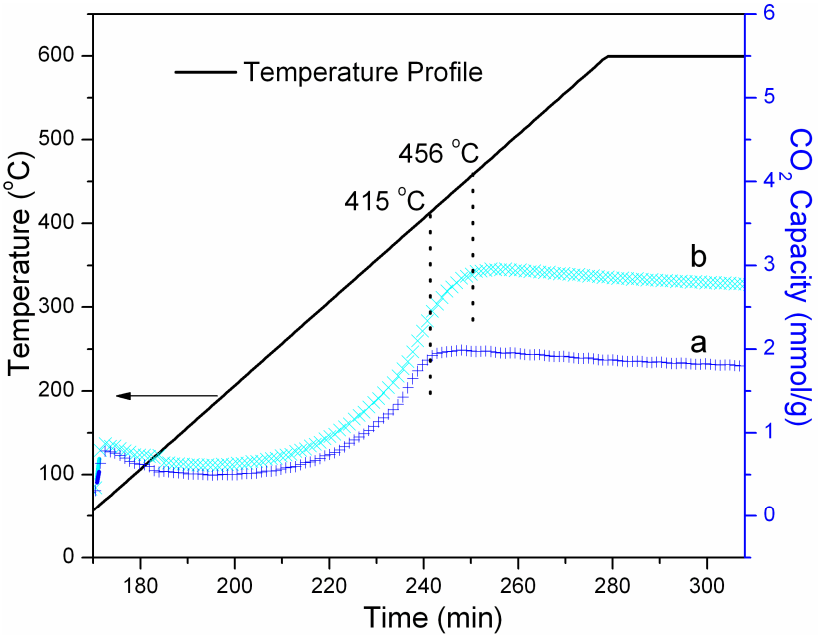

Fig. S9 CO_2 sorption amount as a function of temperature of the mesoporous composites obtained at 700 °C with a calcium content of 13.5 and 18.5 wt %, respectively.

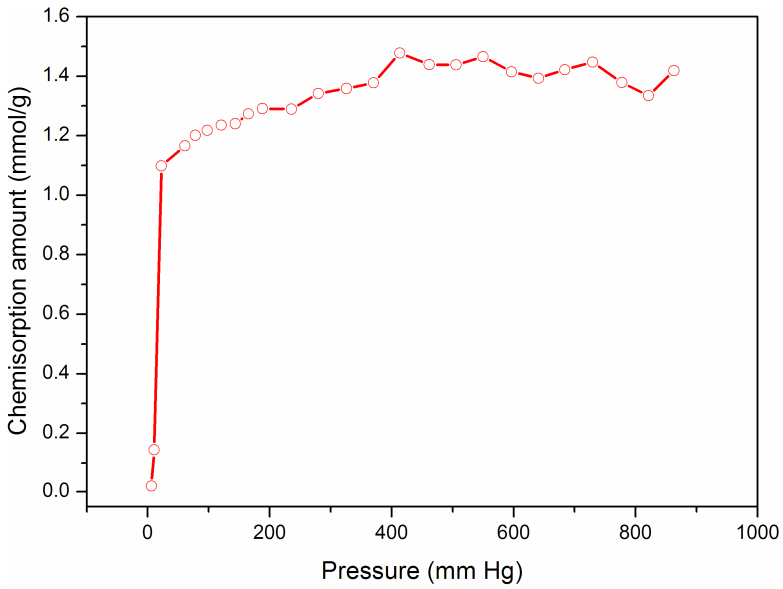

Fig. S10 Chemisorption isotherm of CO_2 at 450 °C of the mesoporous composites CaO/C-4-700 with a CaO content of 18.5 wt %. The capacity is calculated to be ~ 1.5 mmol/g, which is lower than the value obtained by thermogravimetric analysis. The reason is that the activation temperature is only 450 °C limited by the analyzer. In this case, part of the calcium is in the form of carbonate before the analysis.

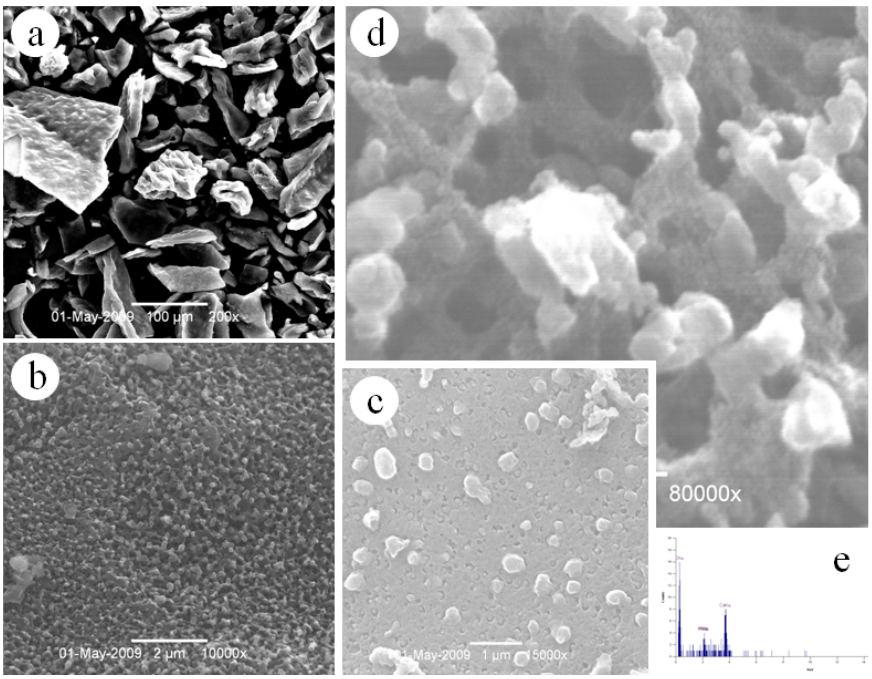

Fig. S11 FE-SEM images (a-d) and EDX spectrum (e) of the residues obtained from the mesoporous composites CaO/C-4-700 after ten cycles of CO_2 absorption and regeneration.

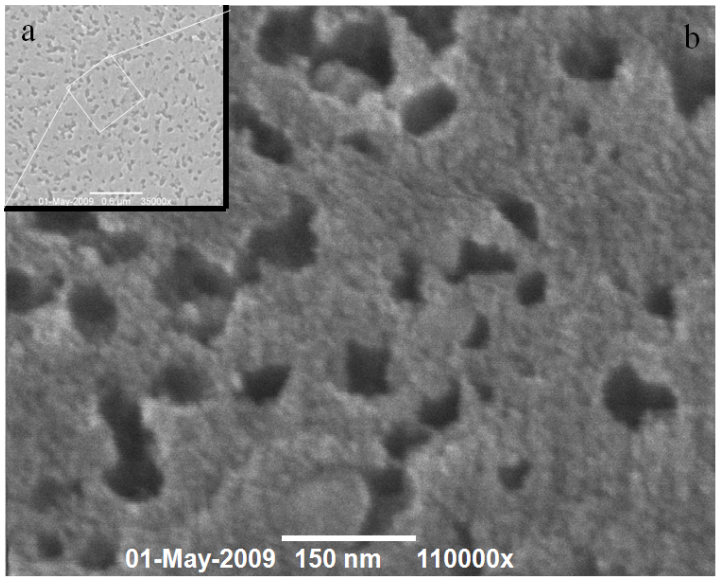

Fig. S12 FE-SEM images of the hierarchically macro-/meso-/micro-porous carbon material after washing away the CaO particles of the composites, which are obtained from the mesoporous composite CaO/C-4-700 after 10 cycles of CO_2 absorption and regeneration.

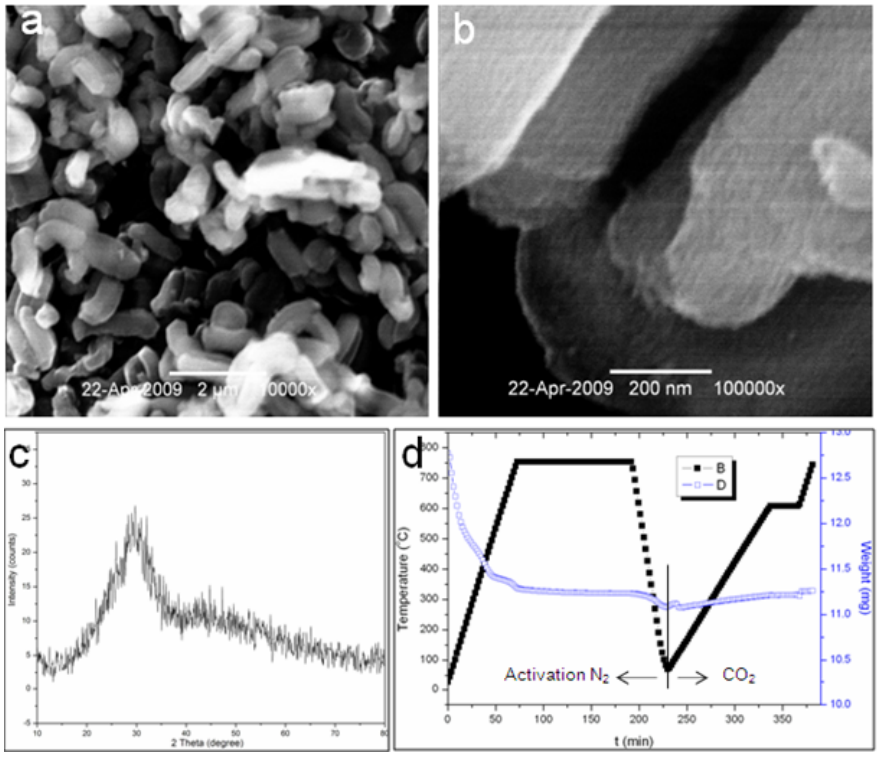

Fig. S13 FE-SEM images (a, b), wide-angle XRD pattern (c) and the temperature-dependent CO_2 sorption behavior (d) of the mesoporous silica SBA-15 loaded with ~ 35 wt % of CaO.