Supporting Information

One-pot Generation of Mesoporous Carbon Supported Nanocrystalline Calcium Oxides Capable of Efficient CO$_2$ Capture at a Wide Range of Temperature

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Supplementary Material for PCCP
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Fig. S1 SAXS patterns (left), N₂ 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 O₂) 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 N₂ 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** CO₂ 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 CO₂ 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.