Electronic Supplementary Information (ESI) for:

Design and synthesis of stable supported-CaO sorbents for CO_2 capture

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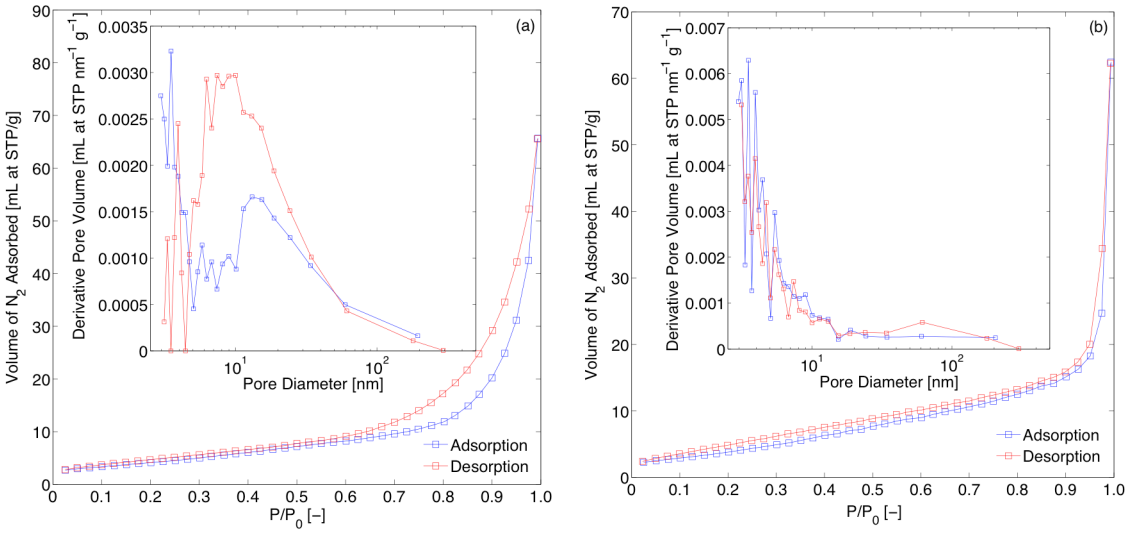

Figure S1. N_2 physisorption isotherms (measured at −196 °C) and pore size distributions (insets) of (a) ***meso*-$Ca_xAl_yO_z$** and (b) **CaO/*meso*-$Ca_xAl_yO_z$**.

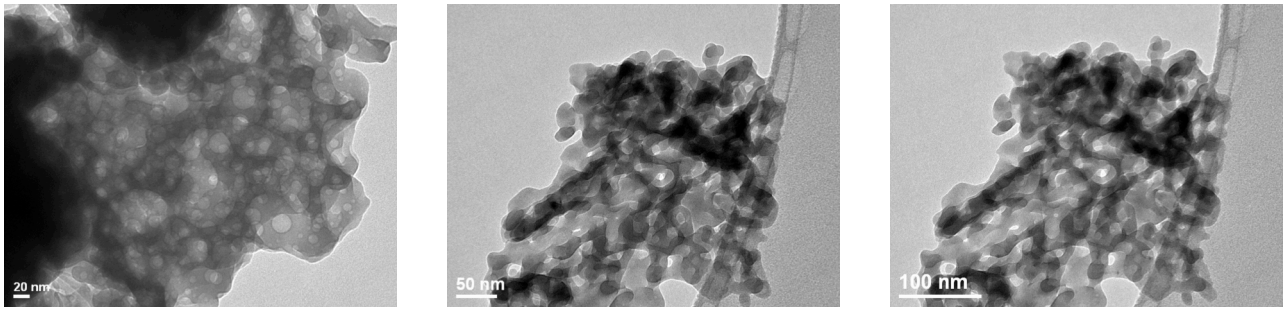

Figure S2. Transmission electron microscope images of ***meso*-$Ca_xAl_yO_z$**.

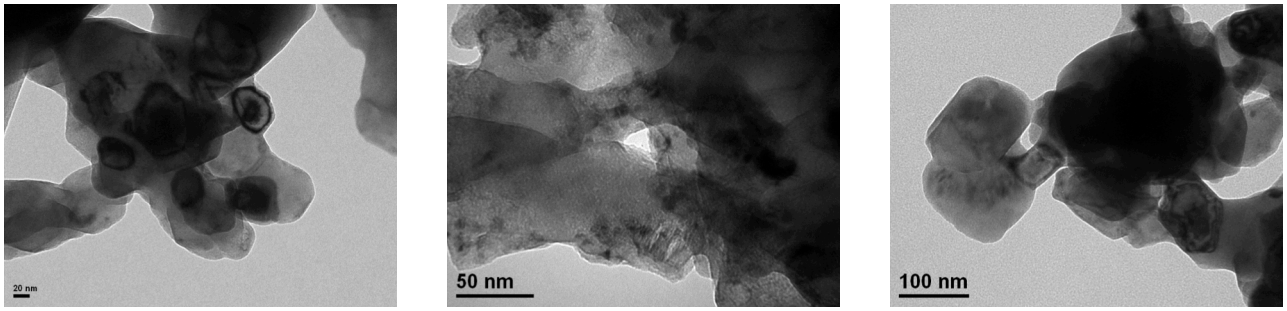

Figure S3. Transmission electron microscope images of **CaO/*meso*-$Ca_xAl_yO_z$**.

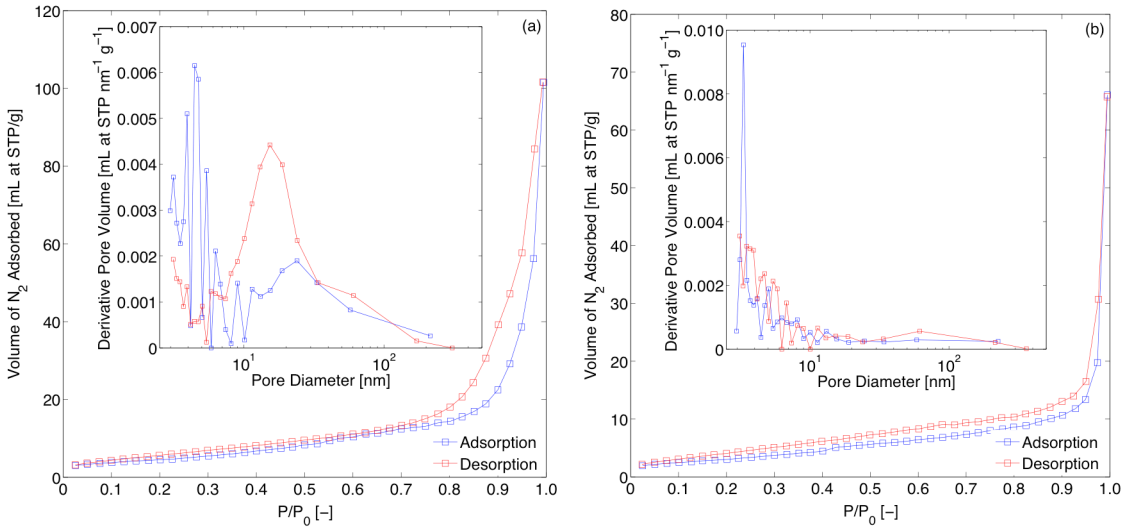

Figure S4. N_2 physisorption isotherms (measured at −196 °C) and pore size distributions (insets) of (a) ***hier*-$Ca_xAl_yO_z$** and (b) **CaO/*hier*-$Ca_xAl_yO_z$**.

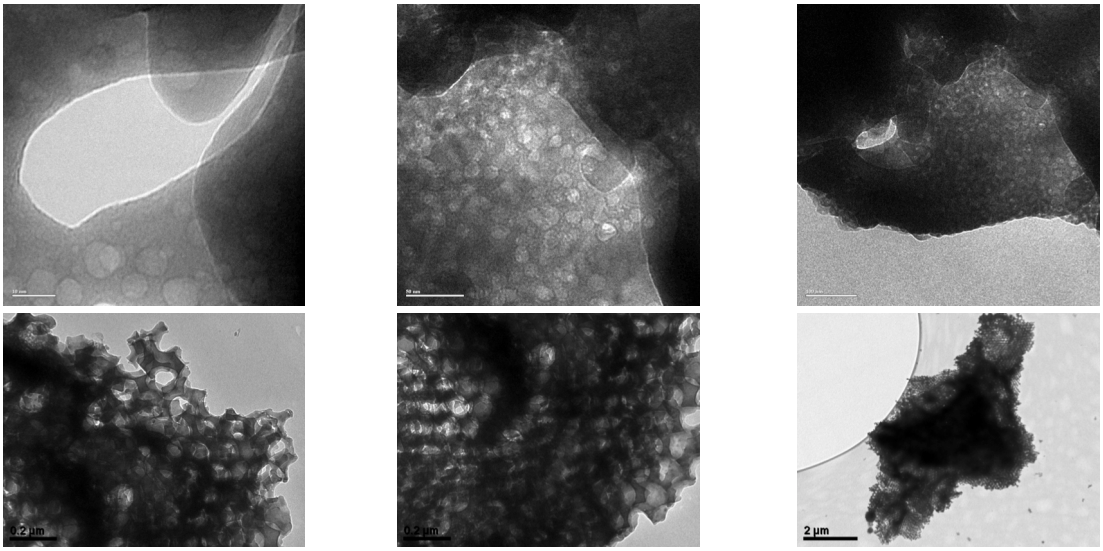

Figure S5. Transmission electron microscope images of ***hier*-$Ca_xAl_yO_z$**.

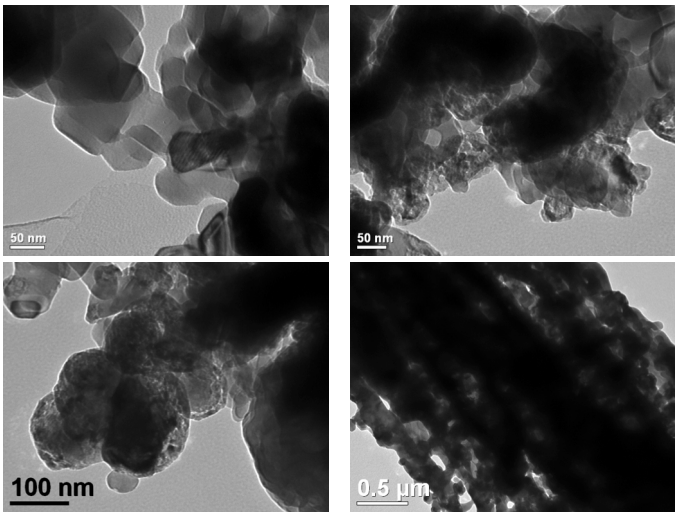

Figure S6. Transmission electron microscope images of **CaO/*hier*-$Ca_xAl_yO_z$**.

2. Characterization data for SiO_2/SiC-based materials

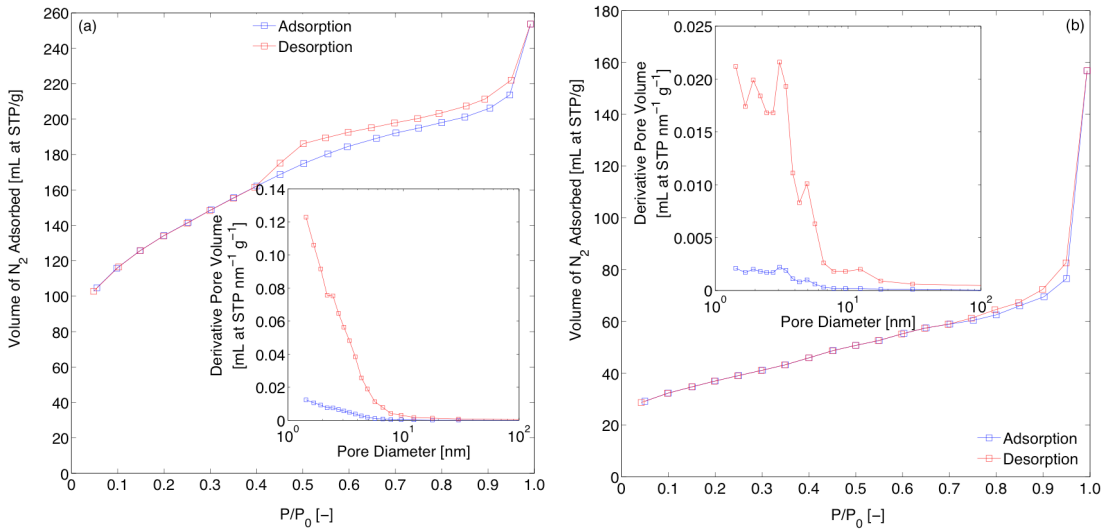

Figure S7. N_2 physisorption isotherms (measured at −196 °C) and pore size distributions (insets) of (a) ***meso*-SiC** and (b) **CaO/*meso*-SiC**.

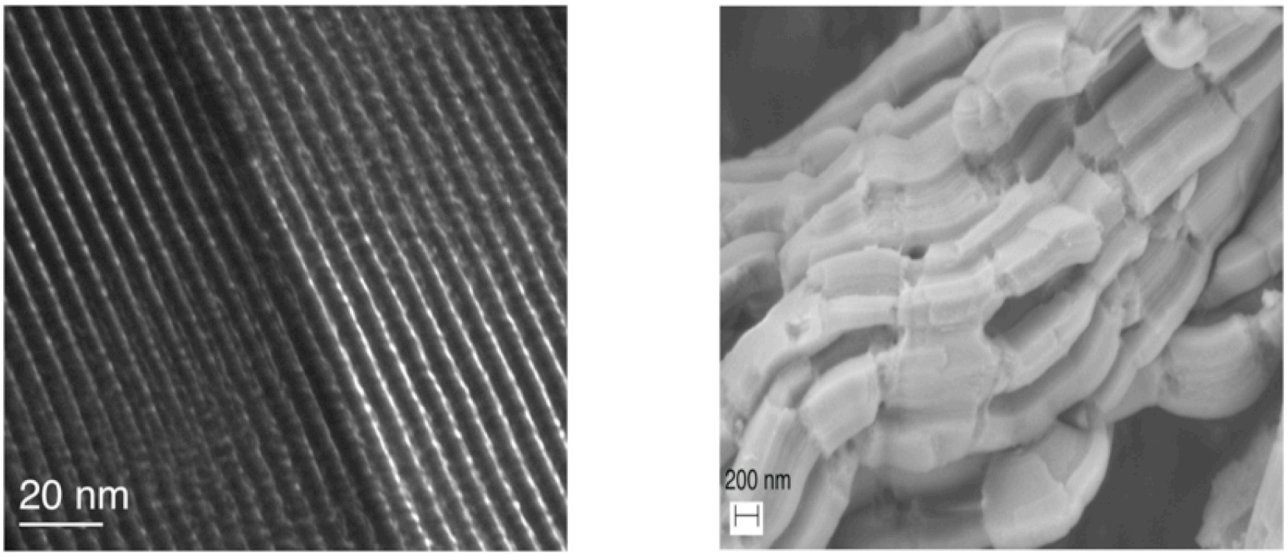

Figure S8. TEM (left) and SEM (right) images of SBA-15. TEM image of SBA-15, showing highly ordered mesoporosity. For TEM, the ultrafine sample that was obtained by sonicating the SBA-15 in ethanol was dispersed on a copper grid for measurement, and imaged using a 120-kV electron beam. For SEM, The sample was put on carbon tape and imaged using a secondary-electron detector and a 2-kV electron beam.

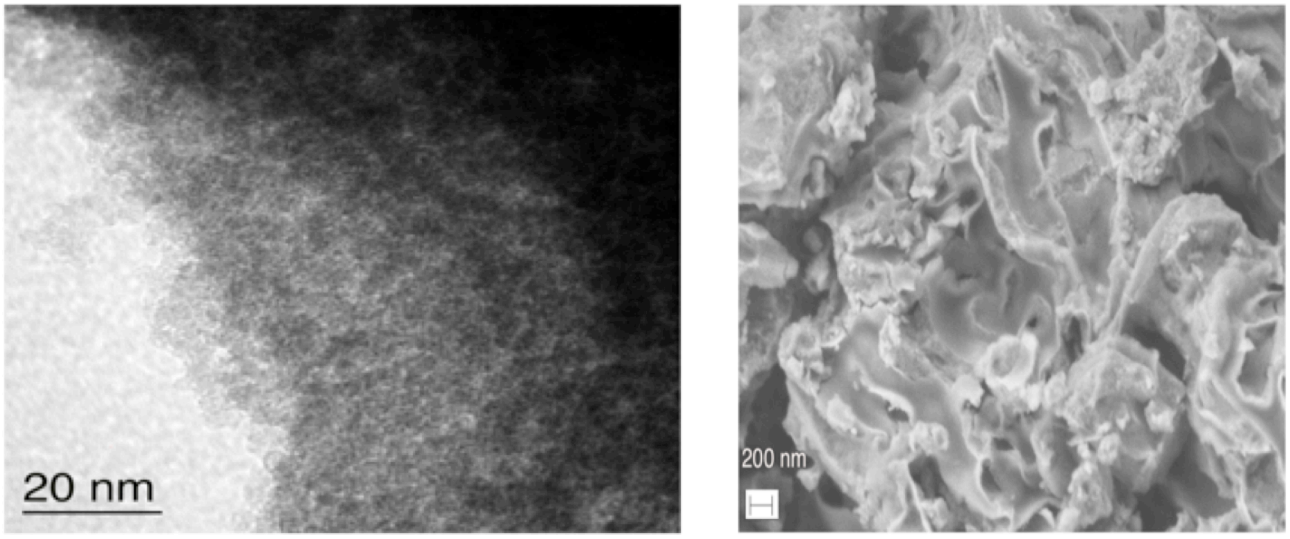

Figure S9. TEM (left) and SEM (right) images of ***meso*-SiC**. The ultrafine sample that was obtained by sonicating ***meso*-SiC** in ethanol was dispersed on a copper grid for measurement, and imaged using a 120-kV electron beam. For SEM, the sample was put on carbon tape and imaged using a secondary-electron detector and a 2-kV electron beam.

3. X-ray diffraction patterns of the supports

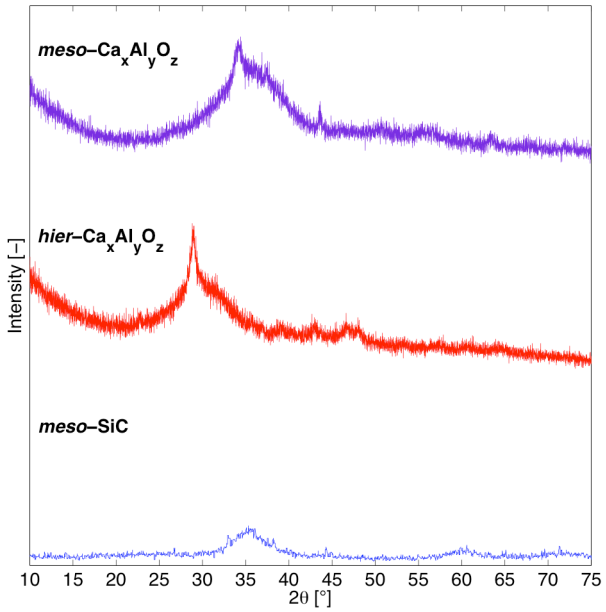

Figure S10. X-ray diffraction patterns of ***meso*-$Ca_xAl_yO_z$**, ***hier*-$Ca_xAl_yO_z$**, and ***meso*-SiC**. Each pattern was normalised and offset for plotting.

4. Determination of maximum carbonation levels for $Ca_xAl_yO_z$ sorbents

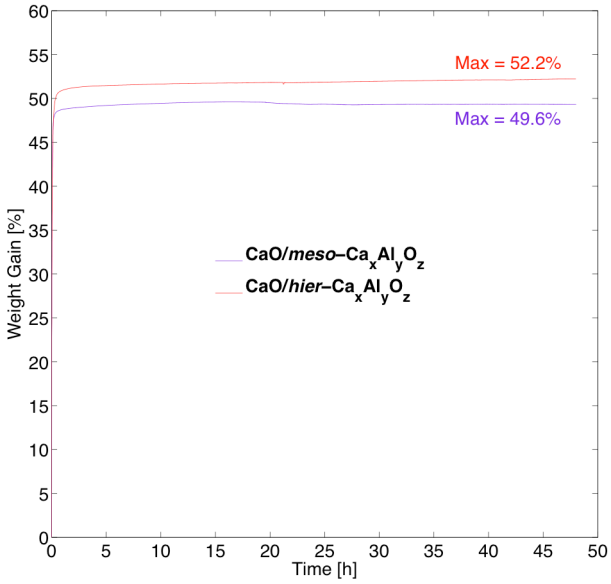

Figure S11. Determination of maximum carbonation levels for calcium aluminate sorbents. Samples were heated to 700 °C in 100% CO_2.

5. Synthesis and characterization of CaO/*meso*-$Ca_xAl_yO_z$(20)

CaO/*meso*-$Ca_xAl_yO_z$(20) was synthesised using a sol-gel synthesis procedure that employed evaporation-induced self-assembly. Calcium aluminate with a 4:1 ratio of Al^{3+} and Ca^{2+} ions was prepared by combining the structure-directing agent $PEO_{20}PPO_{70}PEO_{20}$ (Pluronic P123, Aldrich, 2.0 g, M_n = 5800 g mol^{-1}) and ethanol (absolute, 20.0 mL) and stirring at 40 °C until the surfactant dissolved. $Ca(NO_3)_2$•$4H_2O$ (Fluka, ≥99.0%, 0.95 g) was then added to this solution and stirred vigorously until it dissolved. In a separate solution, aluminium isopropoxide (Sigma Aldrich, 3.27 g) was combined with nitric acid (3.2 mL, 67 wt.%) and ethanol and stirred at 40°C until dissolved. These solutions were then combined and stirred at room temperature for 5 h, then aged at 60 °C for 48 h under flowing N_2. The dried calcium aluminate sol was then calcined by heating in a GSL-1300X tube furnace (MTI) in air at 1.5 °C/min to 700 °C and holding at that temperature for 4 h.

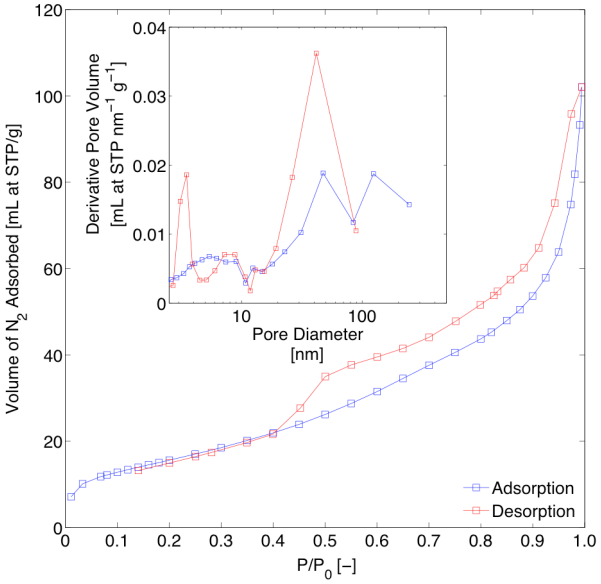

Figure S12. N_2 physisorption isotherm (measured at −196 °C) and pore size distribution (inset) of **CaO/*meso*-$Ca_xAl_yO_z$(20)**.

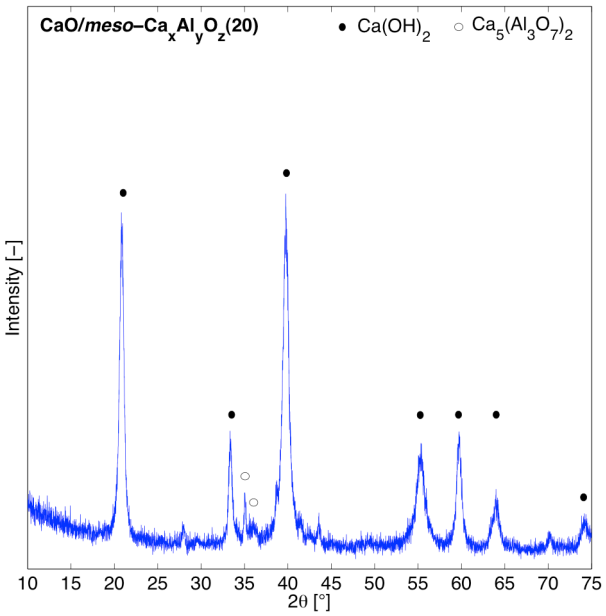

Figure S13. XRD pattern of **CaO/*meso*-$Ca_xAl_yO_z$(20)**.

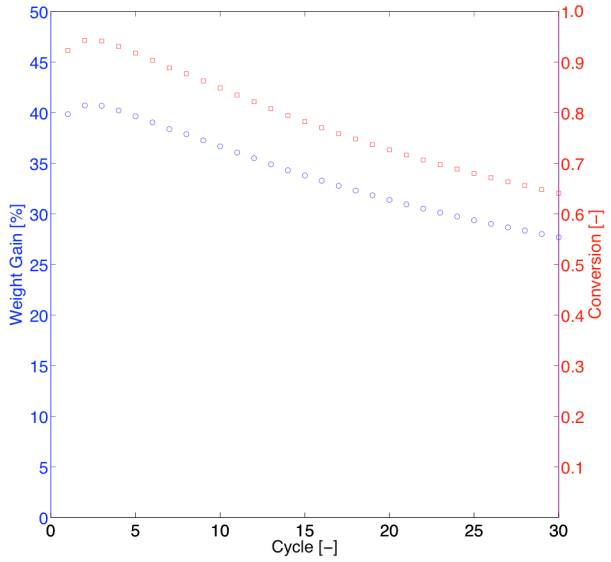

Figure S14. Performance of **CaO/*meso*-$Ca_xAl_yO_z$(20)** in 30 calcination-carbonation cycles (carbonation: 700 °C, 15% CO_2 in N_2, 30 min; calcination: 850 °C, N_2, 10 min).