Synthesis of sintering-resistant sorbents for CO₂ capture

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Figure S1 XRD patterns of solids after first-heating-step (drying) from solution mixture of calcium D-gluconate monohydrate and magnesium D-gluconate hydrate.



Figure S2 The influence of sample mass on the conversion of the solid sorbent CGMG75.



Figure S3 Pore size distribution for sorbent CGMG75.



Figure S4 TGA profiles of CO₂ capture capacity for sorbents synthesized from various precursors.



Figure S5 A schematic description of process of a chemical heat pump.

Principle of a chemical heat pump: Kato et al. (*S1*) proposed a CaO/PbO/CO₂ chemical heat pump, the principle of which is illustrated in Figure S5. The system has CaO and PbO reactors that originally charged with CaCO₃ and PbO. With the operation of this system, a heat that has a higher temperature than the original heat source can be obtained. There are two operation modes in this system:

Storage mode: the CaO reactor received heat from a heat source and CaCO₃ was decarbonated to CaO and CO₂. CO₂ was reacted with PbO in the PbO reactor to form PbCO₃ and the exothermic heat of the carbonation was recovered. It was suggested that over 860 $^{\circ}$ C was the optimal temperature for CaCO₃ decarbonation and 300 $^{\circ}$ C was the optimal for the carbonation.

Heat supply mode: $PbCO_3$ was decomposed by the heat and CO_2 was formed and introduced into the CaO reactor. Then CaO was reacted with the CO_2 to generate heat because of the exothermic reaction. It was suggested that optimal decarbonation temperatures were 440 °C or 450 °C and 880 °C for CaO carbonation.

3D percolation theory: This theory (see the reference for details. Ewen, P. J. S. & Robertson, J. M. A percolation model of conduction in segregated systems of metallic and insulating materials: Application to thick film resistors. *J. Phys. D: Appl. Phys.* **1981**, *14*, 2253-2268.) predicts that minimum weight percentage of support material (MgO) to maintain a continuum framework to separate is 15 vol.%. Therefore the maximum CaO can be calculated to be 85 vol.%. Assuming the particles are spherical, the corresponding weight fraction (82.7 wt.% for CaO and 17.3 wt.% for MgO) then can be calculated by their densities.

Theoretical value of maximum CO₂ **capture capacity** is calculated when assuming 100% CaO conversion. Theoretical value of maximum CO₂ capture capacity= CaO weight friction x (molecular weight of CO₂/molecular weight of CaO). For example when CaO content is 50%, the theoretical value of max CO₂ capture capacity = $0.5 \times (44/56)=0.393$ g-CO2/g-sorbent.

(S1) Kato, Y., Harada, N. & Yoshizawa, Y. Kinetic feasibility of a chemical heat pump for heat utilization of high-temperature processes. *Appl. Therm. Eng.* **1999**, *19*, 239-254.