

## Support Information

# Controllable Synthesis of Two CaO Crystals: Precursors' Template-Free Synthesis and Formation Mechanism

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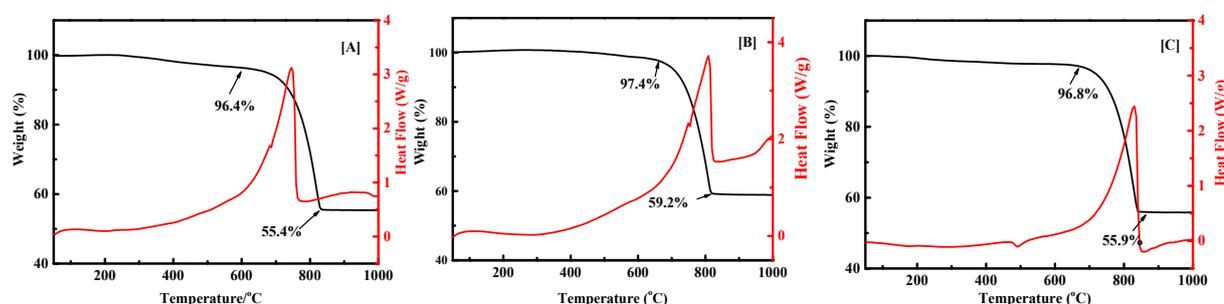


Fig. S1 TGA and DSC plots of dumbbell-like, wheat spike-like and microsphere-like CaCO<sub>3</sub> samples.

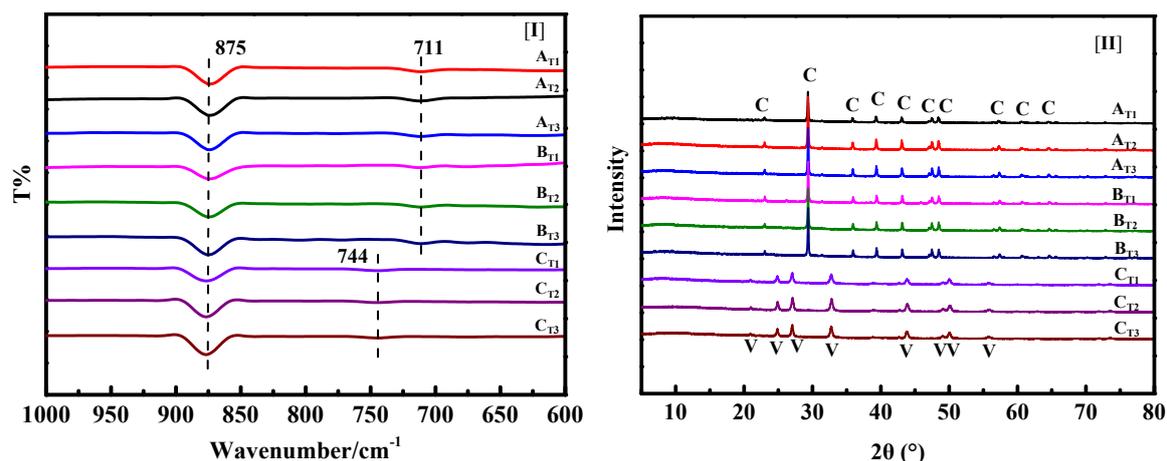


Fig. S2 FTIR spectra and XRD patterns of  $\text{CaCO}_3$  precursors obtained at different reaction temperature with saturated  $\text{Ca}(\text{OH})_2$  solution for 1 h:  $A_{T1}$  80 °C,  $A_{T2}$  120 °C and  $A_{T3}$  150 °C with 1.5 g/L  $\text{CO}_2\text{SM}$  concentration;  $B_{T1}$  80°C,  $B_{T2}$  120 °C and  $B_{T3}$  150 °C with 3.0 g/L  $\text{CO}_2\text{SM}$  concentration;  $C_{T1}$  80 °C,  $C_{T2}$  120 °C and  $C_{T3}$  150 °C with 20.0 g/L  $\text{CO}_2\text{SM}$  concentration. In the second picture, “C” and “V” stand for calcite and vaterite respectively.

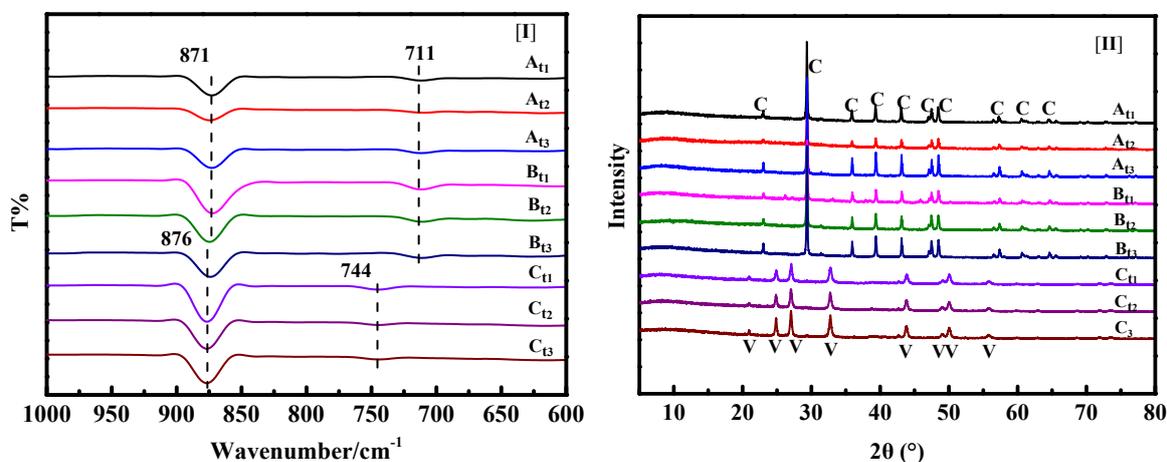


Fig. S3 FTIR spectra (I) and XRD patterns (II) of  $\text{CaCO}_3$  precursors obtained in different reaction time at 100 °C with saturated  $\text{Ca}(\text{OH})_2$  solution:  $A_{t1}$  0.5 h,  $A_{t2}$  2.0 h and  $A_{t3}$  4.0 h with 1.5 g/L  $\text{CO}_2\text{SM}$  concentration;  $B_{t1}$  0.5 h,  $B_{t2}$  2.0 h and  $B_{t3}$  4.0 h with 3.0 g/L  $\text{CO}_2\text{SM}$  concentration;  $C_{t1}$  0.5 h,  $C_{t2}$  2.0 h and  $C_{t3}$  4.0 h with 20.0 g/L  $\text{CO}_2\text{SM}$  concentration. In Figure II, “C” and “V” stand for calcite and vaterite respectively.

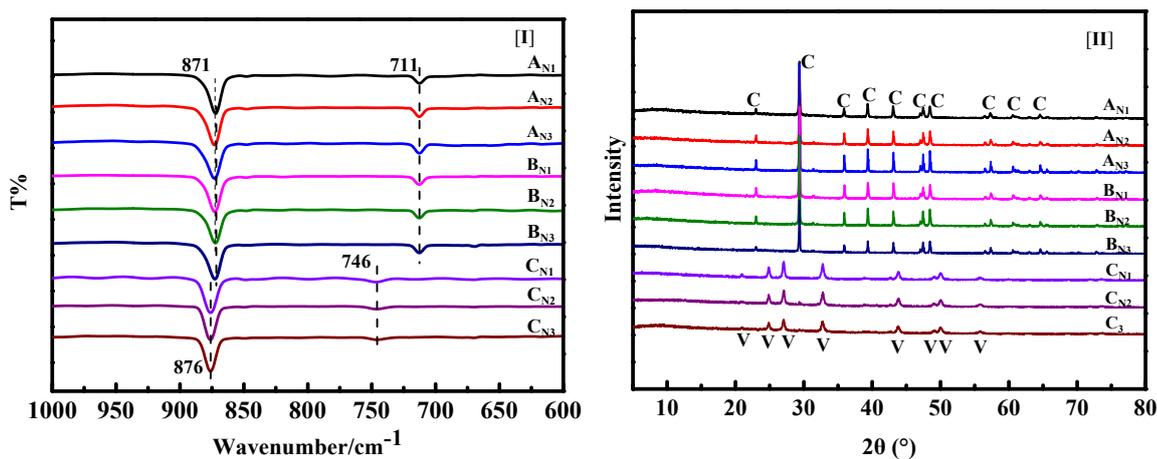


Fig. S4 FTIR spectra (I) and XRD patterns (II) of  $\text{CaCO}_3$  precursors obtained in different  $\text{Ca}(\text{OH})_2$  concentrations at  $100\text{ }^\circ\text{C}$  for 1 h:  $\text{A}_{\text{N}1}$  1.44 g/L,  $\text{A}_{\text{N}2}$  1.28 g/L and  $\text{A}_{\text{N}3}$  1.12 g/L with 1.5 g/L  $\text{CO}_2\text{SM}$ ;  $\text{B}_{\text{N}1}$  1.44 g/L,  $\text{B}_{\text{N}2}$  1.28 g/L and  $\text{B}_{\text{N}3}$  1.12 g/L with 3.0 g/L  $\text{CO}_2\text{SM}$ ;  $\text{C}_{\text{N}1}$  1.44 g/L,  $\text{C}_{\text{N}2}$  1.28 g/L and  $\text{C}_{\text{N}3}$  1.12 g/L with 3.0 g/L  $\text{CO}_2\text{SM}$ . In Figure II, “C” and “V” stand for calcite and vaterite respectively.

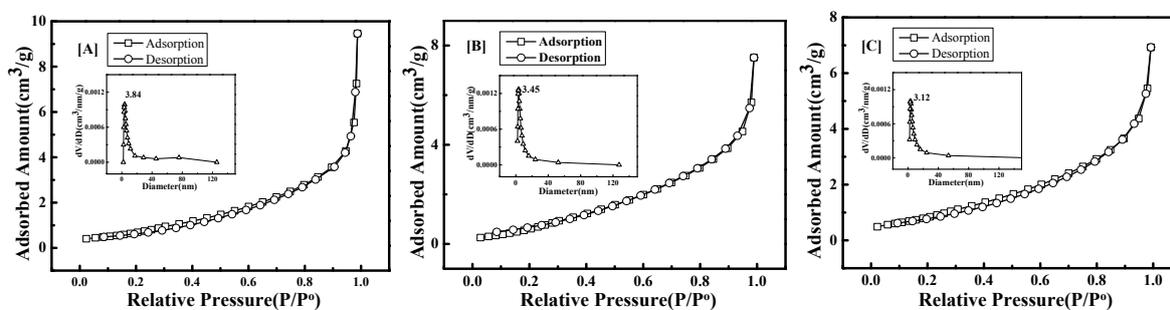


Fig. S5 The adsorption-desorption isotherms and the corresponding pore diameter distribution curves of  $\text{CaO}$  by calcinating A dumbbell-like, B wheat spike-like and C microsphere-like  $\text{CaCO}_3$  precursors, respectively. Typical BET isotherms and pore diameter distribution curves for various samples were plotted.

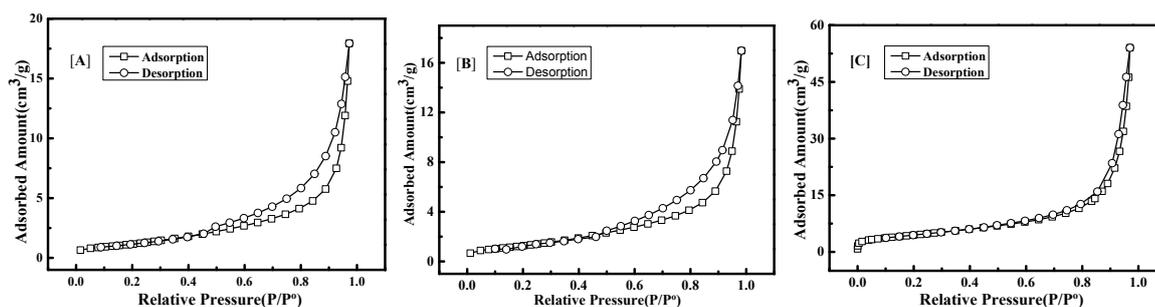


Fig. S6 The  $N_2$  adsorption-desorption isotherms of A dumbbell-like, B wheat spike-like and C microsphere-like  $CaCO_3$  precursors. The values of specific surface area could be obtained were 4.7, 4.9 and  $16.4 \text{ m}^2/\text{g}$  respectively for dumbbell-like, wheat spike-like and microsphere-like  $CaCO_3$  precursors respectively.

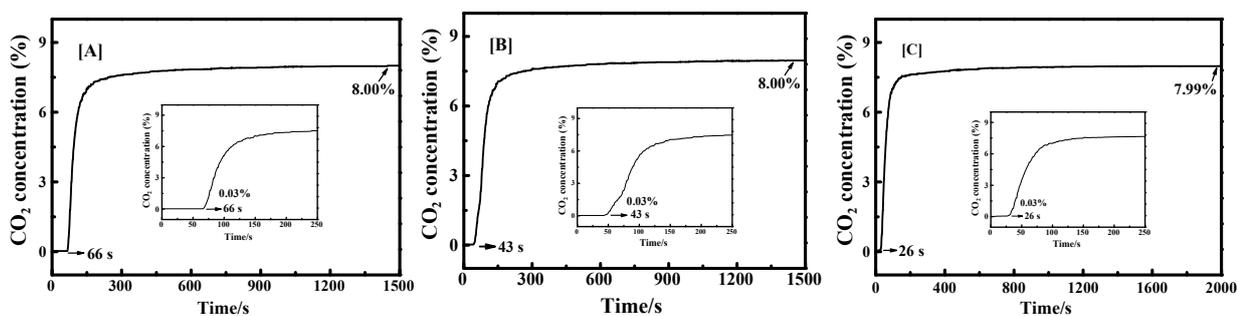


Fig. S7 Breakthrough curves of  $CaO$  crystals: by calcinating A dumbbell-like, B wheat spike-like and C microsphere-like  $CaCO_3$  precursors respectively with 8 %  $CO_2$  under the atmospheric pressure and the gas velocity was  $30 \text{ mL}/\text{min}$ . In addition, the local enlargement of the breakthrough curve is placed in the diagram.