

## Supporting information

### *Yolk-shell-type CaO-based sorbents for CO<sub>2</sub> capture: Assessing the role of nanostructuring for the stabilization of the cyclic CO<sub>2</sub> uptake*

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Table S1: Phases and their respective space groups used in Rietveld refinement

#### Experimental

##### *1.1 Rietveld refinement*

Multiphase Rietveld refinements were carried out with the FullProf Suite software. The space groups of the individual phases that were used for the Rietveld refinements are listed in Table S1. Exemplary fits for Ca-22Zr and Ca@55Zr are shown in Figure S2 a and b, respectively.

Table S1: Space groups of the individual phases that were used for the Rietveld refinements.

CaO	CaZrO <sub>3</sub>	Ca(OH) <sub>2</sub>	CaCO <sub>3</sub>	t-ZrO <sub>2</sub>
F m -3 m	P c m n	P -3 m 1	R -3 c	P 42/n m c

## Figures

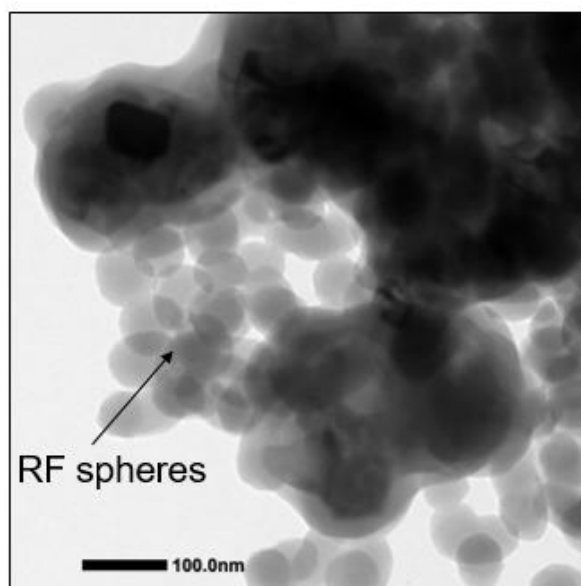


Figure S1: Resorcinol-formaldehyde polymer spheres observed for thicker template depositions at higher temperatures (here: 5 h at 35 °C).

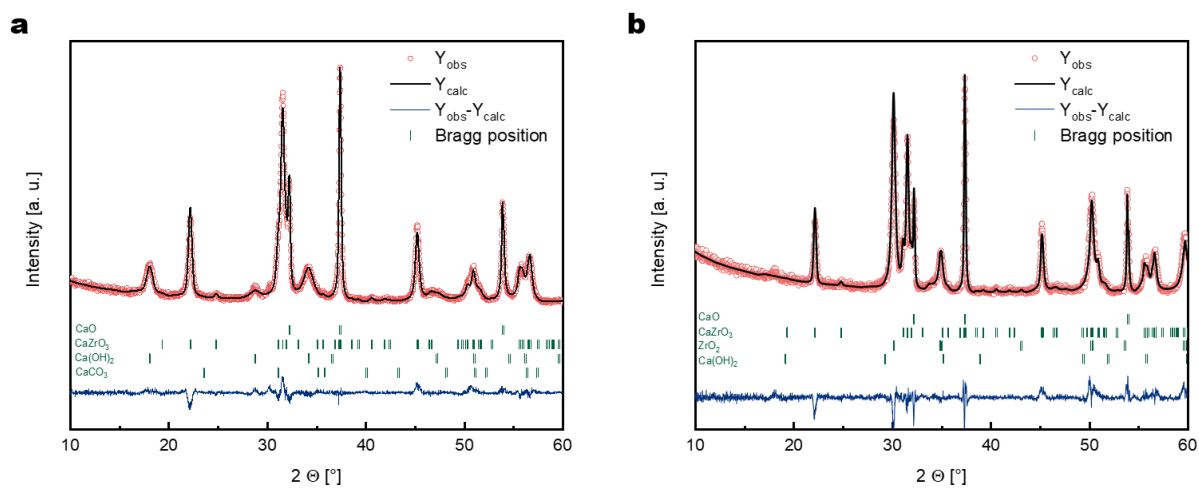


Figure S2: Exemplary Rietveld refinements for as-prepared sorbents: a) Ca-26Zr (core shell) and b) Ca@55Zr (yolk-shell).

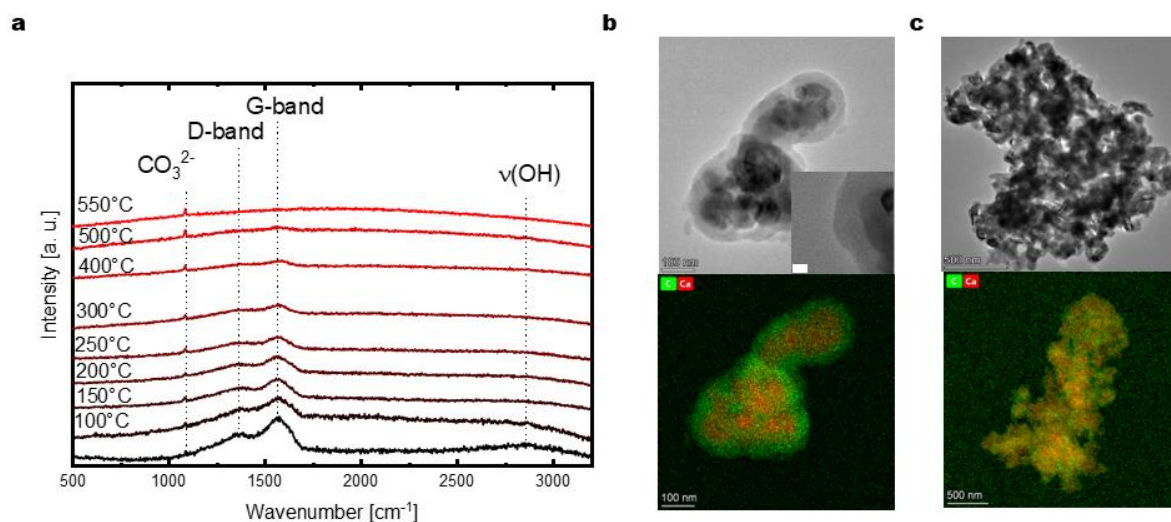


Figure S3: a) In-situ Raman spectroscopy measurements of Ca@C in air (heating ramp  $20\text{ }^{\circ}\text{C min}^{-1}$ ). The vibrational modes of OH disappear at approximately  $100\text{ }^{\circ}\text{C}$ . The D-band and G-band of carbon fully disappear at  $550\text{ }^{\circ}\text{C}$ , confirming complete carbon removal at this temperature; TEM micrograph and EDX-TEM map for Ca and C b) before removal of the carbonaceous template and c) after removal of the carbonaceous template in air at  $900\text{ }^{\circ}\text{C}$  (1 h).

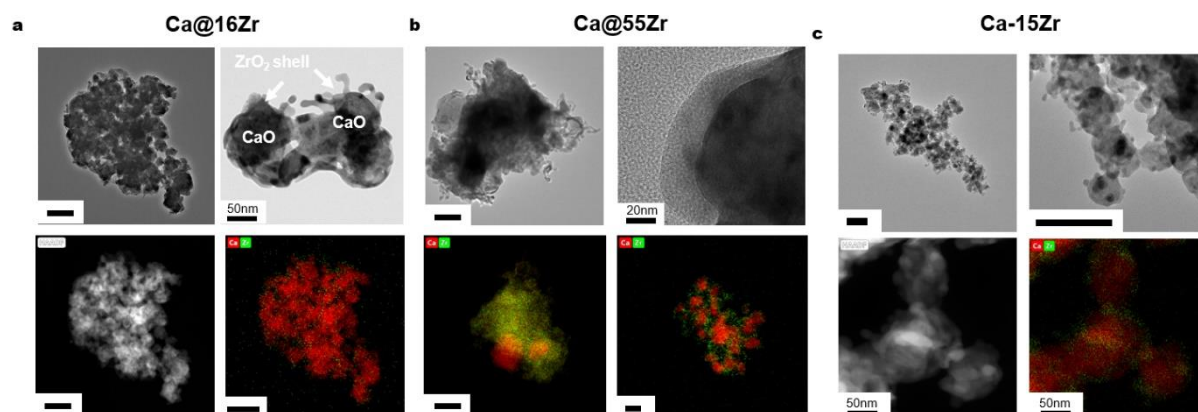


Figure S4: TEM micrographs and EDX-TEM elemental maps for Ca and Zr for zirconia-stabilized sorbents a) yolk-shell-type Ca@16Zr, b) yolk-shell-type Ca@55Zr and c) core-shell-type Ca-15Zr. The scale bars refer to 200 nm if not mentioned otherwise.

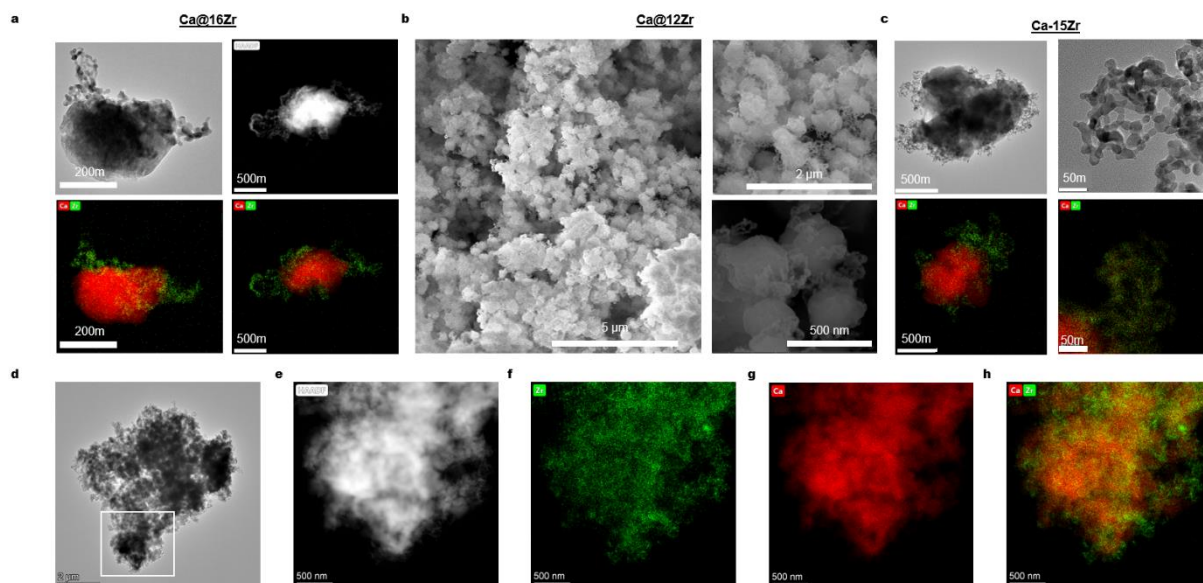


Figure S5: a) TEM micrographs and EDX-TEM elemental mapping of Ca and Zr for Ca@16Zr after 10 cycles, b) SEM micrographs of Ca@12Zr after 10 cycles showing that individual CaO nanoparticles are preserved, c) TEM micrographs and EDX-TEM elemental mapping of Ca and Zr for Ca-15Zr, d) TEM micrograph of Ca@12Zr after 20 cycles with e) – h) HAADF image and elemental mappings for Ca and Zr from EDX-TEM of the inset indicated in d).