Supplementary Materials

Supplement to 3.1. Catalyst performance

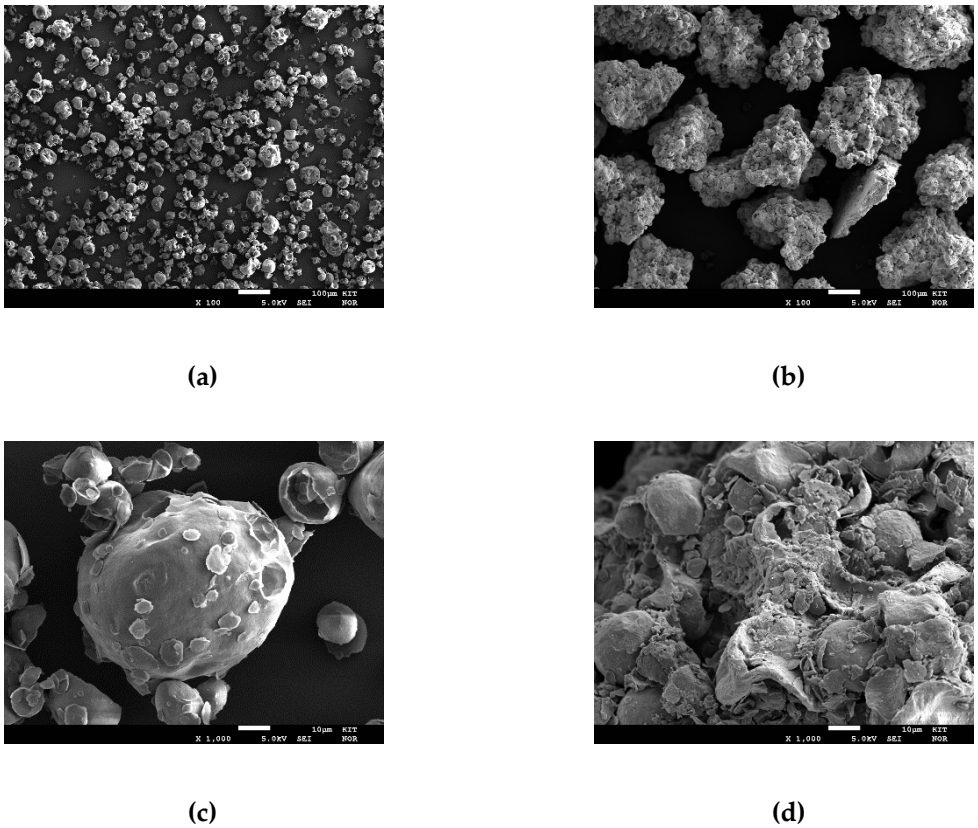
WGS rate equation acc. to Choi et al. with reaction rate r_{CO} in mol/(g·h), temperature T in K, pressure p in atm, and equilibrium constant K_{eq} calculated from Eq. (2).

$$r_{CO} = 2.96 \cdot 10^5 \cdot \exp\left(-\frac{47400}{R \cdot T}\right) \cdot \left(p_{H_2O} \cdot p_{CO} - \frac{p_{CO_2} \cdot p_{H_2}}{K_{eq}}\right) \quad \text{(S1)}$$

Supplement to 3.2. Structural properties and CO_2 capture performance of sorbent

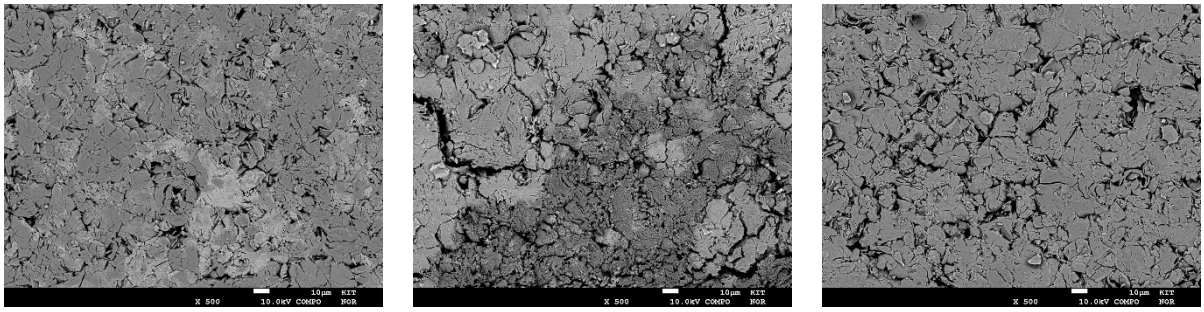
Table S1. BET surface area, BJH average pore width and cumulative pore volume.

Sample	surface area / m²/g	pore width / nm	pore volume / cm³/g
MG70-400	170	3.95	0.123
MG70-K-400	9	12.5	0.027
MG70-400-K-400	18	9.50	0.042



(a) **(b)** **(c)** **(d)**

Figure S1. SE image of (a) untreated sample (MG70) (x100), (b) impregnated – calcined sample (MG70-K-400) (x100), (c) untreated sample (MG70) (x1000), and (d) impregnated – calcined sample (MG70-K-400) (x1000).

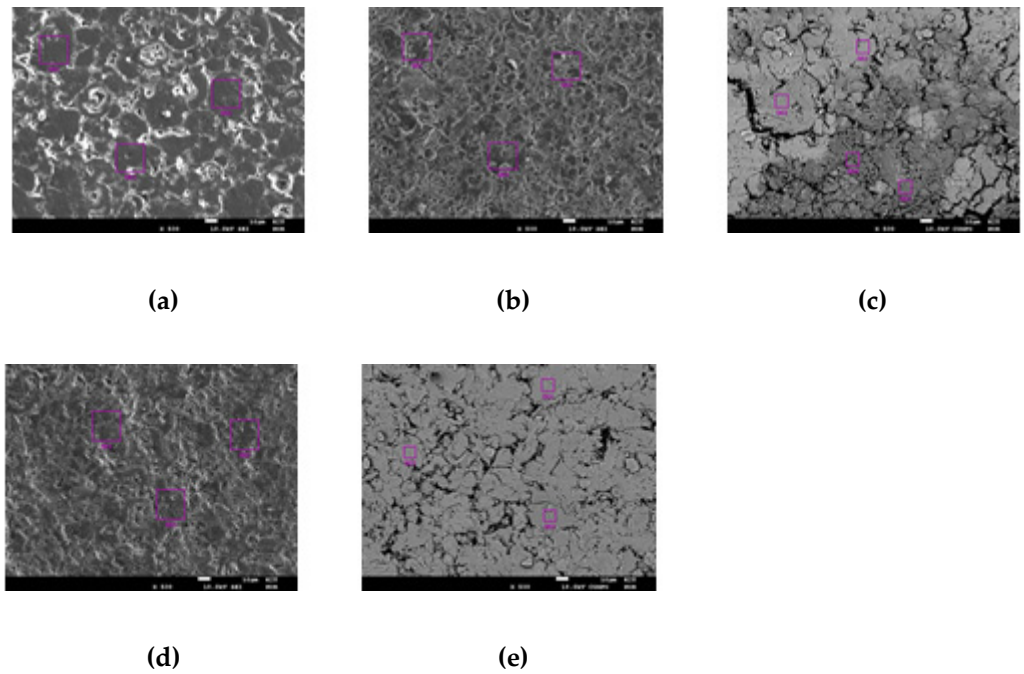


(a) **(b)** **(c)**

Figure S2. BSE image (x500) of pressed powder (a) calcined – impregnated sample (MG70-400-K), (b) impregnated – calcined sample (MG70-K-400), and (c) calcined – impregnated – calcined sample (MG70-400-K-400).

Table S2. EDS results: Mg/Al ratio (average) and potassium content (probe positions in Figure S3).

Sample	Mg/Al / -	K 1 / wt%	K 2 / wt%	K 3 / wt%	K 4 / wt%
MG70	3.11 ± 0.03	-	-	-	
MG70-400	3.18 ± 0.02	-	-	-	
MG70-K-400	3.64 ± 0.54	6.55	2.95	23.97	23.14
MG70-400-K	3.19 ± 0.06	12.85	6.52	26.53	
MG70-400-K-400	3.12 ± 0.02	6.89	3.19	6.55	



(a) **(b)** **(c)**

(d) **(e)**

Figure S3. Probe positions marked for EDS analysis of pressed powder (a) untreated sample (MG70), (b) calcined sample (MG70-400), (c) impregnated – calcined sample (MG70-K-400), (d) calcined – impregnated sample (MG70-400-K), and (e) calcined – impregnated – calcined sample (MG70-400-K-400). Results are listed in Table S2.

Table S3. Freundlich equation (Eq. (S2)) parameters k_F and n, and fitting parameter R^2.

Sample	k_F / mmol/(g·bar$^{1/n}$)	n / -	R^2 / -
MG70-250	0.008	4.687	0.966
MG70-400	0.071	5.280	0.984
MG70-500	0.022	4.399	0.985
MG70-K-400	0.174	9.512	0.97
MG70-400-K-400	0.141	8.924	0.967

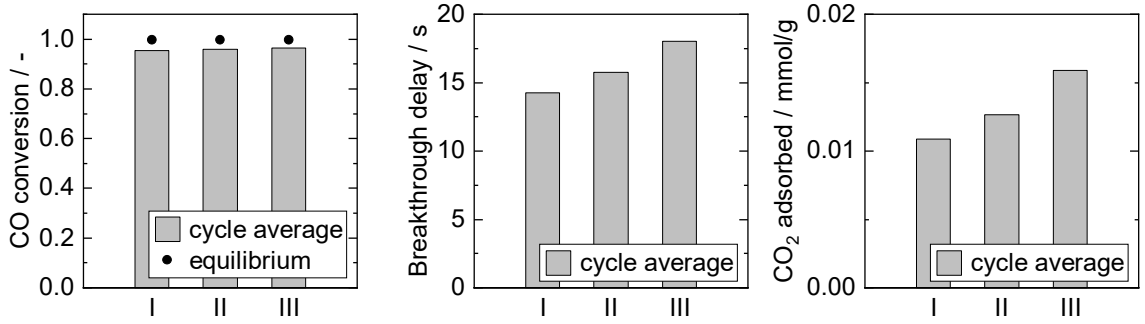
Freundlich model equation for CO_2 adsorption isotherm with n_{ads} being the specific amount of moles CO_2 adsorbed in mmol/g, Freundlich constant k_F in mmol/(g·bar$^{1/n}$), partial pressure of CO_2 p_{CO2} in bar, and adsorption intensity n.

$$n_{ads} = k_F \cdot p_{CO_2}^{1/n} \quad \text{(S2)}$$

Supplement to 3.3 SEWGS performance

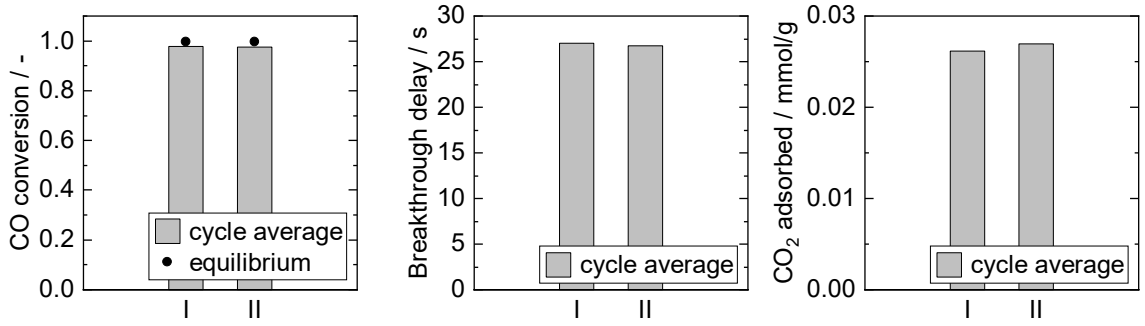
Supplement to 3.3.2 Long term stability

The sorbent in Figure S4 has seen a higher water content (in other experiments at different conditions) between the reproduced experiments (II and III), what resumed in an increase in both, breakthrough time and sorption capacity. This might be due to adsorption site activation caused by steam, see section 3.3.3 and 3.3.4. However, the sorbent in the example depicted in Figure S5 had a comparable time on stream but was not exposed to different conditions between the repetitions, and no noteworthy difference in breakthrough delay or in adsorbed CO_2 could be detected.



(**a**) (**b**) (**c**)

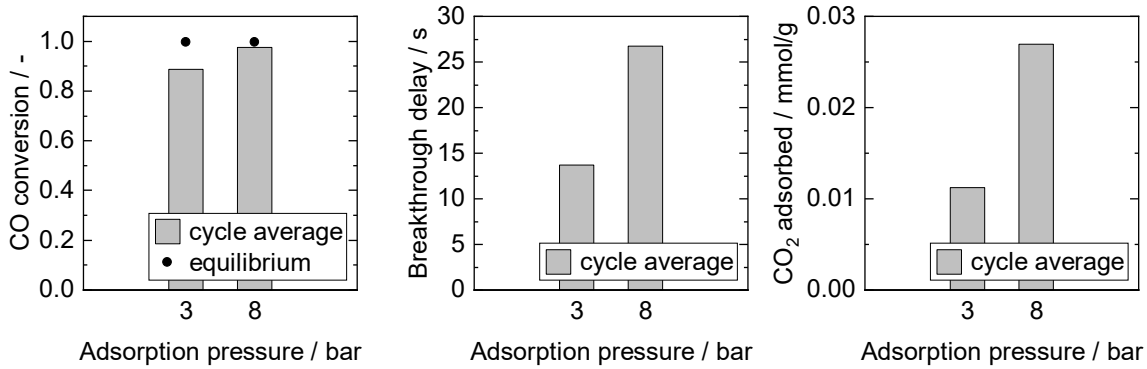
Figure S4. Adsorption cycle average (a) CO conversion, (b) breakthrough delay, and (c) adsorbed amount of CO_2 for reproduced experiments after different sorbent "histories". Adsorption: 15 min, 8 bar, 2000 ml/min, 2.5 % CO, S/G = 4. Desorption: 40 min, 1 bar, 1000 ml/min, 40 % H_2O. PBMR, Mode B, 250 °C.



(**a**) (**b**) (**c**)

Figure S5. Adsorption cycle average (a) CO conversion, (b) breakthrough delay, and (c) adsorbed amount of CO_2 for reproduced experiments after comparable sorbent "histories". Adsorption: 15 min, 8 bar, 2000 ml/min, 2.5 % CO, S/G = 4. Desorption: 40 min, 1 bar, 1000 ml/min, 40 % H_2O. PBMR, Mode C, 250 °C.

Supplement to 3.3.3 Variation of adsorption parameters



(**a**) (**b**) (**c**)

Figure S6. Adsorption cycle average (a) CO conversion, (b) breakthrough delay, and (c) adsorbed amount of CO_2. Adsorption: 15 min, 2000 ml/min, 2.5 % CO, S/G = 4. Desorption: 40 min, 1 bar, 1000 ml/min, 40 % H_2O. PBMR, Mode C, 250 °C.

Supplement to 3.3.5 Variation of reactor configuration

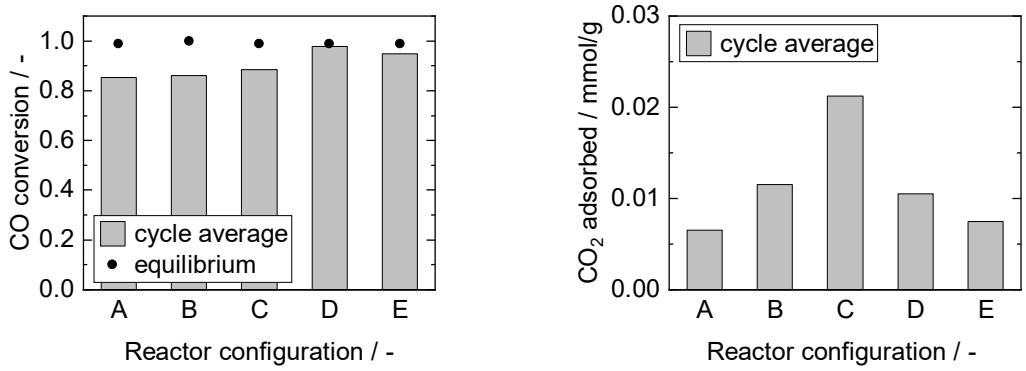


Figure S7. Adsorption cycle average (a) CO conversion, and (b) adsorbed amount of CO_2. Adsorption: 15 min, 8 bar, 2000 ml/min, 10 % CO, S/G = 2. Desorption: 40 min, 1 bar, 1000 ml/min, 40 % H_2O. PBMR, 250 °C.