Supplementary Materials

Effect of alkali metal (Li, Na, K,) on Ni/CaO dual functional materials for integrated CO_2 capture and hydrogenation

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Table S1. Weak, medium, strong and total basicity of the reduced Ni/CaO and M-Ni/CaO (M = Li, Na, K) DFMs.

Materials	Weak basicity (μmol/g)	Medium basicity (μmol/g)	Strong basicity (μmol/g)	Total basicity (μmol/g)
Ni/CaO	55.7	462.1	248.9	766.7
Li-Ni/CaO	120.5	321.0	131.2	572.7
Na-Ni/CaO	135.1	547.1	304.9	987.1
K-Ni/CaO	102.8	493.5	283.2	879.5

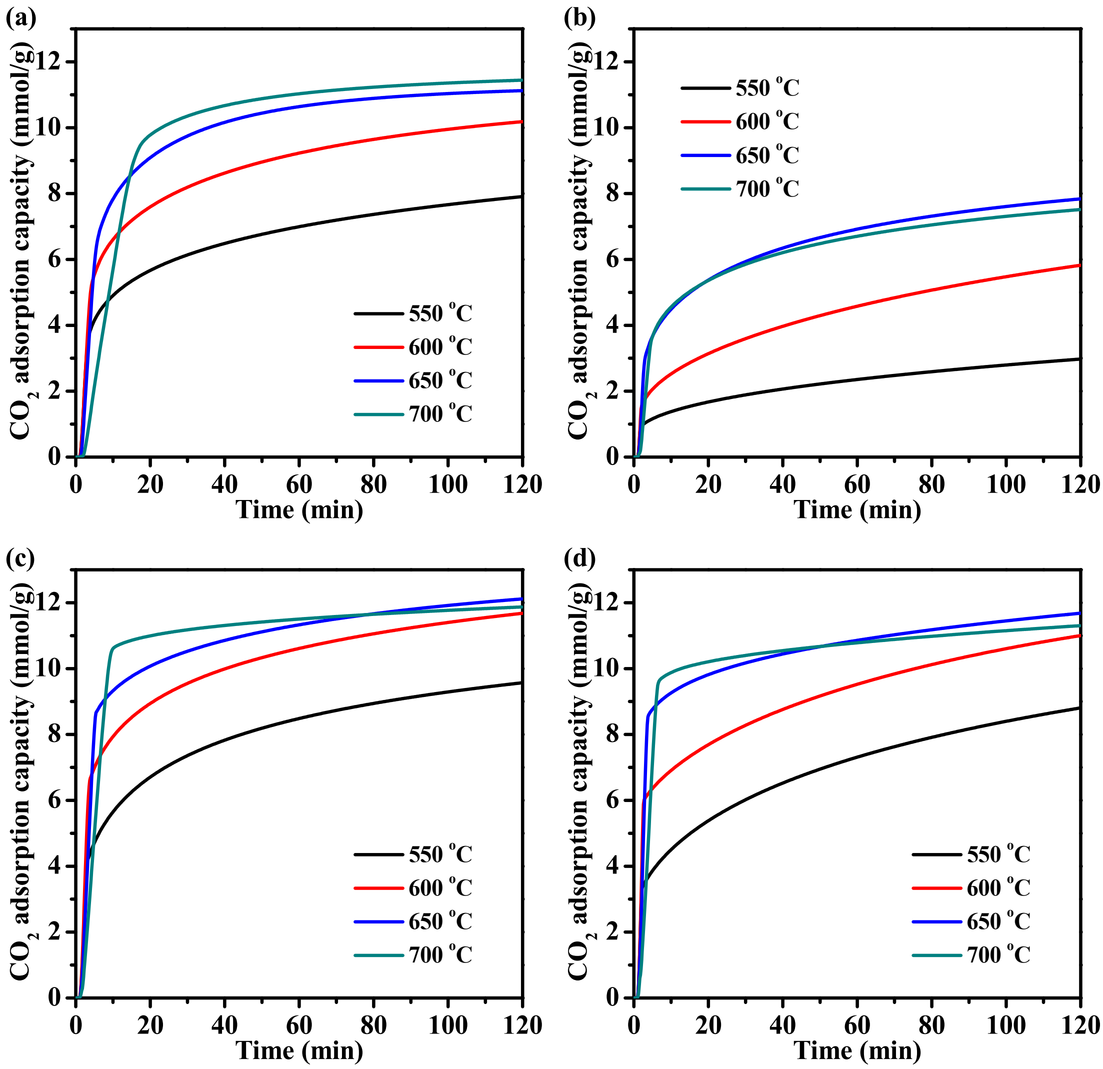

Figure S1. CO_2 capture profiles in the temperature range of 550 to 700 °C for 2 h. (a) Ni/CaO, (b) Li-Ni/CaO, (c) Na-Ni/CaO and (d) K-Ni/CaO.

Table S2. The CO_2 capture capacities of the M-Ni/CaO (M = Li, Na, K) DFMs at the temperature range of 550 – 700 °C for 2 h.

Materials	CO_2 adsorption capacity (mmol/g)				
	Theoretical value	550 °C	600 °C	650 °C	700 °C
Ni/CaO	17.4	7.9	10.2	11.1	11.4
Li-Ni/CaO	16.5	3.0	5.8	7.8	7.5
Na-Ni/CaO	16.5	9.6	11.7	12.1	11.9
K-Ni/CaO	16.5	8.8	11.0	11.7	11.3

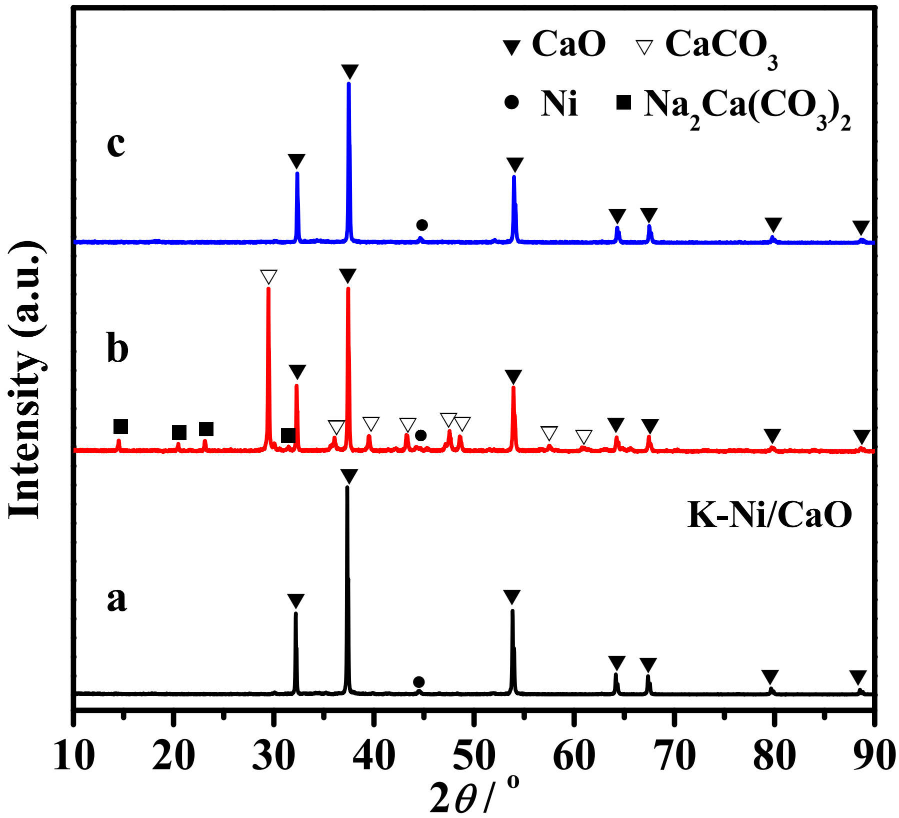

Figure S2. XRD pattern of the Na-Ni/CaO with 15 wt% Na doping after (a) the reduction; (b) the CO_2 capture stage and (c) the integrated CO_2 capture and hydrogenation.

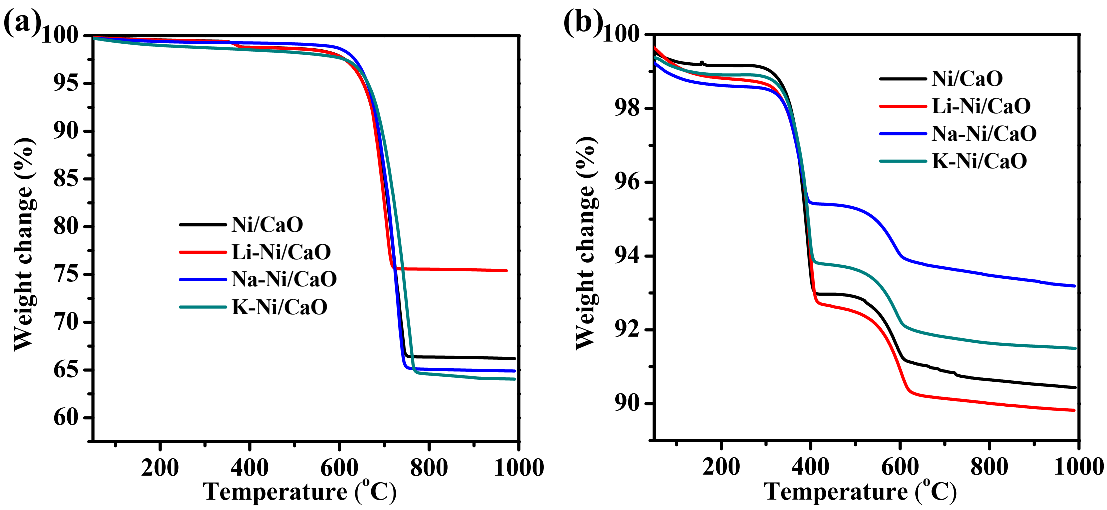

Figure S3. TG profiles of all DFMs (a) after the CO_2 capture stage and (b) the hydrogenation stage.

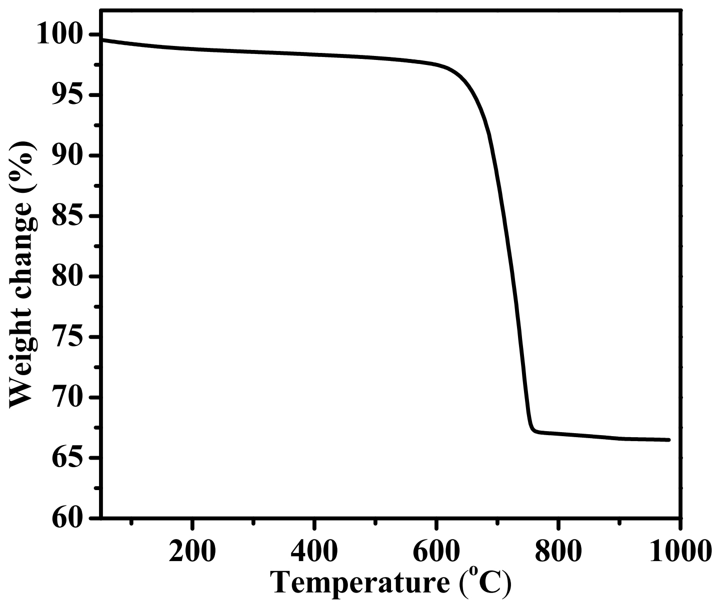

Figure S4. TG profile of the spent Na-Ni/CaO after adsorption in CO_2/Ar for 1 h.

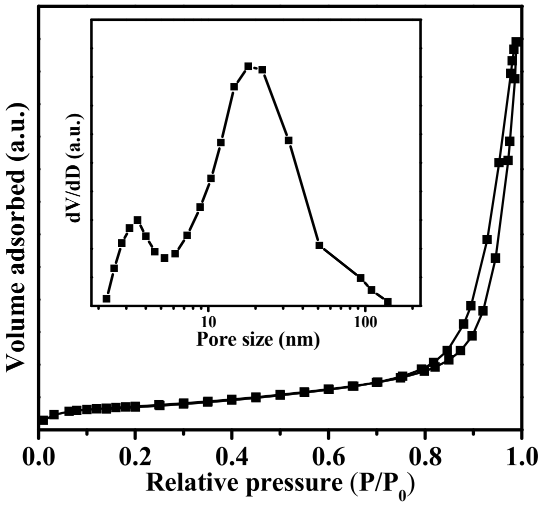

Figure S5. N_2 adsorption-desorption isotherms and pore size distribution (insert part) of spent Na-Ni/CaO DFM after 20 cycles of integrated CO_2 capture and hydrogenation.

Table S3. The combined CO_2 capture and hydrogenation performance over our proposed Na-Ni/CaO DFM and recently documented Ni/CaO based DFMs.

DFMs	Preparation method	Reaction	Temperature (ºC)	Feed composition CO_2 capture + conversion	CO_2 capacity (mmol/g)	Product yield (mmol/g)	Ref.
$Fe_5Co_5Mg_{10}CaO$	One-pot sol-gel	RWGS	650	10% CO_2 + 100% H_2	9.2	8.28	1
15NiCa 15NiNa	Wet impregnation	Methanation	520	10% CO_2 + 10% H_2	- -	0.14 0.19	2
$Ca_1Ni_{0.1}$ $Ca_1Ni_{0.1}Ce_{0.033}$	Sol-gel	RWGS	650	15% CO_2 + 5% H_2	15.0 14.1	6.9 7.3	3
Ni/CaO		Methanation	500	10% CO_2/10% H_2O + 10% H_2	8.96	8.34	4
Ni-CaO/γ-Al_2O_3	impregation	Methanation	320	9.5% CO_2 + 10% H_2	0.31	0.14	5
1%NiCaO 1%Ni/CeO_2-CaO-phy	One-pot physical mixing	Methanation	550	15% CO_2 + 100%	9.2 15.3	2.0 8.0	6
Ni/CS-P30-C Ni/CS-P30-C-P	Sol-gel	RWGS	650	10% CO_2 + 5% H_2	13.86 8.95	5.52 5.33	7
Ni/CeO_2-CaO	physical mixture	RWGS	650	10% CO_2 + 5% H_2	9.6	2.7	8
Ni/CaO Na-Ni/CaO	Precipitation-combustion	RWGS	650	10% CO_2 + 10% H_2	11.1 12.0	6.3 7.0	This work

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