

Supplementary Information

Continuous direct air capture and conversion tandem system applicable to a wide range of CO₂ concentrations

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Supplementary Figures and Texts

1 Introduction

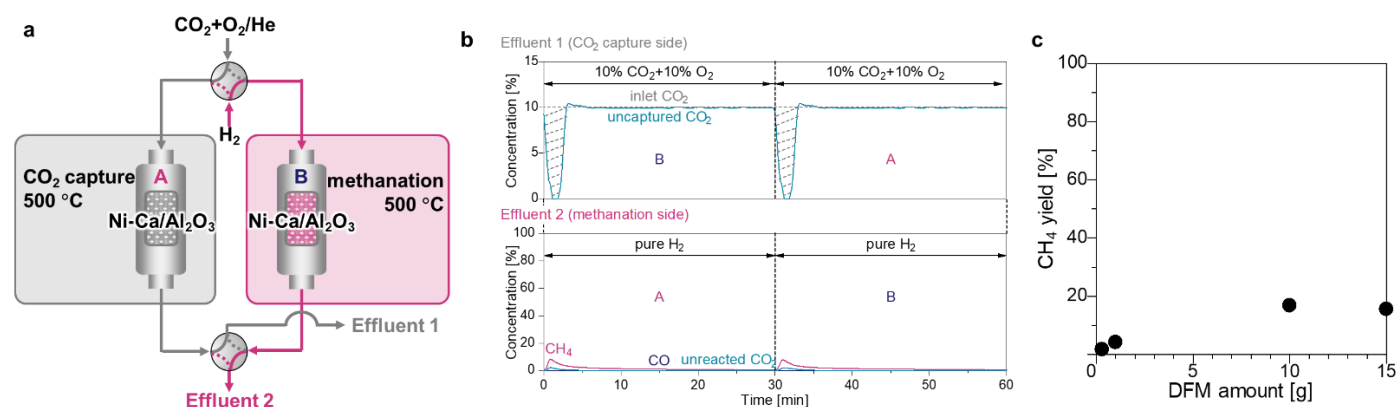


Fig. S1 a. Schematic diagram of the continuous CO₂ capture and methanation system; captured gas (100 mL/min, 10%CO₂+10%O₂/He for 30 min) and hydrogenation gas (100 mL/min, pure H₂ for 30 min) were alternately fed into each reactor containing 10 g of Ni-Ca/Al₂O₃. **b.** Typical time course of the CH₄, CO₂, and CO concentration in effluent 1 and 2, respectively. **c.** Variation in CH₄ yield with DFM amount in continuous CO₂ capture and methanation.

Text S1

Previously, various DFMs for selective CH₄ production were reported. Especially, Ni-Ca based DFMs were well-known as high performance one (Table S1 and S2). Recently, our group developed Ni-Ca/Al₂O₃ DFM, which optimized Ni and Ca loading. Detail characterization revealed that 500 °C is the best condition for CO₂ capture and methanation. For continuous CH₄ production from high concentration of CO₂, we carried out CO₂ capture and methanation using the continuous CCR system (Fig. S1a). First, the CO₂/O₂ mixture was fed into one reactor for 30 min for CO₂ capture. On the other hand, pure H₂ was fed into the other reactor (containing a CO₂-captured Ni-Ca/Al₂O₃). Fig. S1b shows the typical time course of the CH₄, and CO₂ concentrations analyzed by online gas-cell IR in effluent 1 and 2, respectively. Fig. S1c shows the result of the optimization of Ni-Ca/Al₂O₃ amount. These results indicate that increasing the amount of DFM has limited effects on improving CH₄ yield (<20%).

Text S2

The conventional stepwise CCR system illustrated in Fig. 1a was originally developed by Farrauto's group, who pioneered the concept[†]. They successfully demonstrated the capture of CO₂ from emission sources and its subsequent conversion to CH₄ within a single reactor at the same temperature (320 °C), using Ru and nano-dispersed CaO co-supported on porous γ -Al₂O₃. Around the same period, Urakawa's group demonstrated CO₂ capture and subsequent hydrogenation to CO using a DFM composed of earth-abundant elements (FeCrCu/K/MgO–Al₂O₃)[‡]. Furthermore, they proposed a continuous operation concept, illustrated in Fig. 1b, in which CO₂ can be continuously removed from the effluent stream and converted into valuable products (such as syngas) by synchronizing the switching of gas flow directions between two reactors with an appropriate time delay. Following these pioneering studies, our group demonstrated continuous CCR to CO using a dual-functional material composed of Pt nanoparticles coordinated with Na oxides on Al₂O₃, integrated within a reactor configuration similar to that shown in Fig. 1b[¶]. Our previous study demonstrated a groundbreaking CCR process that promotes CO₂ capture and reduction at significantly lower temperatures compared to conventional systems. However, it had a limitation in effectively capturing high concentrations of CO₂, such as those found in simulated exhaust gas. To address this issue, we developed a new process configuration, as shown in Fig. 1c, by integrating a TSA system capable of capturing large amounts of CO₂.

[†] M. Duyar, M. Treviño, R. Farrauto, *Appl. Catal. B*, 2015, 168-169, 370-376

[‡] L. Bobadilla, J. Riesco-García, G. Penelás-Pérez, A. Urakawa, *J. CO₂ Util.*, 2016, 14, 106-111

[¶] S. Miyazaki, L. Li, S. Yasumura, K. Ting, T. Toyao, Z. Maeno, K. Shimizu, *ACS Catal.*, 2022, 12, 2639–2650

2 Results and Discussion

2.1 Screening and optimization of sorbents and catalysts

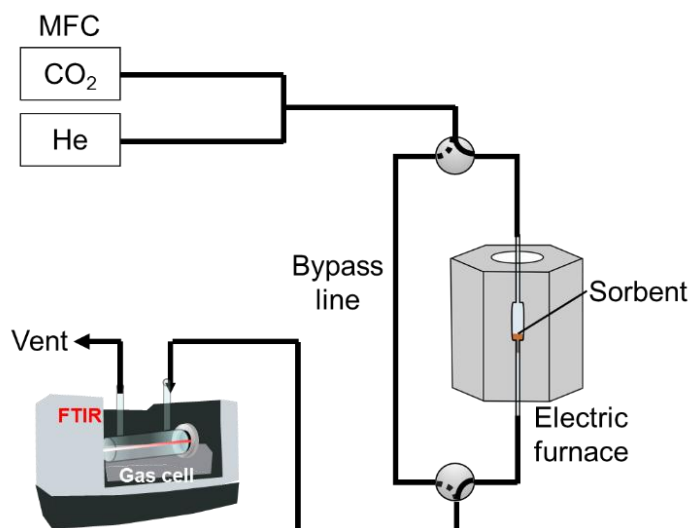


Fig. S2 Illustration of the experimental setup for CO₂ adsorption measurement.

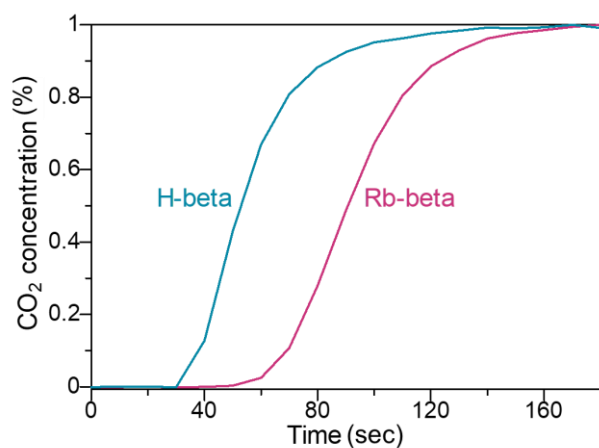


Fig. S3 Profile of the breakthrough experiments over H-beta and Rb-beta under a flow of 1% CO₂ at 50 °C, total flow rate = 100 mL/min, He balance. Weight of adsorbents: 100 mg. The feed gases were introduced to the adsorbents from 0 s.

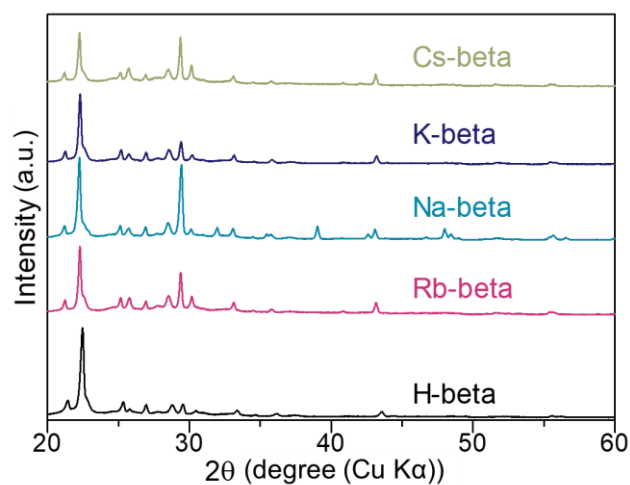


Fig. S4 XRD patterns of H-beta, Rb-beta, Na-beta, K-beta, and Cs-beta.

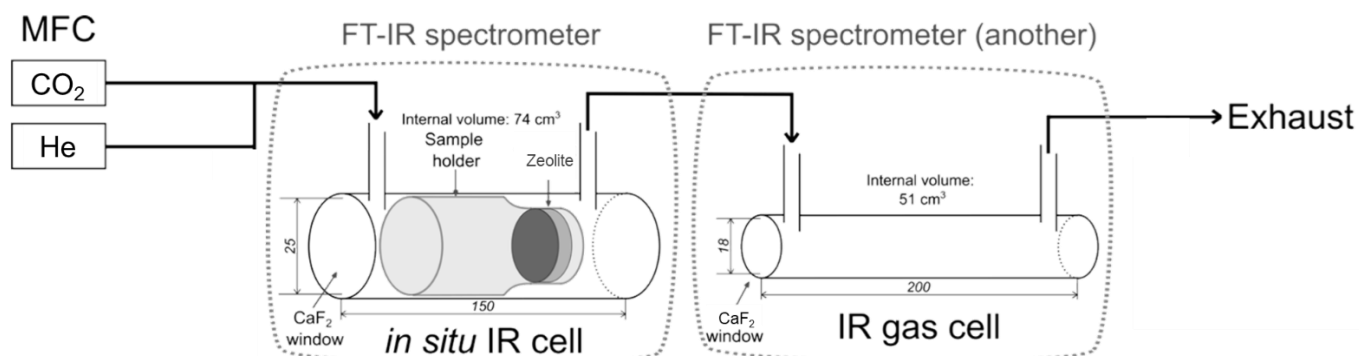


Fig. S5 Schematic view of the setup used for *operando* IR measurement, including *in situ* IR cell and IR gas cell. The inner diameter and length of the cell are provided with a unit of mm.

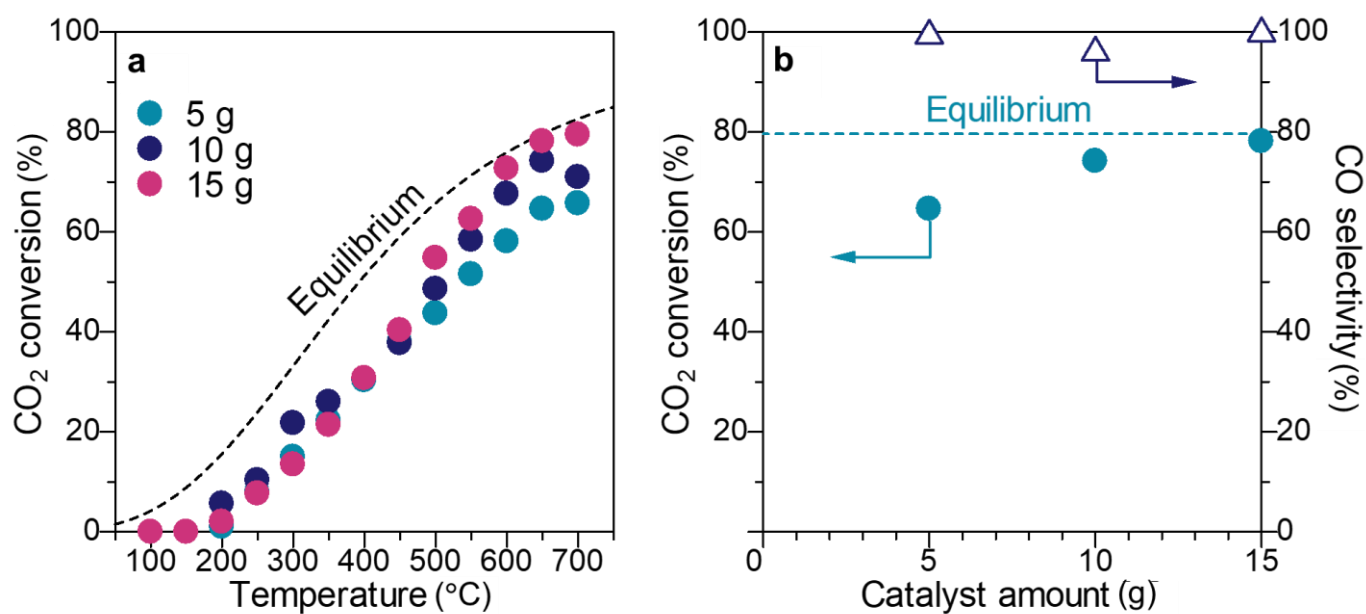


Fig. S6 a. Validation in CO₂ conversion with respect to temperature for different amounts of Cu/ZnO/Al₂O₃. **b.** Variation in CO₂ conversion and CO selectivity with catalyst amount in steady-state RWGS reaction over Cu/ZnO/Al₂O₃.

2.2 Continuous production of O₂-free CO₂ from CO₂/O₂ mixture

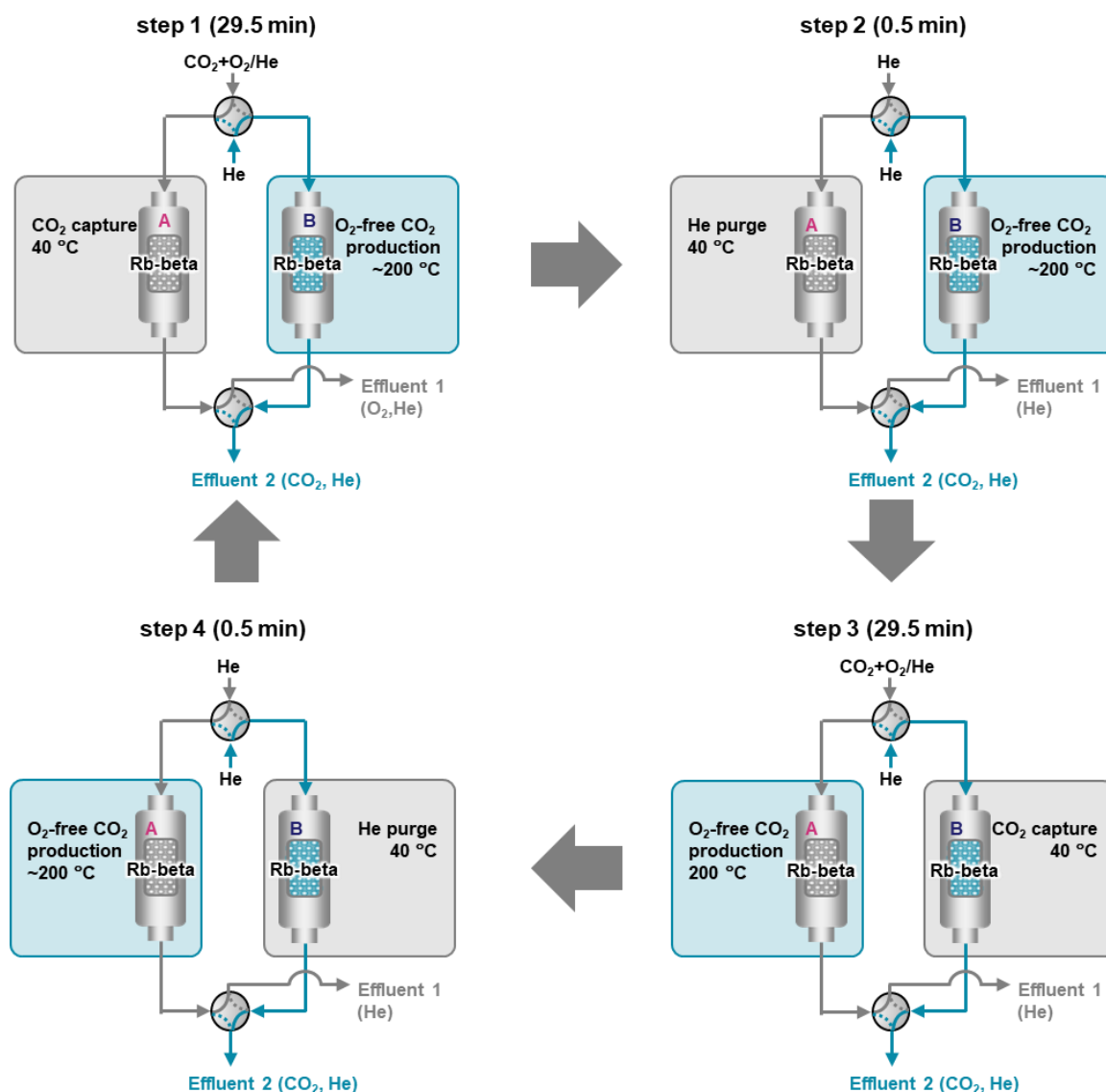


Fig. S7 Schematic diagram and procedure of the continuous CO₂ capture and O₂-free CO₂ production.

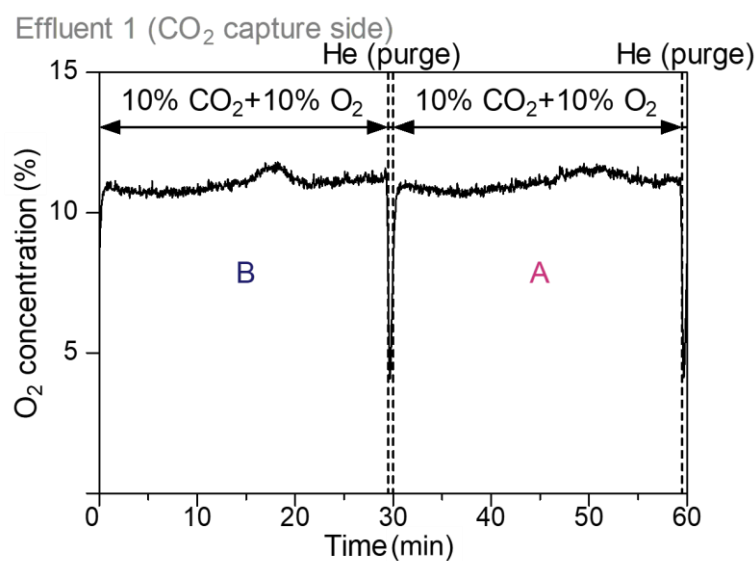


Fig. S8 Typical time course of the O₂ concentration in effluent 1 during continuous CO₂ capture and O₂-free CO₂ production.

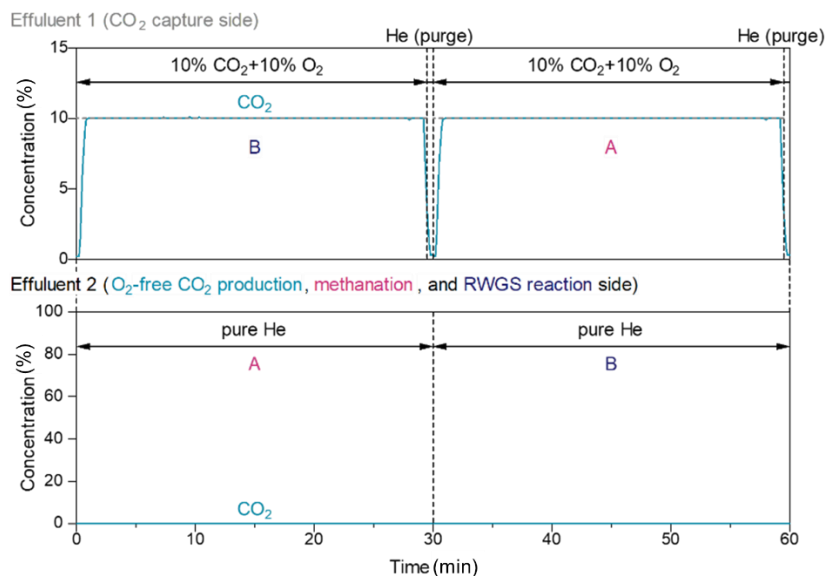


Fig. S9 Effluent concentration profiles of CO₂ for blank during continuous CO₂ capture and O₂-free CO₂ production, methanation and RWGS reaction side production; captured gas (100 mL min⁻¹, 10%CO₂+10%O₂/He for 29.5 min and then pure He for 0.5 min) and other side gas (120 mL min⁻¹, pure He for 30 min).

Text S3

In continuous CO₂ capture and O₂-free CO₂ production, the total flow rate was moderately changed due to O₂-free CO₂ production. The total flow rate in effluent 2 can be shown eq. 1. The produced O₂-free CO₂ flow rate can be calculated in eq. 2. Finally, eq. 3 to derive the total flow rate was calculated from eqs. 1 and 2. The derived total flow rate is close to the measured total flow rate by the soap-film flow meter (HORIBA, Ltd., Fluid Control System SF-1U combined VP-3U, Fig. S7). From eqs. 2 and 6, the amount of produced O₂-free CO₂ every 0.5 min was derived and O₂-free CO₂ yield was also calculated (O₂-free CO₂ yield = 97%, Fig. S7).

$$F_{all}^{out(2)} = F_{CO_2}^{out(2)} + F_{He}^{out(2)} \quad (1)$$

$$F_{CO_2}^{out(2)} = C_{CO_2}^{out(2)} \times F_{all}^{out(2)} \quad (2)$$

$$F_{all}^{out(2)} = \frac{F_{He}^{out(2)}}{1 - C_{CO_2}^{out(2)}} \quad (3)$$

Effluent 2 (O₂-free CO₂ production side)

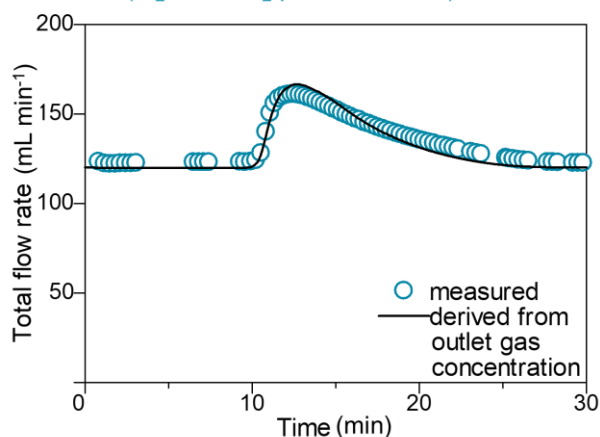


Fig. S10 Comparison of changes about measured total flow rate and derived total flow rate from outlet gas concentration in effluent 2 during CO₂ capture and O₂-free CO₂ production. Conditions are the same as in Fig. 5. The methodology of total flow rate derivation from outlet gas concentration in effluent 2 is shown in Supplementary Text 1.

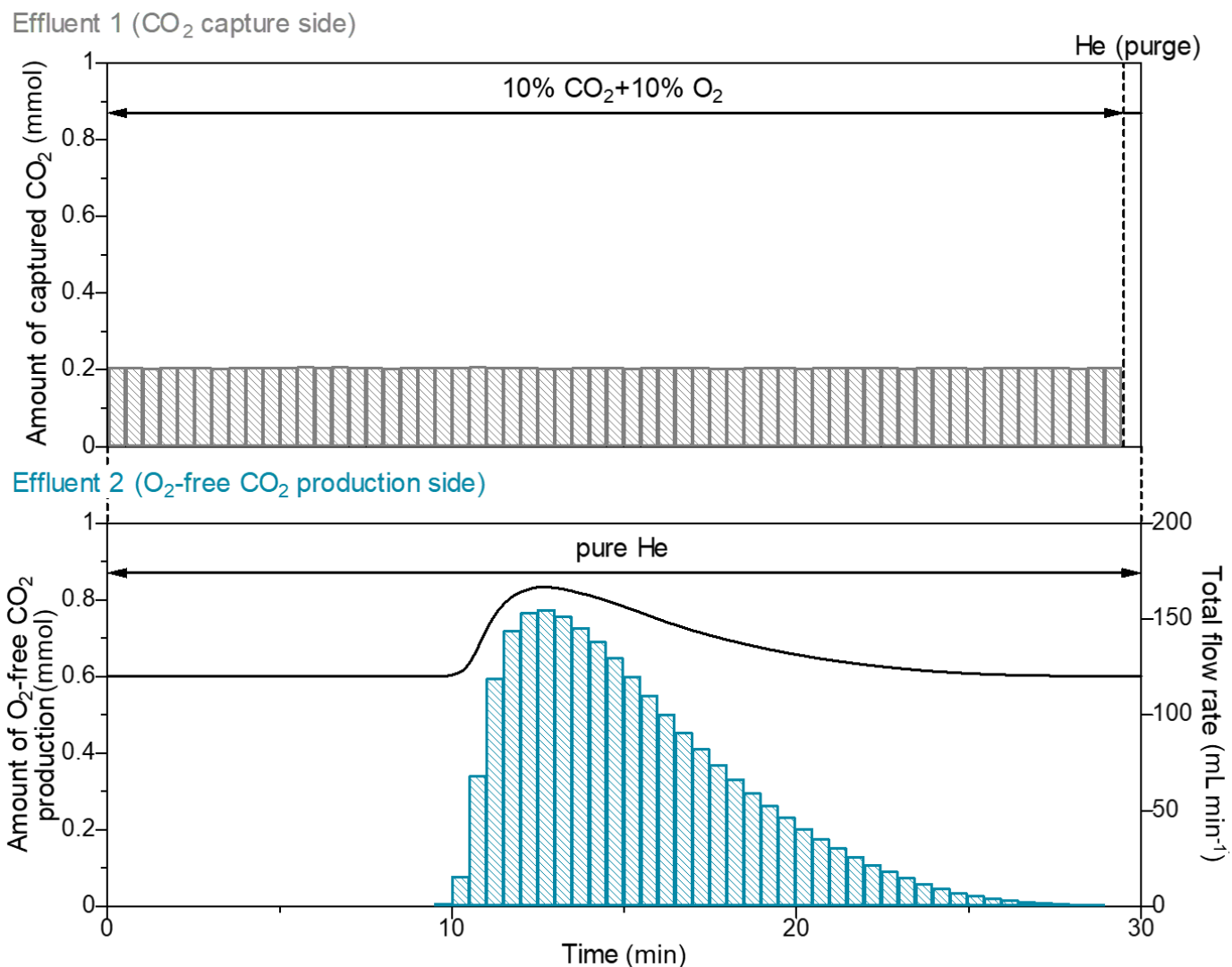


Fig. S11 Time course of amount of captured CO_2 in effluent 1, O_2 -free CO_2 production in effluent 2 every 0.5 min, and total flow rate of effluent 2 for continuous CO_2 capture and RWGS reaction. Conditions are the same as in Fig. 5.

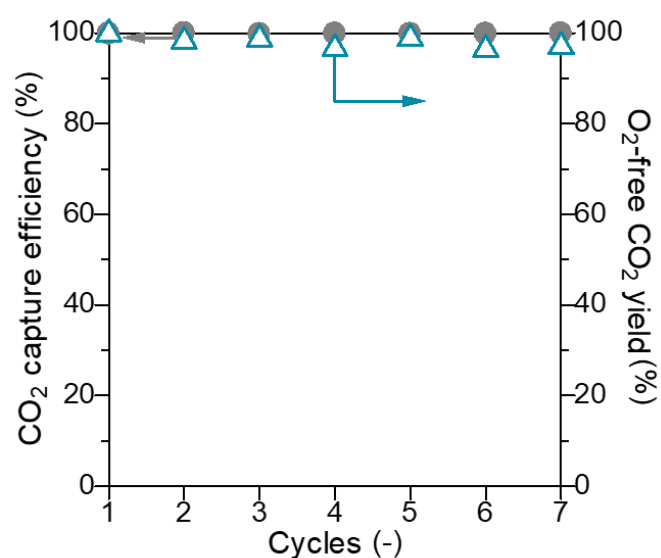


Fig. S12 Transitions of the CO_2 capture efficiency and O_2 -free CO_2 yield during cyclic test of continuous CO_2 capture and O_2 -free CO_2 production. Conditions are the same as in Fig. 5.

2.3 Continuous CO₂ capture and methanation/RWGS reaction using the tandem system

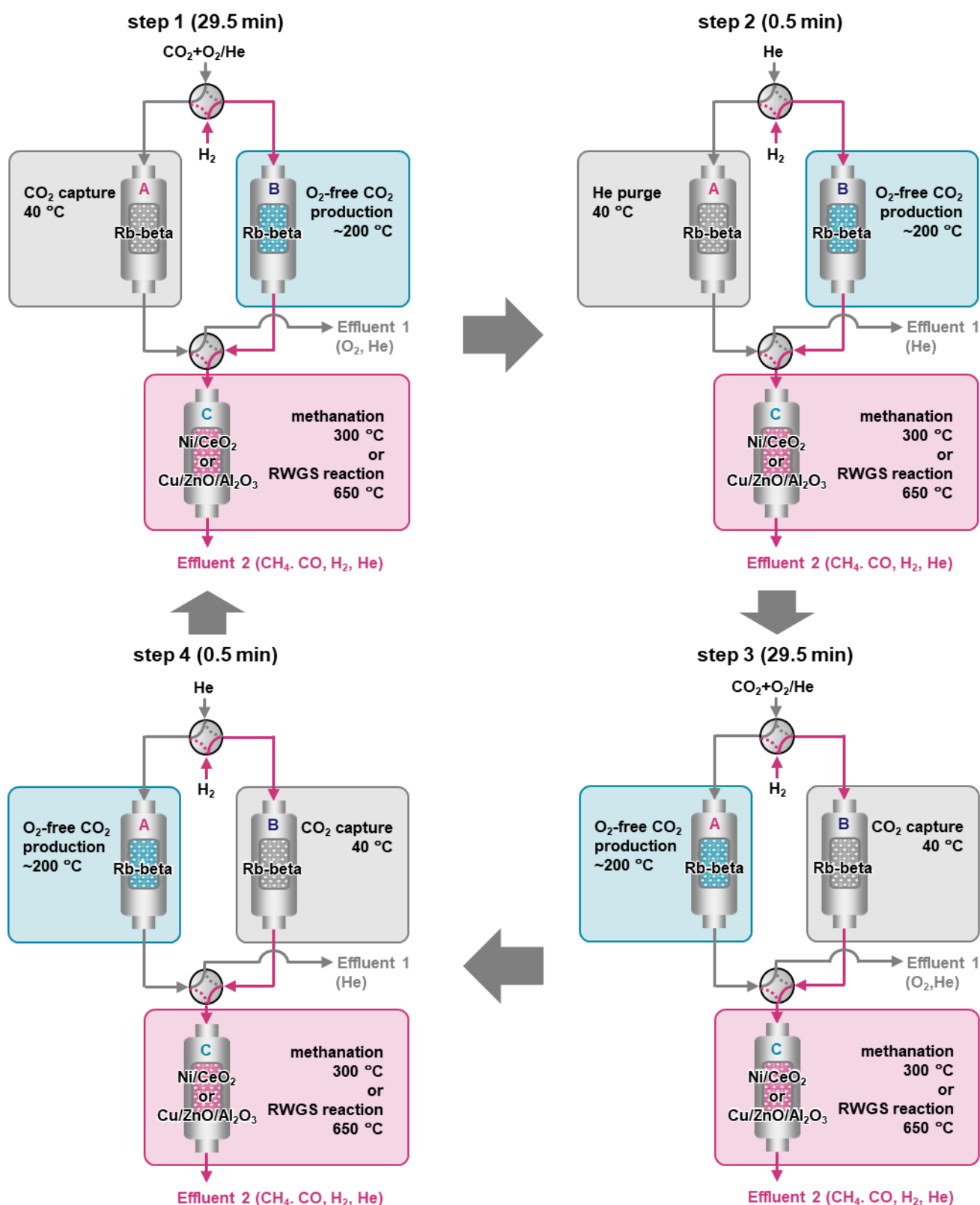


Fig. S13 Schematic diagram and procedure of the continuous CO₂ capture and methanation as well as RWGS reaction.

Text S4

In continuous high-concentration CO₂ capture and methanation, total flow rate was drastically changed due to large amount of CH₄ formation. The total flow rate in effluent 2 can be shown eq. 1, because of produced CO and unreacted CO₂ are hardly observed, and produced H₂O was captured by the cold trap. From the stoichiometric equation of methanation (eq. 3 in Main Text), produced CH₄ and unreacted H₂ flow rates can be shown eqs. 2 and 3, respectively. amount of converted H₂ can be calculated by eq. 4 and eq. 4 can transformed to eq. 5 using eq. 2. Finally, eq. 6 to derive the total flow rate was calculated from eqs. 4 and 5. The derived total flow rate is close to the measured total flow rate by the soap-film flow meter (HORIBA, Ltd., Fluid Control System SF-1U combined VP-3U). From eqs. 2 and 6, the amount of produced CH₄ every 0.5 min was derived and CH₄ yield was also calculated (CH₄ yield = 92%, Fig. S10).

$$F_{all}^{out(2)} = F_{CH_4}^{out} + F_{H_2}^{out} \quad (1)$$

$$F_{CH_4}^{out} = C_{CH_4}^{out} \times F_{all}^{out(2)} \quad (2)$$

$$F_{H_2}^{out} = (1 - C_{CH_4}^{out}) \times F_{all}^{out(2)} \quad (3)$$

$$F_{H_2}^{in} - F_{H_2}^{out} = 4 \times F_{CH_4}^{out} \quad (4)$$

$$F_{H_2}^{in} - F_{H_2}^{out} = 4 \times C_{CH_4}^{out} \times F_{all}^{out(2)} \quad (5)$$

$$F_{all}^{out} = \frac{F_{H_2}^{in}}{1 + 3C_{CH_4}^{out}} \quad (6)$$

Effluent 2 (methanation side)

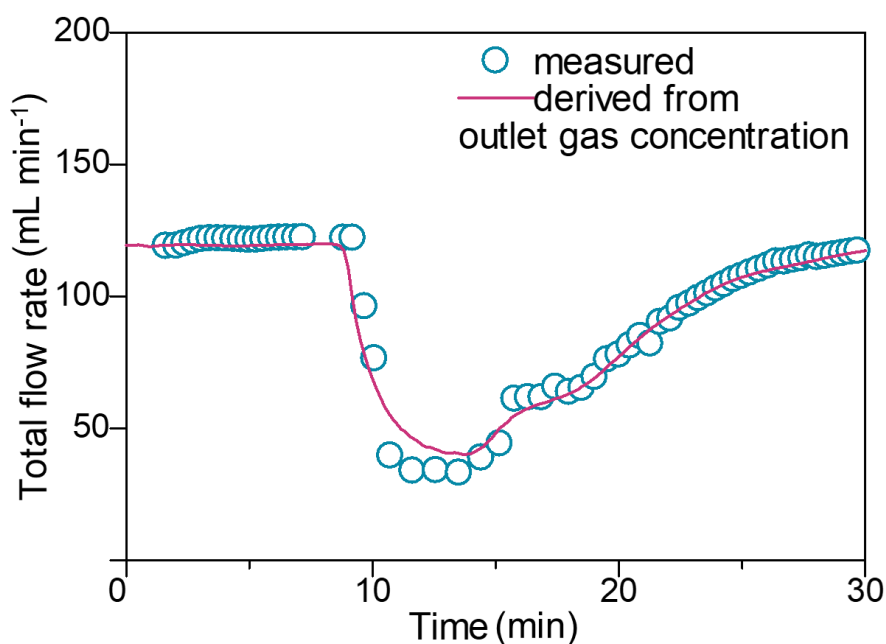


Fig. S14 Comparison of changes about measured total flow rate and derived total flow rate from outlet gas concentration in effluent 2 during CO₂ capture and methanation. Conditions are the same as in Fig. 5. The methodology of total flow rate derivation from outlet gas concentration in effluent 2 is shown in Supplementary Text 3.

Effluent 1 (CO₂ capture side)

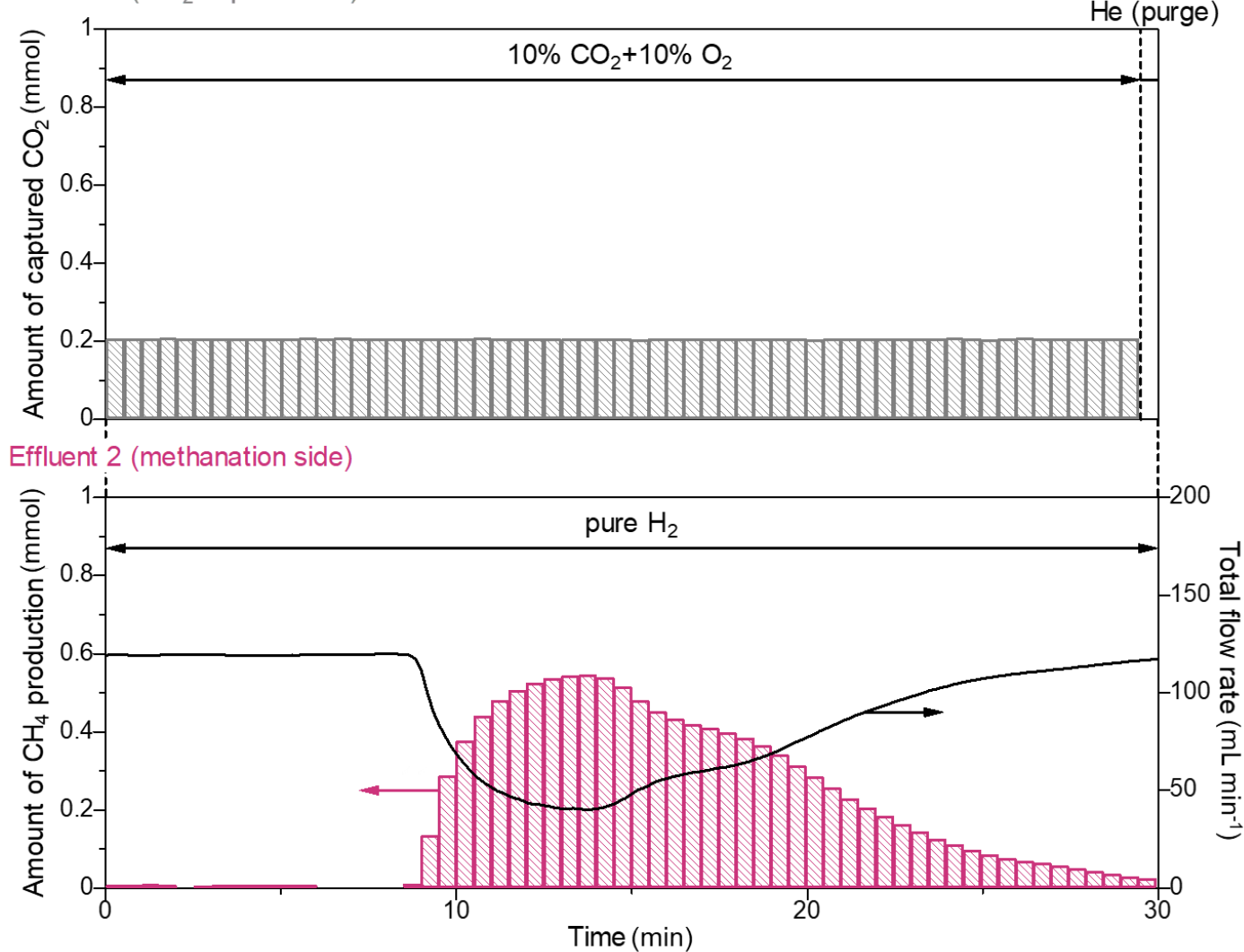


Fig. S15 Time course of amount of captured CO₂ in effluent 1, CH₄ production in effluent 2 every 0.5 min, and total flow rate of effluent 2 for continuous CO₂ capture and methanation. Conditions are the same as in Fig. 5.

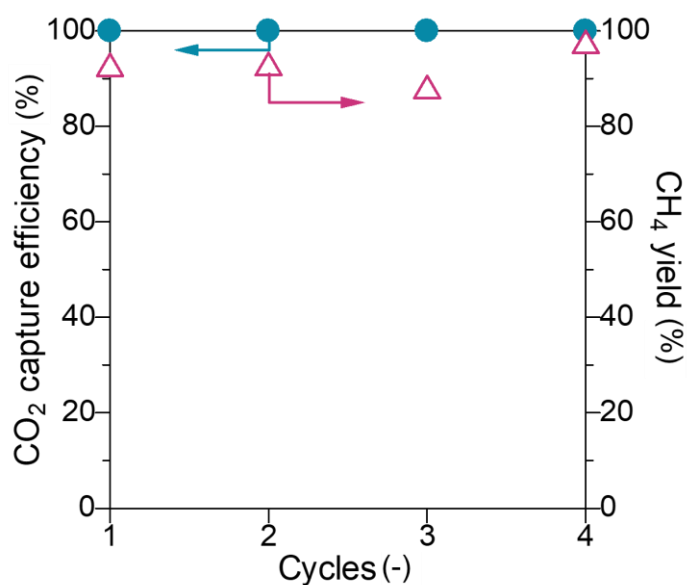


Fig. S16 Transitions of the CO₂ capture efficiency and CH₄ yield during cyclic test of continuous CO₂ capture and RWGS reaction. Conditions are the same as in Fig. 5.

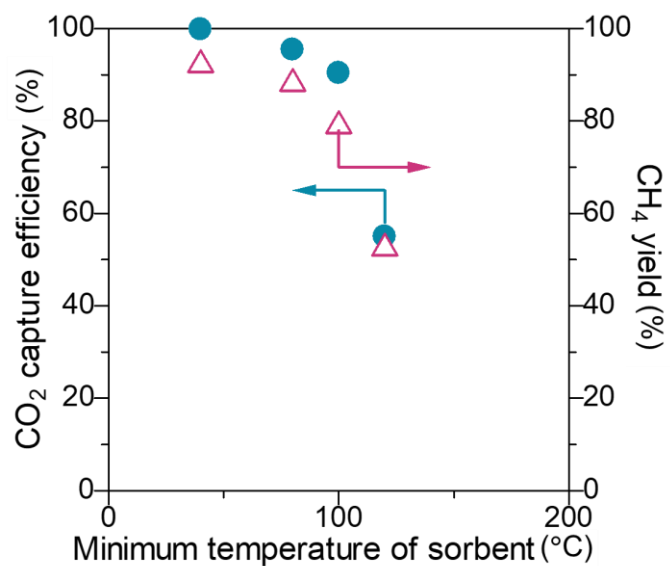


Fig. S17 Variation in CO₂ capture efficiency and CH₄ yield with range of temperature changing (minimum range from 40 °C to 120 °C) in continuous CO₂ capture and CH₄ production.

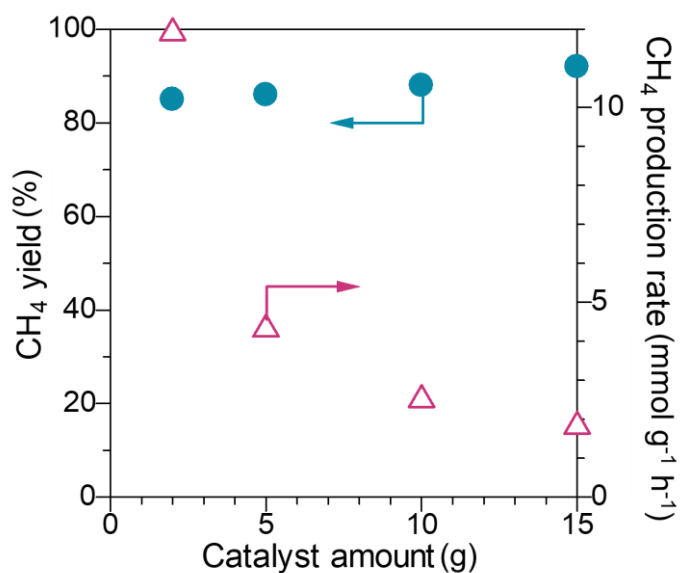


Fig. S18 Variation in CH₄ yield and CH₄ production rate with catalyst amount in continuous CO₂ capture and CH₄ production.

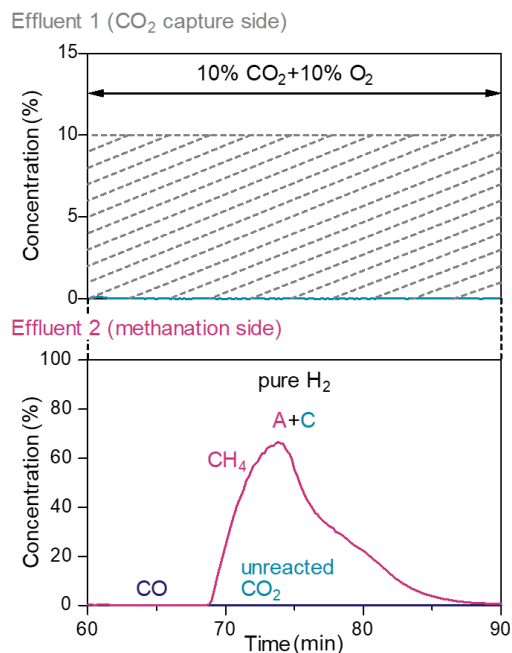


Fig. S19 Typical time course of uncaptured CO₂ in effluent 1 and CH₄, CO, and unreacted CO₂ in effluent 2 for continuous CO₂ capture and methanation under process-relevant conditions (without He purge). Conditions: 14 g of Rb-beta for each upper reactor, temperature swing from 40 °C to 200 °C (heating rate = 5.5 °C/min). 15 g of Ni/CeO₂ for the bottom reactor, 300 °C. 100 mL/min 10% CO₂+10% O₂/He for 30 min, switched to 120 mL/min H₂ for another 30 min.

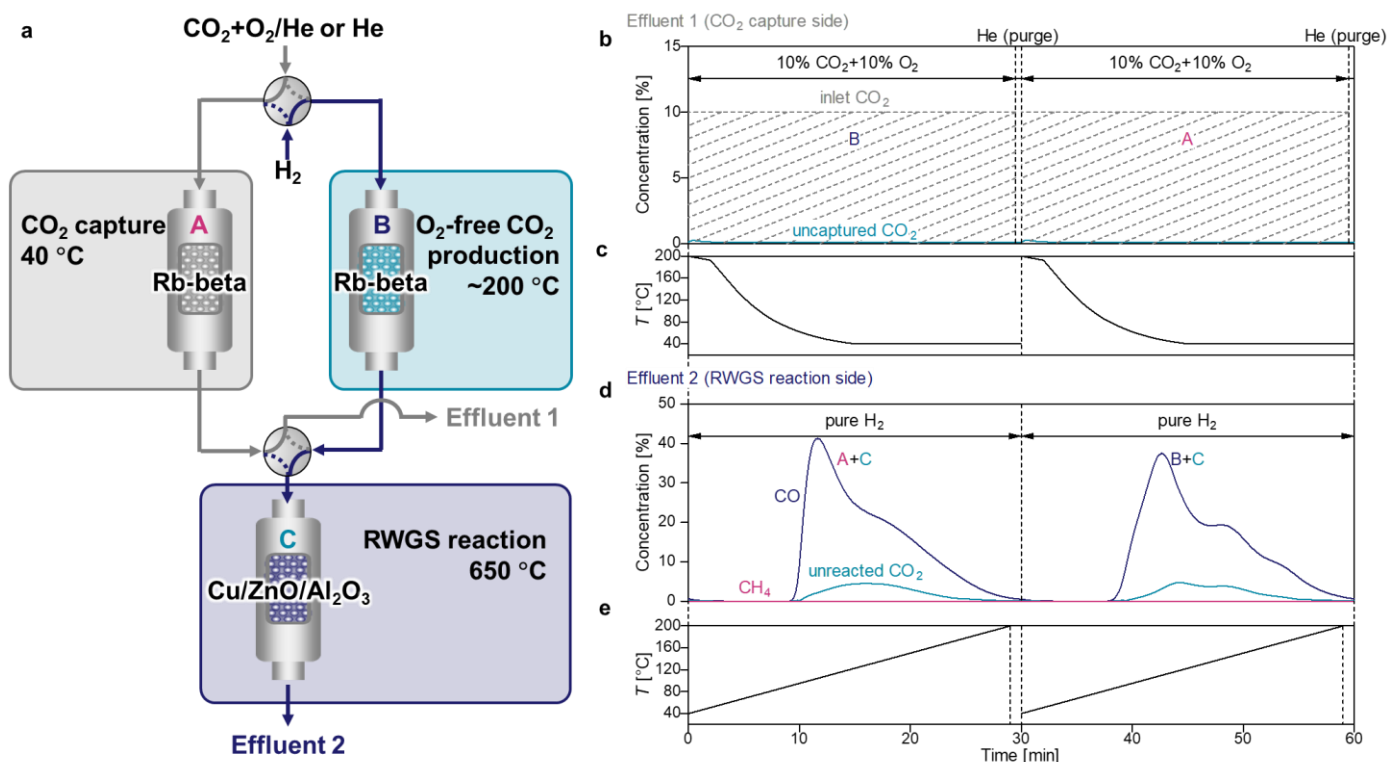


Fig. S20 (a) Schematic diagram of the two-reactor TSA system for continuous CO₂ capture and RWGS reaction; captured gas (100 mL/min, 10%CO₂+10%O₂/He for 29.5 min and then pure He for 0.5 min) and hydrogenation gas (100 mL/min, pure H₂ for 30 min) were alternately fed into each reactor containing 14 g of Rb-beta (b and d) Typical time course of the CO₂ and O₂ concentration in effluent 1 and 2, respectively. (c and e) Typical time course of the temperature changing in reactor a and b, respectively.

Text S5

In continuous high-concentration CO₂ capture and RWGS reaction, total flow rate was slightly changed due to CO formation and CO₂ desorption. Using the soap-film flow meter, total flow rate in effluent 2 was measured and the amount of produced CO every 0.5 min was derived, and CO yield was also calculated (CO yield = 85%, Fig. S18).

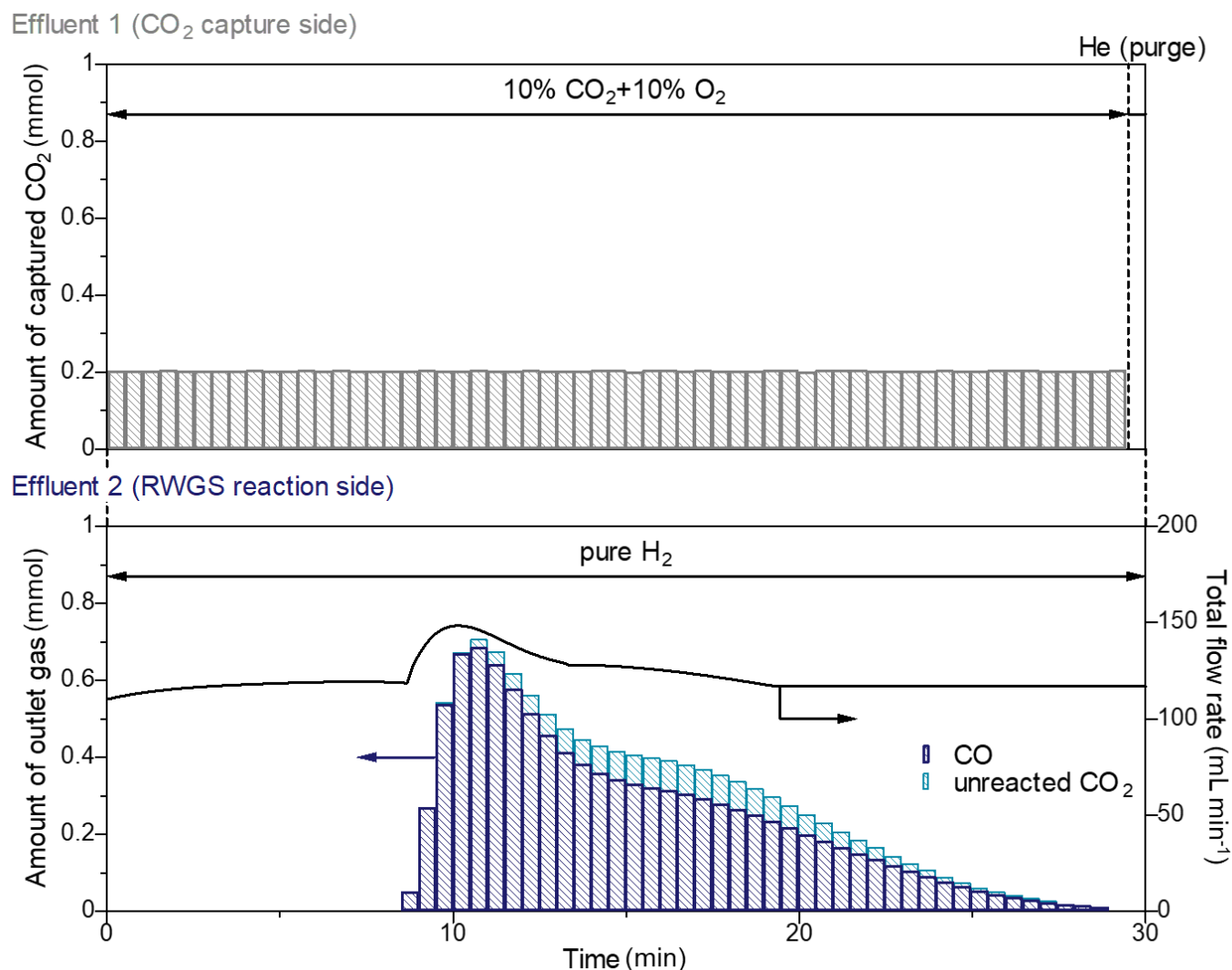


Fig. S21 Time course of amount of captured CO₂ in effluent 1, CO production in effluent 2 every 0.5 min, and total flow rate of effluent 2 for continuous CO₂ capture and RWGS reaction. Conditions are the same as in Fig. S17.

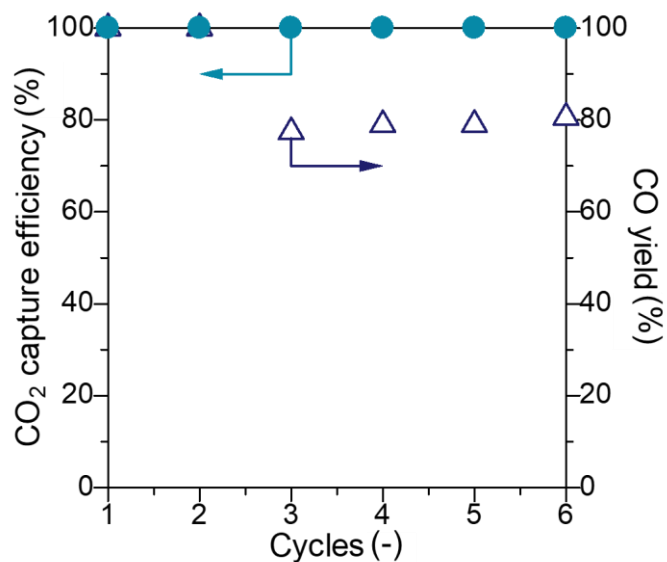


Fig. S22 Transitions of the CO₂ capture efficiency and CO yield during the cyclic test of continuous CO₂ capture and RWGS reaction. Conditions are the same as in Fig. S17.

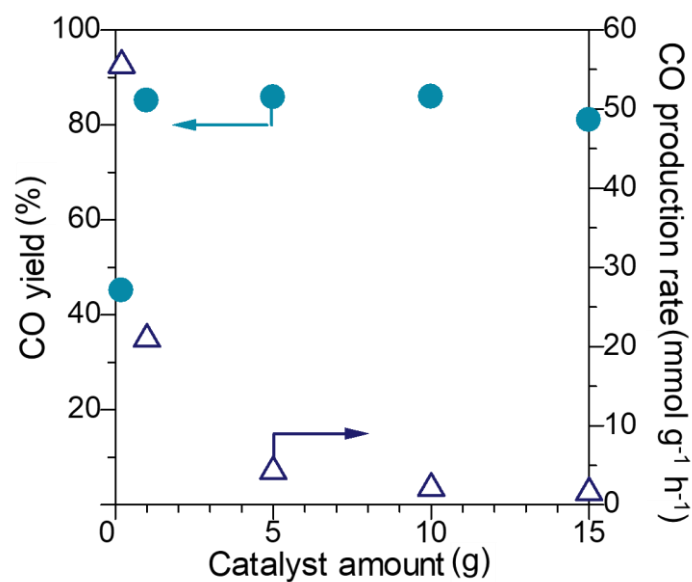


Fig. S23 Variation in CO yield and STY_{CO} with catalyst amount in continuous CO₂ capture and RWGS Reaction.

Text S6

H₂ conversion is also an important property in the CO₂ methanation process. We demonstrated continuous CO₂ capture and methanation, with the H₂ flow time of H₂ reduced from 30 min to 20 min. Similar to Fig. 6, 10% CO₂ + 10% O₂ was fed into one reactor for 29.5 min for CO₂ capture with cooling, and then pure He (0.5 min) was fed to purge the remaining O₂ in the reactor and gas line (Fig. S24a). At the same time, into the other reactor in parallel, pure H₂ was fed at 20 min after 10 min of no gas flow with heating. CO₂ capture efficiency and CH₄ yield were maintained during 4 cycles at 99% and 93%, respectively (Fig. S24e). The effect of H₂ flow time was investigated and H₂ conversion and the average CH₄ concentration were increased from 38% to 58%, and from 9.8% to 15%, respectively.

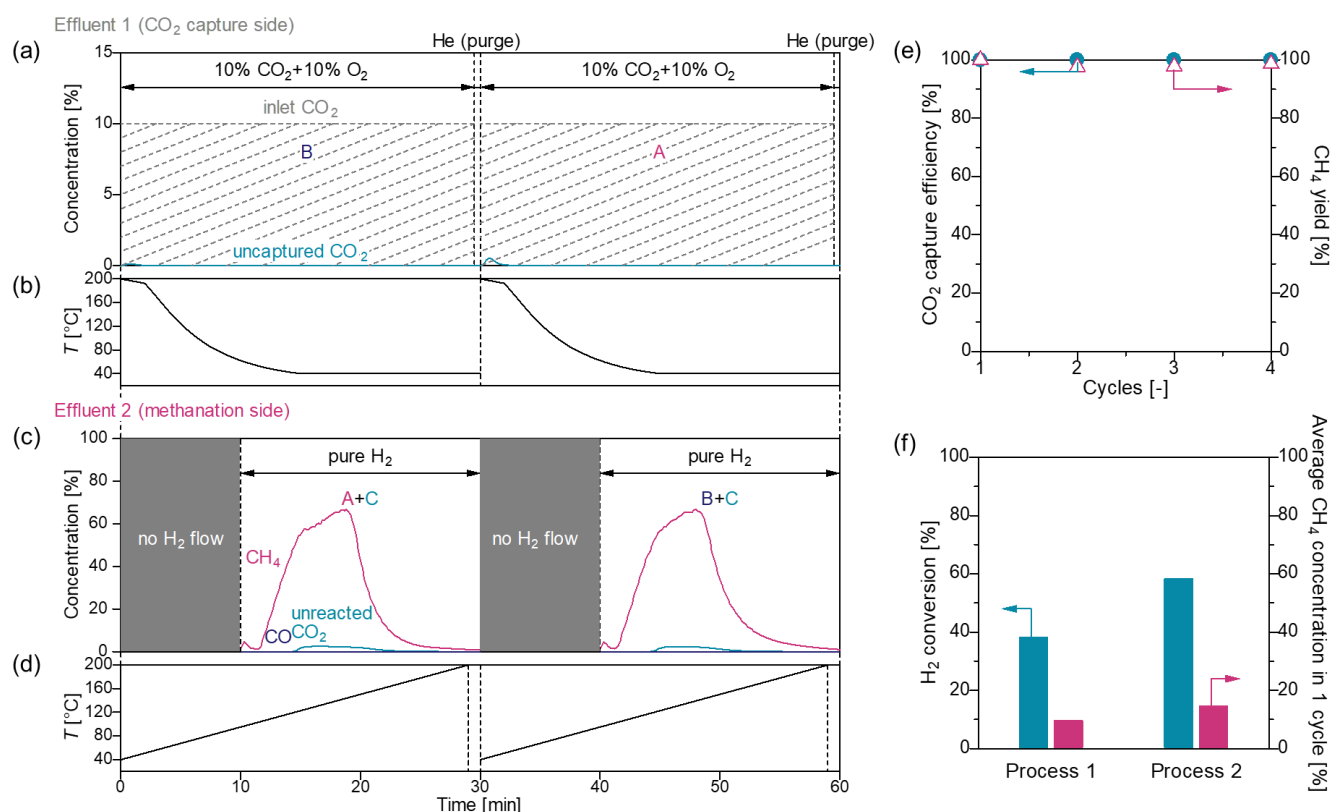


Fig. S24 Continuous CO₂ capture and methanation; captured gas (100 mL min⁻¹, 10% CO₂+10%O₂/He for 29.5 min and then pure He for 0.5 min) and hydrogenation gas (100 mL min⁻¹, pure H₂ for 20 min) were alternately fed into each reactor containing 14 g of Rb-beta. (a and c) Typical time course of the CH₄, CO₂, and CO concentration in effluent 1 and 2, respectively. (b and d) Typical time course of the temperature changing in reactor a and b, respectively. (e) Transitions of the CO₂ capture efficiency and CH₄ yield during cyclic test. (f) Comparison of H₂ conversion and average CH₄ concentration in 1 cycle between conditions of Fig. 6 (denoted as process 1) and conditions of Fig. S24 (denoted as process 2).

2.4 Continuous direct air capture and methanation

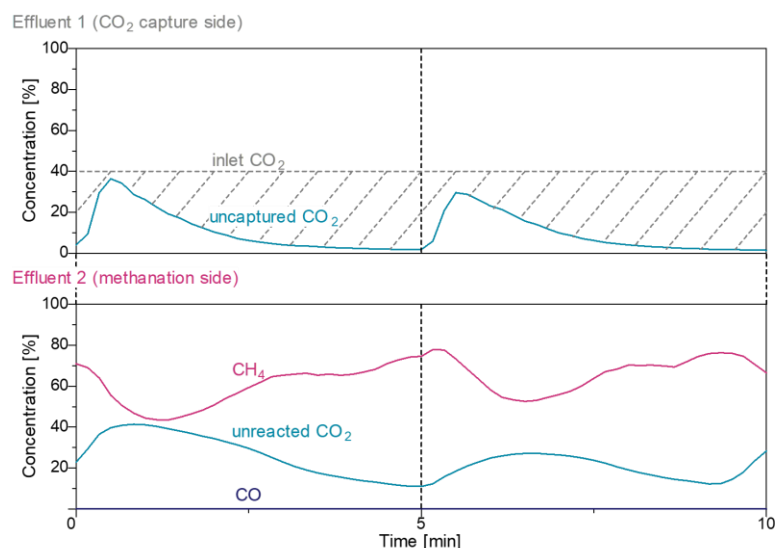


Fig. S25 Typical time course of unaptured CO₂ in effluent 1 and CH₄, CO, and unreacted CO₂ in effluent 2 for continuous high-concentration CO₂ capture and methanation. Conditions: 14 g of Rb-beta for each upper reactor, temperature swing from 80 °C to 100 °C (heating rate = 20 °C/min). 15 g of Ni/CeO₂ for the bottom reactor, 300 °C. 500 mL/min air for 5 min, switched to 10 mL min⁻¹ H₂ for another 5 min.

2.5 Energy efficiency

Text S7

The energy efficiency (denoted as η) was defined as the ratio between the outlet energy based on the low heating value (LHV) of CH₄ and the overall energy. For example, Andrew et al. evaluated the η of CO₂ methanation under both pseudo-adiabatic and adiabatic configurations, and demonstrated that the adiabatic configuration significantly enhances the overall efficiency of the methanation process[†]. While the definition of η excludes the contribution of the LHV of unreacted H₂, it serves as a reliable metric for evaluating and comparing the efficiency of CO₂ conversion. In contrast, the fuel production efficiency (FPE) is defined as the ratio of the total output energy to the total input energy. Both input and output energies were calculated based on the LHV and the applied electrical power (P). Murphy et al. demonstrated a linear correlation between the flow rate–normalized input energy and the fuel production efficiency (FPE)[‡], highlighting the utility of FPE as an indicator of how effectively CO₂ and H₂ are jointly converted into higher-value energy outputs.

[†] M. Biset-Peiró, R. Meyb, J. Guilerá, and T. Andreua, *Chem. Eng. J.* 2020, **393**, 124786

[‡] S. Ullah, Y. Gao, L. Dou, Y. Liu, T. Shao, Y. Yang and A. B. Murphy, *Plasma Chemistry and Plasma Processing*, 2023, **43**, 1335–1383.

Tables

2 Results and Discussion

Table S1. Summary of the continuous CO₂ capture and methanation in this study and other reported CO₂ capture and methanation considering the effect of coexistent O₂. Entries without *, †, or ‡ symbols using Fig. 1a system.

DFMs or catalyst	CO ₂ capture gas	Hydroge- nation gas	CH ₄ yield [%]	T [°C]	ref
Ni(5)/CeO ₂ *	10%CO ₂ +10%O ₂ /He	100% H ₂	92	300	*
Ru(1)-Ni(10)-Na ₂ O(6.1) /Al ₂ O ₃	7.5%CO ₂ +4.5%O ₂ +15%H ₂ O/N ₂	15%H ₂ /N ₂	5.67	320	¹
Pt(1)-Ni(10)-Na ₂ O(6.1) /Al ₂ O ₃	7.5%CO ₂ +4.5%O ₂ +15%H ₂ O/N ₂	15%H ₂ /N ₂	3.73	320	¹
Ru(1)-Na ₂ O(6.1) /Al ₂ O ₃	7.5%CO ₂ +4.5%O ₂ +15%H ₂ O/N ₂	15%H ₂ /N ₂	4.63	320	¹
Ru(5)-Na ₂ O(6.1) /Al ₂ O ₃	7.5%CO ₂ +4.5%O ₂ +15%H ₂ O/N ₂	15%H ₂ /N ₂	4.35	320	²
Ni(10)-Na ₂ O(6.1) /Al ₂ O ₃	7.5%CO ₂ +4.5%O ₂ +15%H ₂ O/N ₂	15%H ₂ /N ₂	4.12	320	²
Ru(0.95)-K(5) /Al ₂ O ₃	1%CO ₂ +3%O ₂ + 2.5%H ₂ O/He	4%H ₂ /He	0.38	350	³
Ru(0.95)-Ca(5.1) /Al ₂ O ₃	1%CO ₂ +3%O ₂ +2.5%H ₂ O/He	4%H ₂ /He	0.48	350	³
Ru(0.84)-Ba(16) /Al ₂ O ₃	1%CO ₂ +3%O ₂ +2.5%H ₂ O/He	4%H ₂ /He	2.22	350	³
Ni(10)-Ca(30)/Al ₂ O ₃ †	1%CO ₂ +10%O ₂ /N ₂	100%H ₂	0.15	450	⁴
Ni(10)/Al ₂ O ₃ †	1%CO ₂ +10%O ₂ /N ₂	100%H ₂	0.002	450	⁴
Ni(10)-Ca(6)/Al ₂ O ₃ †	1%CO ₂ +10%O ₂ /N ₂	100%H ₂	0.08	450	⁴
Ni(10)-Ca(20)/Al ₂ O ₃ †	1%CO ₂ +10%O ₂ /N ₂	100%H ₂	0.09	450	⁴
Ni(10)-Ca(40)/Al ₂ O ₃ †	1%CO ₂ +10%O ₂ /N ₂	100%H ₂	0.15	450	⁴
Ni(5)-Ca(30)/Al ₂ O ₃ †	1%CO ₂ +10%O ₂ /N ₂	100%H ₂	0.13	450	⁴
Ni(20)-Ca(30)/Al ₂ O ₃ †	1%CO ₂ +10%O ₂ /N ₂	100%H ₂	0.14	450	⁴
Ca(30)-Ni(10)/Al ₂ O ₃ †	1%CO ₂ +10%O ₂ /N ₂	100%H ₂	0.064	450	⁴
Ni(10)/Al ₂ O ₃	2.5%CO ₂ +10%O ₂ /N ₂	100%H ₂	0.43	500	⁵
Ni(10)/CaO	2.5%CO ₂ +10%O ₂ /N ₂	100%H ₂	20.4	500	⁵
Ni(10)-Ca(28)/Al ₂ O ₃	2.5%CO ₂ +10%O ₂ /N ₂	100%H ₂	29.9	500	⁵
Ni(10)-Ca(8)/Al ₂ O ₃	2.5%CO ₂ +10%O ₂ /N ₂	100%H ₂	8.66	500	⁵
Ni(10)-Ca(14)/Al ₂ O ₃	2.5%CO ₂ +10%O ₂ /N ₂	100%H ₂	15.54	500	⁵
Ni(10)-Ca(32)/Al ₂ O ₃	2.5%CO ₂ +10%O ₂ /N ₂	100%H ₂	24.1	500	⁵
Ni(30)-Ca(28)/Al ₂ O ₃	2.5%CO ₂ +10%O ₂ /N ₂	100%H ₂	20.7	500	⁵
Ni(50)-Ca(28)/Al ₂ O ₃	2.5%CO ₂ +10%O ₂ /N ₂	100%H ₂	22.63	500	⁵
Ni(10)-Ca(28)/Al ₂ O ₃	2.5%CO ₂ +10%O ₂ /N ₂	100%H ₂	15.1	400	⁵
Ni(10)-Ca(28)/Al ₂ O ₃	2.5%CO ₂ /10%O ₂ /N ₂	100%H ₂	5.91	300	⁵
Ni(10)-Ca(6)/Al ₂ O ₃ †	0.25%CO ₂ +10%O ₂ /N ₂	100%H ₂	14.0	350	⁶
Ni ₂ Ca ₂ -Mg ₂ Al ₂ /LDH	10%CO ₂ +5%O ₂ /He	20%H ₂ /He	15.68	320	⁷
Ni ₂ Ca ₄ -Al ₂ /LDH	10%CO ₂ +5%O ₂ /He	20%H ₂ /He	12.88	320	⁷

Ni ₂ Mg ₂ -Al ₂ -LDH	10%CO ₂ +5%O ₂ /He	20%H ₂ /He	6.72	320	⁷
Ni(30)/CaO(15)-MgO(15)-Al ₂ O ₃	10%CO ₂ +5%O ₂ /He	20%H ₂ /He	6.16	320	⁷
Ru(0.84)-Ba(16)/Al ₂ O ₃	1%CO ₂ +3%O ₂ +2.5%H ₂ O/He	4%H ₂ /He	1.12	350	⁸
Ru(1)/Al ₂ O ₃ + Ba(16)/Al ₂ O ₃	1%CO ₂ +3%O ₂ +2.5%H ₂ O/He	4%H ₂ /He	0.63	350	⁸
Na-Ni/Al ₂ O ₃	5%CO ₂ +4.5%O ₂ +11%H ₂ O/Ar	5%H ₂ /Ar	0.47	350	⁹
K-Ni/Al ₂ O ₃	5%CO ₂ +11%H ₂ O+4.5%O ₂ /Ar	5%H ₂ /Ar	2.333	350	⁹
Ba-Ni/Al ₂ O ₃	5%CO ₂ +11%H ₂ O+4.5%O ₂ /Ar	5%H ₂ /Ar	0.093	350	⁹
Ru(0.5)-Na ₂ O(6.1) /Al ₂ O ₃	7.5%CO ₂ +4.5%O ₂ +15%H ₂ O/N ₂	15%H ₂ /N ₂	13.44	320	¹⁰
Ru(1)-Na ₂ O(6.1) /Al ₂ O ₃	7.5%CO ₂ +4.5%O ₂ +15%H ₂ O/N ₂	15%H ₂ /N ₂	14.93	320	¹⁰
Ru(1)-Na/Al ₂ O ₃	10%CO ₂ +10%O ₂ +10%H ₂ O/He	10%H ₂ /Ar	2.6	300	¹¹
Ru(1)-Na/Al ₂ O ₃	10%CO ₂ +10%O ₂ /He	10%H ₂ /Ar	15.4	300	¹¹
Ru(1)-Na/Al ₂ O ₃	10%CO ₂ +10%O ₂ /He	10%H ₂ /Ar	24.3	300	¹¹
HT-23NiR	7.5%CO ₂ +4.5%O ₂ /He	100%H ₂	0.020	250	¹²
HT-23NiR	7.5%CO ₂ +4.5%O ₂ /He	100%H ₂	0.028	300	¹²
HT-23NiR	7.5%CO ₂ +4.5%O ₂ /He	100%H ₂	0.022	320	¹²
HT-46NiR	7.5%CO ₂ +4.5%O ₂ /He	100%H ₂	0.032	250	¹²
HT-46NiR	7.5%CO ₂ +4.5%O ₂ /He	100%H ₂	0.038	300	¹²
HT-46NiR	7.5%CO ₂ +4.5%O ₂ /He	100%H ₂	0.032	320	¹²
Ni-Pr/CeO ₂	10%CO ₂ +10%O ₂ /Ar	10%H ₂ /Ar	3.11	300	¹³
RuNi-Pr/CeO ₂	10%CO ₂ +10%O ₂ /Ar	10%H ₂ /Ar	3.70	300	¹³

*This study, † using Fig. 2b system.

Table S2. Summary of the reported CO₂ capture and methanation. Entries without *, †, or ‡ symbols using Fig. 1a system.

DFMs or catalyst	CO ₂ capture gas	Hydrogenation gas	CH ₄ yield [%]	T [°C]	ref
Ru(5)-Na ₂ CO ₃ (10)/Al ₂ O ₃	5%CO ₂ /N ₂	5%H ₂ /N ₂	5.23	320	14
Ru(5)-K ₂ CO ₃ (10)/Al ₂ O ₃	5%CO ₂ /N ₂	5%H ₂ /N ₂	4.53	320	14
Ni(1)/CeCaCO ₃	15%CO ₂ /N ₂	100%H ₂	8.96	550	15
Ni(10)/CaO	15%CO ₂ /N ₂	100%H ₂	3.73	550	15
Ni(1)/CeO ₂ -CaOphy	15%CO ₂ /N ₂	100%H ₂	11.9	550	15
Ni(10)/g-Al ₂ O ₃	5%CO ₂ /N ₂	100%H ₂	0.19	450	16
Ni(10)-Na(15)/g-Al ₂ O ₃	5%CO ₂ /N ₂	100%H ₂	3.33	450	16
Ni(10)-K(15)/g-Al ₂ O ₃	5%CO ₂ /N ₂	100%H ₂	2.51	450	16
Ni(10)-Ca(15)/g-Al ₂ O ₃	5%CO ₂ /N ₂	100%H ₂	1.08	450	16
Ni(10)-Na(15)/g-Al ₂ O ₃	5%CO ₂ /N ₂	100%H ₂	4.77	450	16
Ni(10)-Na(15)/g-Al ₂ O ₃	0.04%CO ₂ /N ₂	100%H ₂	17.8	450	16
Ni(10)-Na(15)/g-Al ₂ O ₃	0.01%CO ₂ /N ₂	100%H ₂	49.7	450	16
Ni(10)-Na(15)/g-Al ₂ O ₃ [‡]	2%CO ₂ /N ₂	100%H ₂	99	400	17
Ru(10)/CaO	1.4%CO ₂ /Ar	10%H ₂ /Ar	31.1	370	18
Ru(10)/Na ₂ CO ₃	1.4%CO ₂ /Ar	10%H ₂ /Ar	48.9	370	18
Ru(5)/CaO	11%CO ₂ /Ar	10%H ₂ /Ar	2.04	280	18
Ru(5)/CaO	11%CO ₂ /Ar	10%H ₂ /Ar	1.95	310	18
Ru(5)/CaO	11%CO ₂ /Ar	10%H ₂ /Ar	1.87	340	18
Ru(5)/CaO	11%CO ₂ /Ar	10%H ₂ /Ar	1.70	370	18
Ru(5)/CaO	11%CO ₂ /Ar	10%H ₂ /Ar	1.53	400	18
Ru(10)/CaO	11%CO ₂ /Ar	10%H ₂ /Ar	3.39	280	18
Ru(10)/CaO	11%CO ₂ /Ar	10%H ₂ /Ar	3.56	310	18
Ru(10)/CaO	11%CO ₂ /Ar	10%H ₂ /Ar	3.73	340	18
Ru(10)/CaO	11%CO ₂ /Ar	10%H ₂ /Ar	3.90	370	18
Ru(10)/CaO	11%CO ₂ /Ar	10%H ₂ /Ar	4.07	400	18
Ru(15)/CaO	11%CO ₂ /Ar	10%H ₂ /Ar	3.05	280	18
Ru(15)/CaO	11%CO ₂ /Ar	10%H ₂ /Ar	4.50	310	18
Ru(15)/CaO	11%CO ₂ /Ar	10%H ₂ /Ar	5.43	340	18
Ru(15)/CaO	11%CO ₂ /Ar	10%H ₂ /Ar	6.11	370	18
Ru(15)/CaO	11%CO ₂ /Ar	10%H ₂ /Ar	6.45	400	18
Ru(5)/Na ₂ CO ₃	11%CO ₂ /Ar	10%H ₂ /Ar	2.55	280	18
Ru(5)/Na ₂ CO ₃	11%CO ₂ /Ar	10%H ₂ /Ar	2.21	310	18
Ru(5)/Na ₂ CO ₃	11%CO ₂ /Ar	10%H ₂ /Ar	1.87	340	18
Ru(5)/Na ₂ CO ₃	11%CO ₂ /Ar	10%H ₂ /Ar	1.70	370	18

Ru(5)/Na ₂ CO ₃	11%CO ₂ /Ar	10%H ₂ /Ar	1.53	400	18
Ru(10)/Na ₂ CO ₃	11%CO ₂ /Ar	10%H ₂ /Ar	6.11	280	18
Ru(10)/Na ₂ CO ₃	11%CO ₂ /Ar	10%H ₂ /Ar	6.19	310	18
Ru(10)/Na ₂ CO ₃	11%CO ₂ /Ar	10%H ₂ /Ar	6.11	340	18
Ru(10)/Na ₂ CO ₃	11%CO ₂ /Ar	10%H ₂ /Ar	5.85	370	18
Ru(10)/Na ₂ CO ₃	11%CO ₂ /Ar	10%H ₂ /Ar	5.18	400	18
Ru(15)/Na ₂ CO ₃	11%CO ₂ /Ar	10%H ₂ /Ar	5.77	280	18
Ru(15)/Na ₂ CO ₃	11%CO ₂ /Ar	10%H ₂ /Ar	6.11	310	18
Ru(15)/Na ₂ CO ₃	11%CO ₂ /Ar	10%H ₂ /Ar	6.11	340	18
Ru(15)/Na ₂ CO ₃	11%CO ₂ /Ar	10%H ₂ /Ar	6.02	370	18
Ru(15)/Na ₂ CO ₃	11%CO ₂ /Ar	10%H ₂ /Ar	5.85	400	18
Ni(5)-CaO(15)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	1.70	520	19
Ni(10)-CaO(15)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	1.96	520	19
Ni(10)-CaO(15)/Al ₂ O ₃ coimp	10%CO ₂ /Ar	10%H ₂ /Ar	2.05	520	19
Ni(15)-CaO(15)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	2.65	520	19
Ni(5)-CaO(15)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	1.31	480	19
Ni(10)-CaO(15)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	1.90	480	19
Ni(10)-CaO(15)/Al ₂ O ₃ coimp	10%CO ₂ /Ar	10%H ₂ /Ar	1.87	480	19
Ni(15)-CaO(15)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	2.43	480	19
Ni(5)-CaO(15)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	1.4	440	19
Ni(10)-CaO(15)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	1.50	440	19
Ni(10)-CaO(15)/Al ₂ O ₃ coimp	10%CO ₂ /Ar	10%H ₂ /Ar	1.50	440	19
Ni(15)-CaO(15)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	2.33	440	19
Ni(5)-CaO(15)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	1.4	400	19
Ni(10)-CaO(15)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	1.49	400	19
Ni(10)-CaO(15)/Al ₂ O ₃ coimp	10%CO ₂ /Ar	10%H ₂ /Ar	1.31	400	19
Ni(15)-CaO(15)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	2.24	400	19
Ni(5)-CaO(15)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	1.31	360	19
Ni(10)-CaO(15)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	1.4	360	19
Ni(10)-CaO(15)/Al ₂ O ₃ coimp	10%CO ₂ /Ar	10%H ₂ /Ar	0.93	360	19
Ni(15)-CaO(15)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	2.05	360	19
Ni(5)-CaO(15)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	0.93	320	19
Ni(10)-CaO(15)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	1.12	320	19
Ni(10)-CaO(15)/Al ₂ O ₃ coimp	10%CO ₂ /Ar	10%H ₂ /Ar	0.47	320	19
Ni(15)-CaO(15)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	1.49	320	19
Ni(5)-CaO(15)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	0.19	280	19
Ni(10)-CaO(15)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	0.56	280	19
Ni(10)-CaO(15)/Al ₂ O ₃ coimp	10%CO ₂ /Ar	10%H ₂ /Ar	0.19	280	19

Ni(15)-CaO(15)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	0.93	280	19
Ni(5)-Na ₂ CO ₃ (10)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	1.87	520	19
Ni(10)-Na ₂ CO ₃ (10)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	1.96	520	19
Ni(10)-Na ₂ CO ₃ (10)/Al ₂ O ₃ coimp	10%CO ₂ /Ar	10%H ₂ /Ar	1.96	520	19
Ni(15)-Na ₂ CO ₃ (10)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	2.05	520	19
Ni(5)-Na ₂ CO ₃ (10)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	2.24	480	19
Ni(10)-Na ₂ CO ₃ (10)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	2.61	480	19
Ni(10)-Na ₂ CO ₃ (10)/Al ₂ O ₃ coimp	10%CO ₂ /Ar	10%H ₂ /Ar	2.52	480	19
Ni(15)-Na ₂ CO ₃ (10)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	2.52	480	19
Ni(5)-Na ₂ CO ₃ (10)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	2.33	440	19
Ni(10)-Na ₂ CO ₃ (10)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	2.8	440	19
Ni(10)-Na ₂ CO ₃ (10)/Al ₂ O ₃ coimp	10%CO ₂ /Ar	10%H ₂ /Ar	2.8	440	19
Ni(15)-Na ₂ CO ₃ (10)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	2.8	440	19
Ni(5)-Na ₂ CO ₃ (10)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	2.8	400	19
Ni(10)-Na ₂ CO ₃ (10)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	3.45	400	19
Ni(10)-Na ₂ CO ₃ (10)/Al ₂ O ₃ coimp	10%CO ₂ /Ar	10%H ₂ /Ar	3.43	400	19
Ni(15)-Na ₂ CO ₃ (10)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	3.47	400	19
Ni(5)-Na ₂ CO ₃ (10)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	3.36	360	19
Ni(10)-Na ₂ CO ₃ (10)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	3.17	360	19
Ni(10)-Na ₂ CO ₃ (10)/Al ₂ O ₃ coimp	10%CO ₂ /Ar	10%H ₂ /Ar	2.8	360	19
Ni(15)-Na ₂ CO ₃ (10)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	3.27	360	19
Ni(5)-Na ₂ CO ₃ (10)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	1.4	320	19
Ni(10)-Na ₂ CO ₃ (10)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	2.05	320	19
Ni(10)-Na ₂ CO ₃ (10)/Al ₂ O ₃ coimp	10%CO ₂ /Ar	10%H ₂ /Ar	1.49	320	19
Ni(15)-Na ₂ CO ₃ (10)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	2.33	320	19
Ni(5)-Na ₂ CO ₃ (10)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	0.093	280	19
Ni(10)-Na ₂ CO ₃ (10)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	0.56	280	19
Ni(10)-Na ₂ CO ₃ (10)/Al ₂ O ₃ coimp	10%CO ₂ /Ar	10%H ₂ /Ar	0.37	280	19
Ni(15)-Na ₂ CO ₃ (10)/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	0.65	280	19
Ru(2.5)/CeO ₂	65%CO ₂ /N ₂	5%H ₂ /N ₂	2.96	300	20
Ru(5)/CeO ₂	65%CO ₂ /N ₂	5%H ₂ /N ₂	3.41	300	20
Ru(10)/CeO ₂	65%CO ₂ /N ₂	5%H ₂ /N ₂	3.65	300	20
Ni(20)/MgO	10%CO ₂ /Ar	5%H ₂ /Ar	0.17	250	21
Ni(20)/MgO	10%CO ₂ /Ar	5%H ₂ /Ar	0.50	300	21
Ni(20)/MgO	10%CO ₂ /Ar	5%H ₂ /Ar	0.45	350	21
Ni(50)/MgO	10%CO ₂ /Ar	5%H ₂ /Ar	0.34	250	21
Ni(50)/MgO	10%CO ₂ /Ar	5%H ₂ /Ar	1.23	300	21
Ni(50)/MgO	10%CO ₂ /Ar	5%H ₂ /Ar	1.06	350	21

Ni(80)/MgO	10%CO ₂ /Ar	5%H ₂ /Ar	0.45	250	21
Ni(80)/MgO	10%CO ₂ /Ar	5%H ₂ /Ar	1.51	300	21
Ni(80)/MgO	10%CO ₂ /Ar	5%H ₂ /Ar	1.29	350	21
Com-Ni(50)/MgO	10%CO ₂ /Ar	5%H ₂ /Ar	0.017	250	21
Com-Ni(50)/MgO	10%CO ₂ /Ar	5%H ₂ /Ar	0.45	300	21
Com-Ni(50)/MgO	10%CO ₂ /Ar	5%H ₂ /Ar	0.039	350	21
Ni/CaO	10%CO ₂ /N ₂	10%H ₂ /N ₂	0.67	400	22
Ni/CaO-MgO	10%CO ₂ /N ₂	10%H ₂ /N ₂	0.69	400	22
Ni/CaO-MgO	10%CO ₂ /N ₂	10%H ₂ /N ₂	0.24	400	22
Ni/MgO	10%CO ₂ /N ₂	10%H ₂ /N ₂	0.15	400	22
AMS-Ni/MgO	65%CO ₂ /N ₂	20%H ₂ /N ₂	0.040	450	23
AMS-Ni/MgO	65%CO ₂ /N ₂	60%H ₂ /N ₂	0.052	450	23
AMS-Ni/MgO	65%CO ₂ /N ₂	100%H ₂	0.046	450	23
AMS-Ni/MgO	65%CO ₂ /N ₂	100%H ₂	0.034	400	23
AMS-Ni/MgO	65%CO ₂ /N ₂	100%H ₂	0.044	500	23
Ru(0.84)-Ba(16)/Al ₂ O ₃	1%CO ₂ /He	4%H ₂ /He	2.06	350	24
Ru(1)/Al ₂ O ₃ + Ba(16)/Al ₂ O ₃	1%CO ₂ /He	4%H ₂ /He	1.32	350	24
Ru(1)/Al ₂ O ₃	1%CO ₂ /He	4%H ₂ /He	0.027	350	3
Ru(0.99)-Li(1)/Al ₂ O ₃	1%CO ₂ /He	4%H ₂ /He	0.027	350	3
Ru(0.97)-Na(3)/Al ₂ O ₃	1%CO ₂ /He	4%H ₂ /He	0.63	350	3
Ru(0.95)-K(5)/Al ₂ O ₃	1%CO ₂ /He	4%H ₂ /He	2.53	350	3
Ru(0.97)-Mg(3.2)/Al ₂ O ₃	1%CO ₂ /He	4%H ₂ /He	0.054	350	3
Ru(0.95)-Ca(5.1)/Al ₂ O ₃	1%CO ₂ /He	4%H ₂ /He	1.85	350	3
Ru(0.84)-Ba(16)/Al ₂ O ₃	1%CO ₂ /He	4%H ₂ /He	3.02	350	3
Ru/rod-CeO ₂ -MgO	35%CO ₂ /N ₂	5%H ₂ /N ₂	0.22	300	25
Ru/particle-CeO ₂ -MgO	35%CO ₂ /N ₂	5%H ₂ /N ₂	0.24	300	25
Ru/cube-CeO ₂ -MgO	35%CO ₂ /N ₂	5%H ₂ /N ₂	0.032	300	25
Ni-Na ₂ CO ₃ /Al ₂ O ₃	9.5%CO ₂ /N ₂	10%H ₂ /N ₂	1.12	320	26
Ni-CaO/Al ₂ O ₃	9.5%CO ₂ /N ₂	10%H ₂ /N ₂	0.88	320	26
Li ₄ SiO ₄ @Ni(2.5)/CeO ₂	15%CO ₂ /N ₂	100%H ₂	9.16	560	27
Li ₄ SiO ₄ @Ni(5)/CeO ₂	15%CO ₂ /N ₂	100%H ₂	9.96	560	27
Li ₄ SiO ₄ @Ni(7.5)/CeO ₂	15%CO ₂ /N ₂	100%H ₂	9.56	560	27
Na-Ni/Al ₂ O ₃	5%CO ₂ /Ar	5%H ₂ /Ar	0.16	250	28
K-Ni/Al ₂ O ₃	5%CO ₂ /Ar	5%H ₂ /Ar	0.17	250	28
Ba-Ni/Al ₂ O ₃	5%CO ₂ /Ar	5%H ₂ /Ar	0.084	250	28
Na-Ni/Al ₂ O ₃	5%CO ₂ /Ar	5%H ₂ /Ar	1.87	300	28
K-Ni/Al ₂ O ₃	5%CO ₂ /Ar	5%H ₂ /Ar	2.43	300	28

Ba-Ni/Al ₂ O ₃	5%CO ₂ /Ar	5%H ₂ /Ar	0.47	300	28
Ni/Al ₂ O ₃	5%CO ₂ /Ar	5%H ₂ /Ar	0.47	300	28
Na-Ni/Al ₂ O ₃	5%CO ₂ /Ar	5%H ₂ /Ar	3.17	350	28
K-Ni/Al ₂ O ₃	5%CO ₂ /Ar	5%H ₂ /Ar	1.49	350	28
Ba-Ni/Al ₂ O ₃	5%CO ₂ /Ar	5%H ₂ /Ar	0.93	350	28
Na-Ni/Al ₂ O ₃	5%CO ₂ /Ar	5%H ₂ /Ar	2.61	400	28
K-Ni/Al ₂ O ₃	5%CO ₂ /Ar	5%H ₂ /Ar	1.59	400	28
Ba-Ni/Al ₂ O ₃	5%CO ₂ /Ar	5%H ₂ /Ar	0.75	400	28
Na-Ni/Al ₂ O ₃	5%CO ₂ /Ar	5%H ₂ /Ar	2.89	450	28
K-Ni/Al ₂ O ₃	5%CO ₂ /Ar	5%H ₂ /Ar	2.29	450	28
Ba-Ni/Al ₂ O ₃	5%CO ₂ /Ar	5%H ₂ /Ar	0.65	450	28
Ru(0.89)-Li(5)/Al ₂ O ₃	10%CO ₂ /N ₂	10%H ₂ /N ₂	2.02	280	29
Ru(3)-K(10)/Al ₂ O ₃	5%CO ₂ /Ar	5%H ₂ /Ar	4.85	300	29
Ru(3)-K(10)/Al ₂ O ₃	5%CO ₂ /Ar	5%H ₂ /Ar	0.93	350	29
Ru(3)-K(10)/Al ₂ O ₃	5%CO ₂ /Ar	5%H ₂ /Ar	7.09	400	29
Ru(3)-K(10)/Al ₂ O ₃	5%CO ₂ /Ar	5%H ₂ /Ar	4.57	450	29
Ru(3)-Na(10)/Al ₂ O ₃	5%CO ₂ /Ar	5%H ₂ /Ar	10.4	300	29
Ru(3)-Na(10)/Al ₂ O ₃	5%CO ₂ /Ar	5%H ₂ /Ar	9.15	350	29
Ru(3)-Na(10)/Al ₂ O ₃	5%CO ₂ /Ar	5%H ₂ /Ar	7.65	400	29
Ru(3)-Na(10)/Al ₂ O ₃	5%CO ₂ /Ar	5%H ₂ /Ar	3.45	450	29
Ru(3)-Ba(10)/Al ₂ O ₃	5%CO ₂ /Ar	5%H ₂ /Ar	1.49	250	29
Ru(3)-Ba(10)/Al ₂ O ₃	5%CO ₂ /Ar	5%H ₂ /Ar	6.35	300	29
Ru(3)-Ba(10)/Al ₂ O ₃	5%CO ₂ /Ar	5%H ₂ /Ar	4.85	350	29
Ru(3)-Ba(10)/Al ₂ O ₃	5%CO ₂ /Ar	5%H ₂ /Ar	2.05	400	29
Ru(3)-Ba(10)/Al ₂ O ₃	5%CO ₂ /Ar	5%H ₂ /Ar	2.61	450	29
Ru(1)-Na(20)/Al ₂ O ₃	10%CO ₂ /N ₂	5%H ₂ /N ₂	1.53	340	30
Ru(0.5)-Na ₂ O(6.1)/Al ₂ O ₃	7.5%CO ₂ /N ₂	15%H ₂ /N ₂	9.71	320	31
Ru(0.5)-Na ₂ O(6.1)/Al ₂ O ₃	7.5%CO ₂ + 15%H ₂ O/N ₂	15%H ₂ /N ₂	7.47	320	31
Ni/Hydrotalcite	15%CO ₂ /N ₂	100%H ₂	3.88	400	32
Ni/Hydrotalcite	15%CO ₂ /N ₂	100%H ₂	4.85	450	32
Ni/Hydrotalcite	15%CO ₂ /N ₂	100%H ₂	4.78	500	32
Ni/Hydrotalcite	15%CO ₂ /N ₂	100%H ₂	3.96	550	32
Ni/Hydrotalcite	15%CO ₂ /N ₂	100%H ₂	3.58	600	32
Ni-Cs(10)/Hydrotalcite	15%CO ₂ /N ₂	100%H ₂	4.93	350	32
Ni(1)/CaO	15%CO ₂ /N ₂	100%H ₂	2.99	550	33
Ni(10)/CaO	15%CO ₂ /N ₂	100%H ₂	3.73	550	33
Ni(1)/CeCaO	15%CO ₂ /N ₂	100%H ₂	4.93	550	33

Ni(1)/CeCaCO ₃	15%CO ₂ /N ₂	100%H ₂	8.96	550	33
Ni(1)/CeO ₂ + CaO	15%CO ₂ /N ₂	100%H ₂	11.95	550	33
LaNiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	1.16	280	34
LaNiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	1.77	320	34
LaNiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	1.87	360	34
LaNiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	2.02	400	34
LaNiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	2.05	440	34
LaNiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	2.00	480	34
LaNiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	1.92	520	34
La _{0.7} Ca _{0.3} NiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	1.4	280	34
La _{0.7} Ca _{0.3} NiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	1.87	320	34
La _{0.7} Ca _{0.3} NiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	2.24	360	34
La _{0.7} Ca _{0.3} NiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	2.39	400	34
La _{0.7} Ca _{0.3} NiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	2.61	440	34
La _{0.7} Ca _{0.3} NiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	2.8	480	34
La _{0.7} Ca _{0.3} NiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	2.37	520	34
La _{0.7} Ba _{0.3} NiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	1.16	280	34
La _{0.7} Ba _{0.3} NiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	1.77	320	34
La _{0.7} Ba _{0.3} NiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	2.02	360	34
La _{0.7} Ba _{0.3} NiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	2.05	400	34
La _{0.7} Ba _{0.3} NiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	1.98	440	34
La _{0.7} Ba _{0.3} NiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	1.59	480	34
La _{0.7} Ba _{0.3} NiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	1.4	520	34
La _{0.7} Na _{0.3} NiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	1.08	280	34
La _{0.7} Na _{0.3} NiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	1.21	320	34
La _{0.7} Na _{0.3} NiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	1.30	360	34
La _{0.7} Na _{0.3} NiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	1.16	400	34
La _{0.7} Na _{0.3} NiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	1.10	440	34
La _{0.7} Na _{0.3} NiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	1.03	480	34
La _{0.7} Na _{0.3} NiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	0.75	520	34
La _{0.7} K _{0.3} NiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	0.65	280	34
La _{0.7} K _{0.3} NiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	0.896	320	34
La _{0.7} K _{0.3} NiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	0.93	360	34
La _{0.7} K _{0.3} NiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	0.93	400	34
La _{0.7} K _{0.3} NiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	0.91	440	34
La _{0.7} K _{0.3} NiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	0.90	480	34
La _{0.7} K _{0.3} NiO ₃ (20)/CeO ₂	10%CO ₂ /Ar	5%H ₂ /Ar	0.84	520	34

Ru/CeO ₂ -CaCO ₃	20%CO ₂ /N ₂	100%H ₂	1.79	350	³⁵
Ru/CeO ₂ -KNO ₃ CaCO ₃	20%CO ₂ /N ₂	100%H ₂	2.52	350	³⁵
Ru/CeO ₂ -LiNO ₃ CaCO ₃	20%CO ₂ /N ₂	100%H ₂	1.456	350	³⁵
Ru/CeO ₂ -(Li-K)NO ₃ CaCO ₃	20%CO ₂ /N ₂	100%H ₂	2.184	350	³⁵
Ru/CeO ₂ -KNO ₃ CaCO ₃	20%CO ₂ /N ₂	100%H ₂	3.192	400	³⁵
Ru/CeO ₂ -KNO ₃ CaCO ₃	20%CO ₂ /N ₂	100%H ₂	5.32	450	³⁵
Ru/CeO ₂ -KNO ₃ CaCO ₃	20%CO ₂ /N ₂	100%H ₂	3.81	500	³⁵
Ru(0.25)-Na/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	0.72	300	³⁶
Ru(0.5)-Na/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	2.27	300	³⁶
Ru(1)-Na/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	2.688	300	³⁶
Ru(2)-Na/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	2.68	300	³⁶
Ru(4)-Na/Al ₂ O ₃	10%CO ₂ /Ar	10%H ₂ /Ar	2.66	300	³⁶
Ni-Pr/CeO ₂	10%CO ₂ /Ar	10%H ₂ /Ar	4.66	300	¹³
RuNi-Pr/CeO ₂	10%CO ₂ /Ar	10%H ₂ /Ar	5.62	300	¹³
Ni/MgO	65%CO ₂ /N ₂	50%H ₂ /N ₂	0.046	500	³⁷

‡ using a circulating fluidized system similar to Fig. 2b

Table S3. Summary of the continuous CO₂ capture and RWGS reaction in this study and other reported CO₂ capture and RWGS reaction considering the effect of coexistent O₂. Entries without *, †, or ‡ symbols using Fig. 1a system.

DFMs or catalyst	CO ₂ capture gas	Hydrogenation gas	CO yield [%]	T [°C]	ref
Cu/ZnO/Al ₂ O ₃ *	10%CO ₂ +10%O ₂ /He	100%H ₂	85.1	650	*
Pt(1)-Na(3)/Al ₂ O ₃	1%CO ₂ +10%O ₂ /N ₂	5%H ₂ /N ₂	8.8	350	38
Pt(1)-Na(3)/Al ₂ O ₃ †	1%CO ₂ +10%O ₂ /N ₂	100% H ₂	89.0	350	38
Pt(1)-Na(3)/MgO	1%CO ₂ +10%O ₂ /N ₂	5%H ₂ /N ₂	3.4	350	38
Pt(1)-Ca(6)/Al ₂ O ₃	1%CO ₂ +10%O ₂ /N ₂	5%H ₂ /N ₂	2.3	350	38
Pt(1)-Mg(3)/Al ₂ O ₃	1%CO ₂ /10%O ₂ /N ₂	5%H ₂ /N ₂	0.56	350	38
Pt(1)-K(6)/Al ₂ O ₃	1%CO ₂ +10%O ₂ /N ₂	5%H ₂ /N ₂	0.39	350	38
Ru(1)-Na(3)/Al ₂ O ₃	1%CO ₂ +10%O ₂ /N ₂	5%H ₂ /N ₂	0.34	350	38
Cu(1)-Na(3)/Al ₂ O ₃	1%CO ₂ +10%O ₂ /N ₂	5%H ₂ /N ₂	0.17	350	38
Pt(1)-Na(3)/SiO ₂	1%CO ₂ +10%O ₂ /N ₂	5%H ₂ /N ₂	0.34	350	38
Pt(1)-Na(3)/TiO ₂	1%CO ₂ +10%O ₂ /N ₂	5%H ₂ /N ₂	1.68	350	38
Rb-Ni/Al ₂ O ₃ †	0.5%CO ₂ +10%O ₂ /N ₂	20%H ₂ /N ₂	47.8	450	39
Pt-Na/Al ₂ O ₃ †	0.5%CO ₂ +10%O ₂ /N ₂	20%H ₂ /N ₂	34.7	450	39
Ni-Rb/Al ₂ O ₃ †	0.5%CO ₂ +10%O ₂ /N ₂	20%H ₂ /N ₂	28.2	450	39
Na-Ni/Al ₂ O ₃ †	0.5%CO ₂ +10%O ₂ /N ₂	20%H ₂ /N ₂	12.9	450	39
Mg-Ni/Al ₂ O ₃ †	0.5%CO ₂ +10%O ₂ /N ₂	20%H ₂ /N ₂	0.54	450	39
Na/Al ₂ O ₃	0.5%CO ₂ +10%O ₂ /N ₂	20%H ₂ /N ₂	15.1	450	39
Rb/Al ₂ O ₃	0.5%CO ₂ +10%O ₂ /N ₂	20%H ₂ /N ₂	16.1	450	39
Fe(6.91)Cr(0.58)Cu(0.20)-K(9.98)/hydrotalcite	5.8%CO ₂ +5%O ₂ +4%H ₂ O/N ₂	100%H ₂	41.3	450	40
Fe(6.91)Cr(0.58)Cu(0.20)-K(9.98)/hydrotalcite	5.8%CO ₂ +5%O ₂ +4%H ₂ O/N ₂	100%H ₂	52.2	500	40
Fe(6.91)Cr(0.58)Cu(0.20)-K(9.98)/hydrotalcite	5.8%CO ₂ +5%O ₂ +4%H ₂ O/N ₂	100%H ₂	56.0	530	40

*This study † using Fig. 2b system.

Table S4. Summary of reported CO₂ capture and RWGS reaction. Entries without *, †, or ‡ symbols using Fig. 1a system.

DFMs or catalyst	CO ₂ capture gas	Hydrogenation gas	CO yield. [%]	T [°C]	ref
Ca ₁ Ni _{0.1} O	15%CO ₂ /N ₂	5%H ₂ /N ₂	10.3	650	41
Ca ₁ Ni _{0.1} Ce _{0.017} O	15%CO ₂ /N ₂	5%H ₂ /N ₂	10.8	650	41
Ca ₁ Ni _{0.1} Ce _{0.033} O	15%CO ₂ /N ₂	5%H ₂ /N ₂	10.9	650	41
Fe(5)Co(5)Mg(10)/CaO	10%CO ₂ /He	100%H ₂	30.9	650	42
CaO	10%CO ₂ /He	100%H ₂	10.3	650	42
Fe(10)Mg(10)/CaO	10%CO ₂ /He	100%H ₂	26.9	650	42
Fe(8)Co(2)Mg(10)CaO	10%CO ₂ /He	100%H ₂	26.5	650	42
Fe(7.5)Co(2.5)Mg(10)CaO	10%CO ₂ /He	100%H ₂	27.7	650	42
Fe(6.7)Co(3.3)Mg(10)CaO	10%CO ₂ /He	100%H ₂	29.7	650	42
Fe(3.3)Co(6.7)Mg(10)CaO	10%CO ₂ /He	100%H ₂	26.0	650	42
Co(10)Mg(10)CaO	10%CO ₂ /He	100%H ₂	24.4	650	42
Ni(10)/CaO	10%CO ₂ /N ₂	5%H ₂ /N ₂	10.6	650	43
Ni(10)/Carbide slag(CS)	10%CO ₂ /N ₂	5%H ₂ /N ₂	10.6	650	43
Rb-Ni/Al ₂ O ₃	0.5%CO ₂ /N ₂	20%H ₂ /N ₂	22.0	450	39
Ni(10)/CaZr(O)	15%CO ₂ /N ₂	66.7%H ₂ /N ₂	4.80	600	44
Ni(10)/CaAl(O)	15%CO ₂ /N ₂	66.7%H ₂ /N ₂	6.95	600	44
Ni(10)/CaO	15%CO ₂ /N ₂	66.7%H ₂ /N ₂	6.09	600	44
Ni(10)/CaMg(O)	15%CO ₂ /N ₂	66.7%H ₂ /N ₂	5.37	600	44
Cu(11)-K(10)/Al ₂ O ₃	4.4%CO ₂ /He	100%H ₂	27.6	450	45
Fe(6.91)Cr(0.58)Cu(0.20)-K(9.98)/hydrotalcite	5.8%CO ₂ /N ₂	100%H ₂	72.7	550	40
Fe(6.91)Cr(0.58)Cu(0.20)-K(9.98)/hydrotalcite	5.8%CO ₂ +4%H ₂ O/N ₂	100%H ₂	64.3	550	40
Fe(6.91)Cr(0.58)Cu(0.20)-K(9.98)/hydrotalcite	5.8%CO ₂ /4%O ₂ /N ₂	100%H ₂	50.8	550	40
Fe(6.91)Cr(0.58)Cu(0.20)-K(9.98)/hydrotalcite	7.6%CO ₂ /N ₂	100%H ₂	54.7	550	40
Fe(6.91)Cr(0.58)Cu(0.20)-K(9.98)/hydrotalcite	7.6%CO ₂ /4%H ₂ O/N ₂	100%H ₂	44.9	550	40
Fe(6.91)Cr(0.58)Cu(0.20)-K(9.98)/hydrotalcite	9.5%CO ₂ /N ₂	100%H ₂	38.7	550	40
Fe(6.91)Cr(0.58)Cu(0.20)-K(9.98)/hydrotalcite	9.5%CO ₂ /4%H ₂ O/N ₂	100%H ₂	32.4	550	40
Fe(6.91)Cr(0.58)Cu(0.20)-K(9.98)/hydrotalcite	5.8%CO ₂ /N ₂	100%H ₂	72.3	450	40
Fe(6.91)Cr(0.58)Cu(0.20)-K(9.98)/hydrotalcite	5.8%CO ₂ /N ₂	100%H ₂	72.7	470	40
Fe(6.91)Cr(0.58)Cu(0.20)-K(9.98)/hydrotalcite	5.8%CO ₂ /N ₂	100%H ₂	78.5	500	40
Fe(6.91)Cr(0.58)Cu(0.20)-K(9.98)/hydrotalcite	5.8%CO ₂ /N ₂	100%H ₂	83.1	530	40
Na(16)/Al ₂ O ₃	5%CO ₂ /N ₂	100%H ₂	5.08	450	46

K(21)/Al ₂ O ₃	5%CO ₂ /N ₂	100%H ₂	5.97	450	46
Ca(15)/Al ₂ O ₃	5%CO ₂ /N ₂	100%H ₂	2.7	450	46
Al ₂ O ₃	5%CO ₂ /N ₂	100%H ₂	0.30	450	46
Na(16)/Al ₂ O ₃	5%CO ₂ /N ₂	100%H ₂	2.33	350	46
Na(16)/Al ₂ O ₃	5%CO ₂ /N ₂	100%H ₂	4.928	400	46
Na(16)/Al ₂ O ₃	5%CO ₂ /N ₂	100%H ₂	5.36	500	46
CaO	15%CO ₂ /N ₂	15%H ₂	1.58	600	47
CaO	15%CO ₂ /N ₂	15%H ₂	7.1	650	47
CaO	15%CO ₂ /N ₂	15%H ₂	19.3	700	47
CeO ₂ (33)/CaO	17%CO ₂ /N ₂	5%H ₂ /N ₂	2.16	650	47
CeO ₂ (33)/CaO	17%CO ₂ /N ₂	5%H ₂ /N ₂	0.98	600	47
CeO ₂ (33)/CaO	17%CO ₂ /N ₂	5%H ₂ /N ₂	3.63	700	47
CeO ₂ (33)/CaO	17%CO ₂ /N ₂	5%H ₂ /N ₂	1.81	750	47
CeO ₂ (10)/CaO	17%CO ₂ /N ₂	5%H ₂ /N ₂	0.84	600	47
CeO ₂ (10)/CaO	17%CO ₂ /N ₂	5%H ₂ /N ₂	3.56	650	47
CeO ₂ (10)/CaO	17%CO ₂ /N ₂	5%H ₂ /N ₂	4.88	700	47
CeO ₂ (10)/CaO	17%CO ₂ /N ₂	5%H ₂ /N ₂	2.23	750	47
CeO ₂ (16)/CaO	17%CO ₂ /N ₂	5%H ₂ /N ₂	0.70	600	47
CeO ₂ (16)/CaO	17%CO ₂ /N ₂	5%H ₂ /N ₂	2.51	650	47
CeO ₂ (16)/CaO	17%CO ₂ /N ₂	5%H ₂ /N ₂	4.74	700	47
CeO ₂ (16)/CaO	17%CO ₂ /N ₂	5%H ₂ /N ₂	2.65	750	47
CeO ₂ (50)/CaO	17%CO ₂ /N ₂	5%H ₂ /N ₂	0.56	600	47
CeO ₂ (50)/CaO	17%CO ₂ /N ₂	5%H ₂ /N ₂	2.23	650	47
CeO ₂ (50)/CaO	17%CO ₂ /N ₂	5%H ₂ /N ₂	2.93	700	47
CeO ₂ (50)/CaO	17%CO ₂ /N ₂	5%H ₂ /N ₂	0.84	750	47
CeO ₂ (67)/CaO	17%CO ₂ /N ₂	5%H ₂ /N ₂	0.35	600	47
CeO ₂ (67)/CaO	17%CO ₂ /N ₂	5%H ₂ /N ₂	1.39	650	47
CeO ₂ (67)/CaO	17%CO ₂ /N ₂	5%H ₂ /N ₂	1.39	700	47
CeO ₂ (67)/CaO	17%CO ₂ /N ₂	5%H ₂ /N ₂	0.56	750	47
Ni/CaO	10%CO ₂ /10%H ₂ O/N ₂	10%H ₂ /N ₂	21.5	700	48
La(15)-Ni(2.5)/CaO	10%CO ₂ /Ar	10%H ₂ /Ar	43.3	650	49
Mg(15)-Ni(2.5)/CaO	10%CO ₂ /Ar	10%H ₂ /Ar	38.8	650	49
Zr(15)-Ni(2.5)/CaO	10%CO ₂ /Ar	10%H ₂ /Ar	36.3	650	49
Ce(15)-Ni(2.5)/CaO	10%CO ₂ /Ar	10%H ₂ /Ar	41.3	650	49
Ni(2.5)/CaO	10%CO ₂ /Ar	10%H ₂ /Ar	31.36	650	49

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