Design Rules for Ternary CO_2 Hydrogenation Catalysts via Literature-Sourced Network Construction and Analysis

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K-means clusters

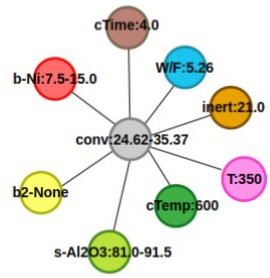
Base wt%	0.0-7.0%, 7.5-15.0%, 17.0-26.0%, 27.5-40.0% , 47.0-100.0%
Base 2 wt%	0.0-3.0%, 4.0-10.0%, 12.5-17.0%, 25.0-37.5%
Support wt%	0.0-60.7%, 62.5-80.0%, 81.0-91.5%, 92.0-100.0%
CO_2 Conversion %	0.0-5.61%, 5.63-14.15%, 14.19-24.52%, 24.62-35.37%, 35.4-46.03%, 46.08-56.88%, 57.0-67.53%, 67.59-77.5%, 77.52-88.38%, 88.41-100.0%

Knowledge network built from the literature

features	Value
Base	Ni
Base wt%	10.0
Base 2	None
Base 2 wt%	0.0
Support	Al_2O_3
Support wt %	90.0
Calcination Temperature (C)	600
Calcination time (h)	4.0
Temperature (C)	350
W/F	5.26
Inert in feed %	21.0
CO_2 Conversion %	25.552

features	clusters
Base	Ni
Base wt%	7.5-15.0%
Base 2	None
Base 2 wt%	0.0-3.0%
Support	Al_2O_3
Support wt %	81.0-91.5%
Calcination Temperature (C)	600
Calcination time (h)	4.0
Temperature (C)	350
W/F	5.26
Inert in feed %	21.0
CO_2 Conversion %	24.62-35.37%

Nodes	Labels
Base	b-Ni:7.5-15.0
Base 2	b2-None
Support	s-Al_2O_3:81.0-91.5
Calcination Temperature (C)	cTemp:600
Calcination time (h)	cTime:4.0
Temperature (C)	T:350
W/F	W/F:5.26
Inert in feed %	inert:21.0
CO_2 Conversion %	conv:24.62-35.37



FigureS1 Conversion from literature data to network for N-1. This is an example of network construction for one data(Base: Ni, Base wt%: 10.0, Base 2: None, Base 2 wt%: 0.0, Support: Al_2O_3, Support: wt%: 90.0, Calcination Temperature (C): 600, Calcination time (h): 4.0, Temperature(C): 350, W/F: 5.26, Inert in feed %: 21.0, CO_2 Conversion %: 25.552). The data points transformed by the corresponding clusters for each variable are shown in red.

K-means clusters

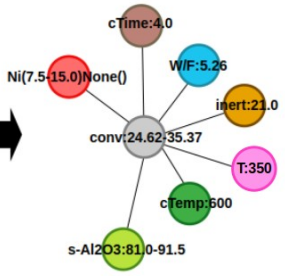
Base wt%	0.0-7.0%, 7.5-15.0%, 17.0-26.0%, 27.5-40.0%, 47.0-100.0%
Base 2 wt%	0.0-3.0%, 4.0-10.0%, 12.5-17.0%, 25.0-37.5%
Support wt%	0.0-60.7%, 62.5-80.0%, 81.0-91.5%, 92.0-100.0%
CO2 Conversion %	0.0-5.61%, 5.63-14.15%, 14.19-24.52%, 24.62-35.37%, 35.4-46.03%, 46.08-56.88%, 57.0-67.53%, 67.59-77.5%, 77.52-88.38%, 88.41-100.0%

Knowledge network built from the literature

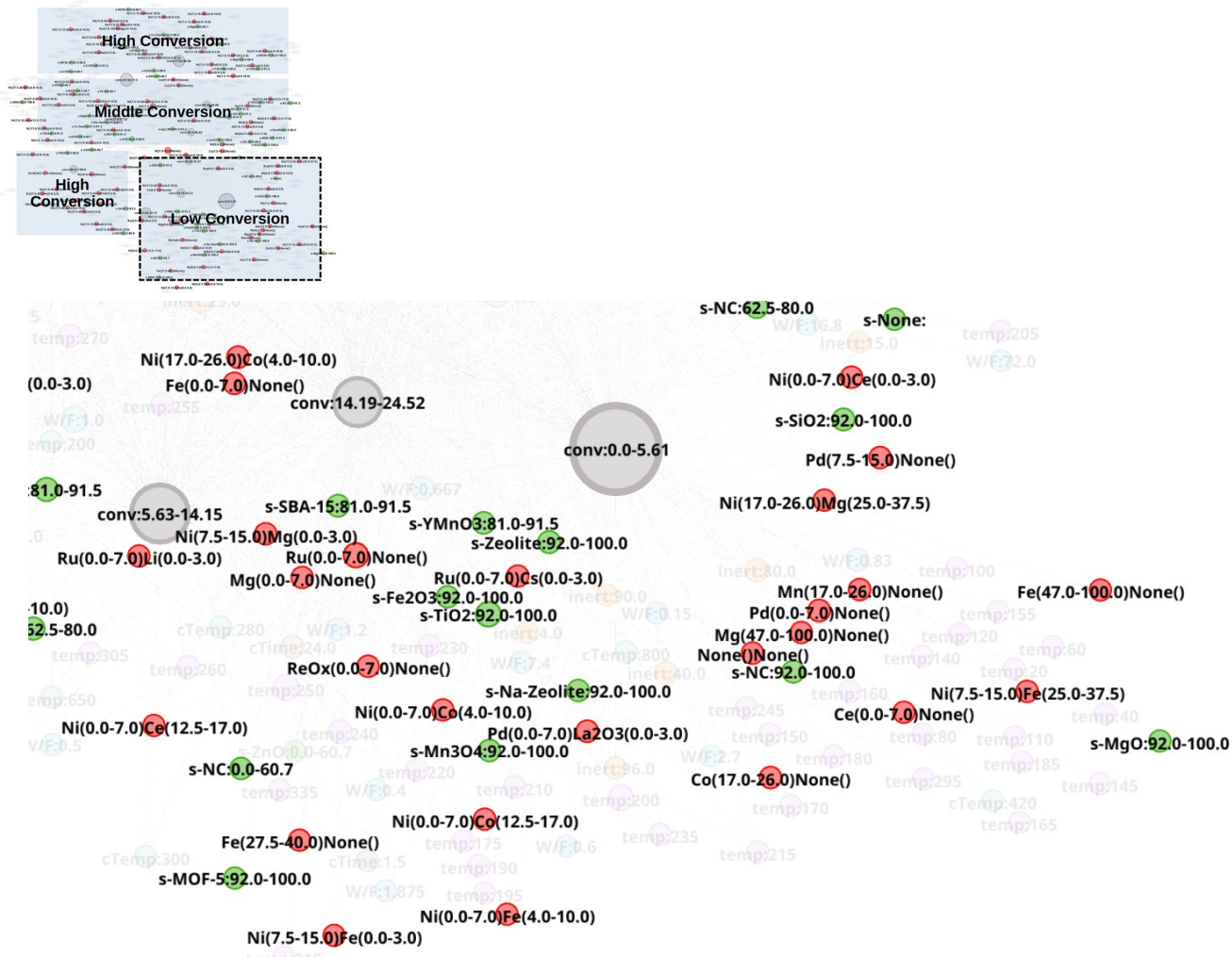
features	Value
Base	Ni
Base wt%	10.0
Base 2	None
Base 2 wt%	0.0
Support	Al_2O_3
Support wt %	90.0
Calcination Temperature (C)	600
Calcination time (h)	4.0
Temperature (C)	350
W/F	5.26
Inert in feed %	21.0
CO_2 Conversion %	25.552

features	clusters
Base	Ni
Base wt%	7.5-15.0%
Base 2	None
Base 2 wt%	0.0-3.0%
Support	Al_2O_3
Support wt %	81.0-91.5%
Calcination Temperature (C)	600
Calcination time (h)	4.0
Temperature (C)	350
W/F	5.26
Inert in feed %	21.0
CO_2 Conversion %	24.62-35.37%

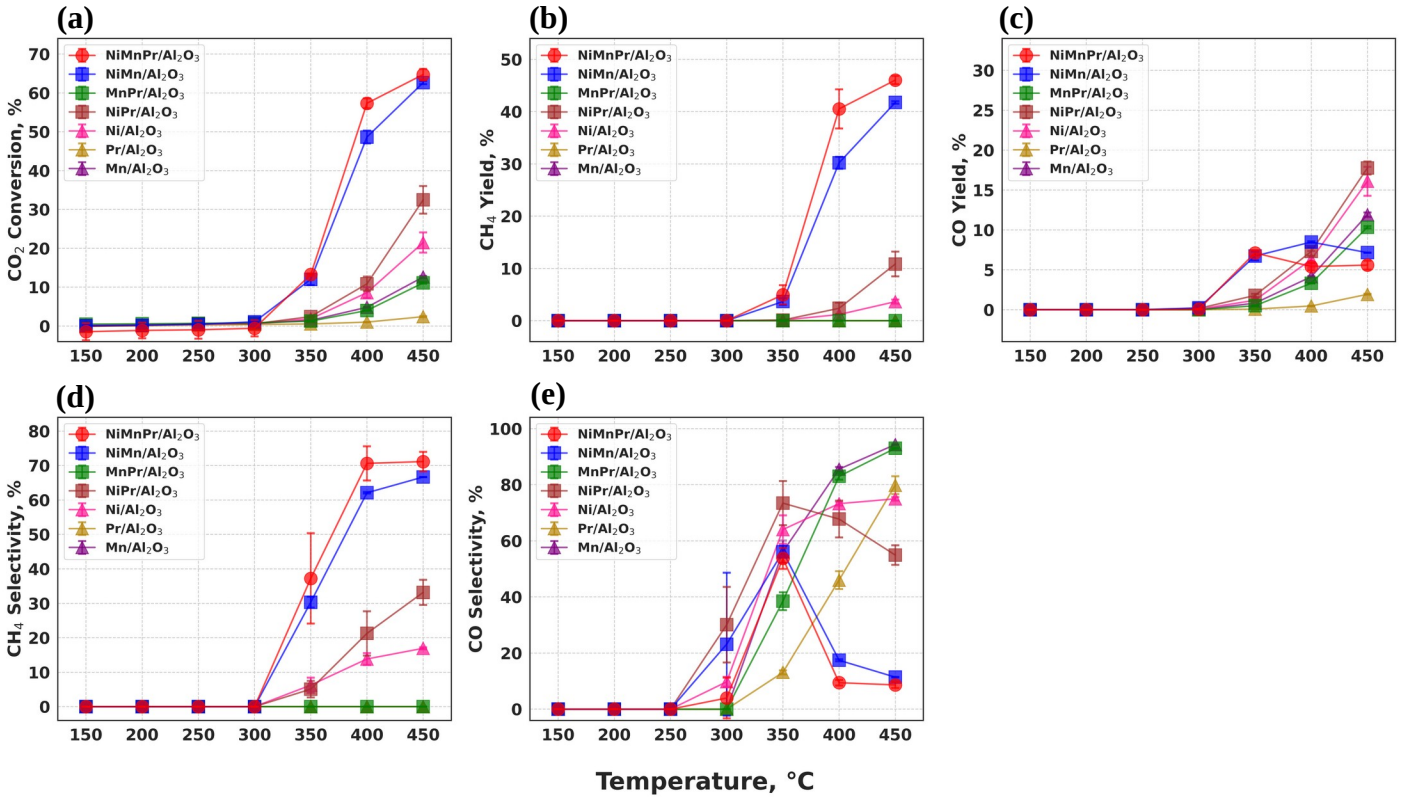
Nodes	Labels
Catalyst	Ni(7.5-15.0) None()
Support	s-Al_2O_3:81.0-91.5
Calcination Temperature (C)	cTemp:600
Calcination time (h)	cTime:4.0
Temperature (C)	T:350
W/F	W/F:5.26
Inert in feed %	inert:21.0
CO_2 Conversion %	conv:24.62-35.37



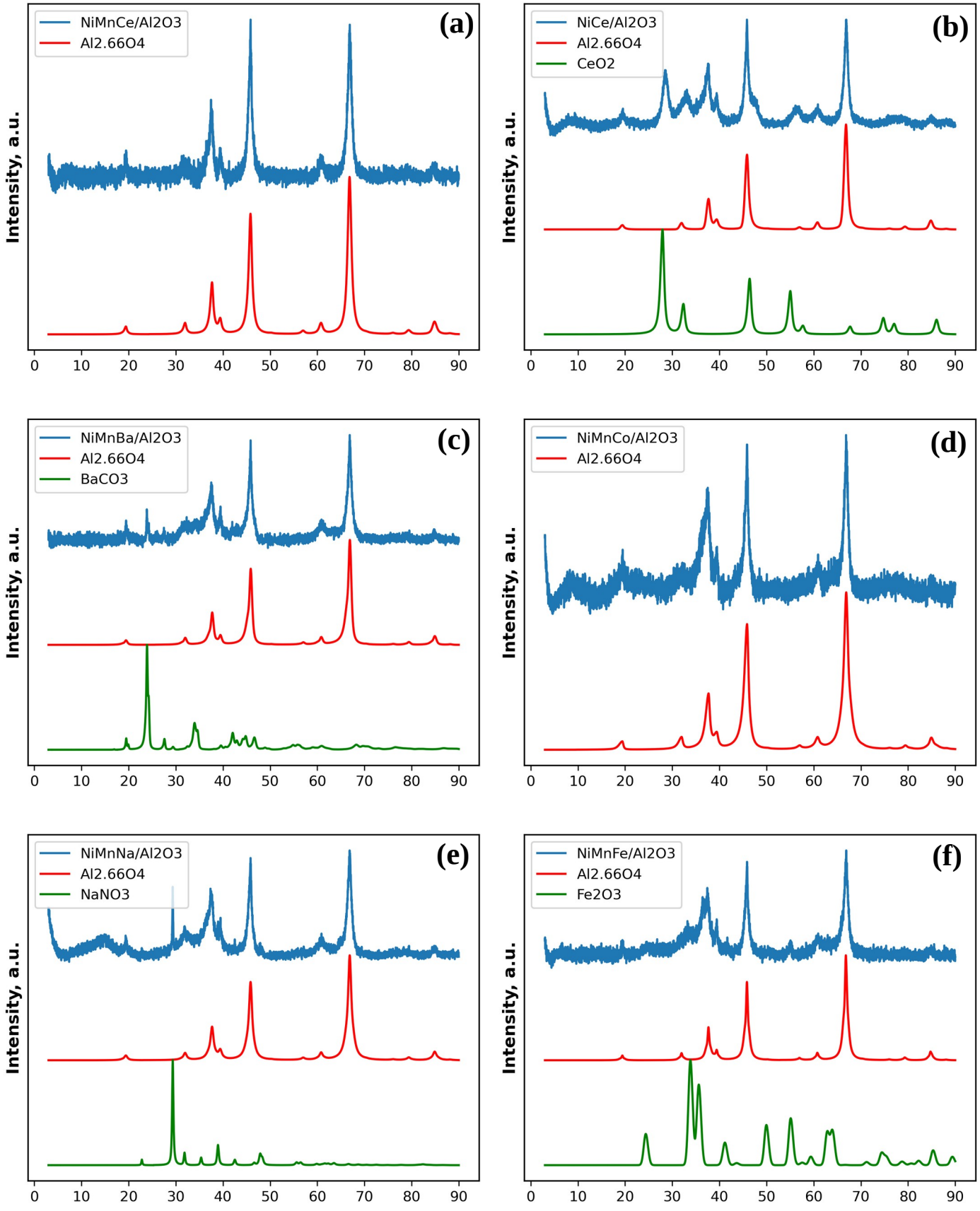
FigureS2 Conversion from literature data to network for N-2. This is an example of network construction for one data(Base: Ni, Base wt%: 10.0, Base 2: None, Base 2 wt%: 0.0, Support: Al_2O_3, Support: wt%: 90.0, Calcination Temperature (C): 600, Calcination time (h): 4.0, Temperature(C): 350, W/F: 5.26, Inert in feed %: 21.0, CO_2 Conversion %: 25.552). The data points transformed by the corresponding clusters for each variable are shown in red.



FigureS3 Details of low CO_2 conversion area in the N-2 network. Catalyst(pink), Support(green), and CO_2 conversion(gray) are highlighted. Note that labels are adjusted, and node size is determined by degree ranking within a range of 10 to 40 for visualization purposes.

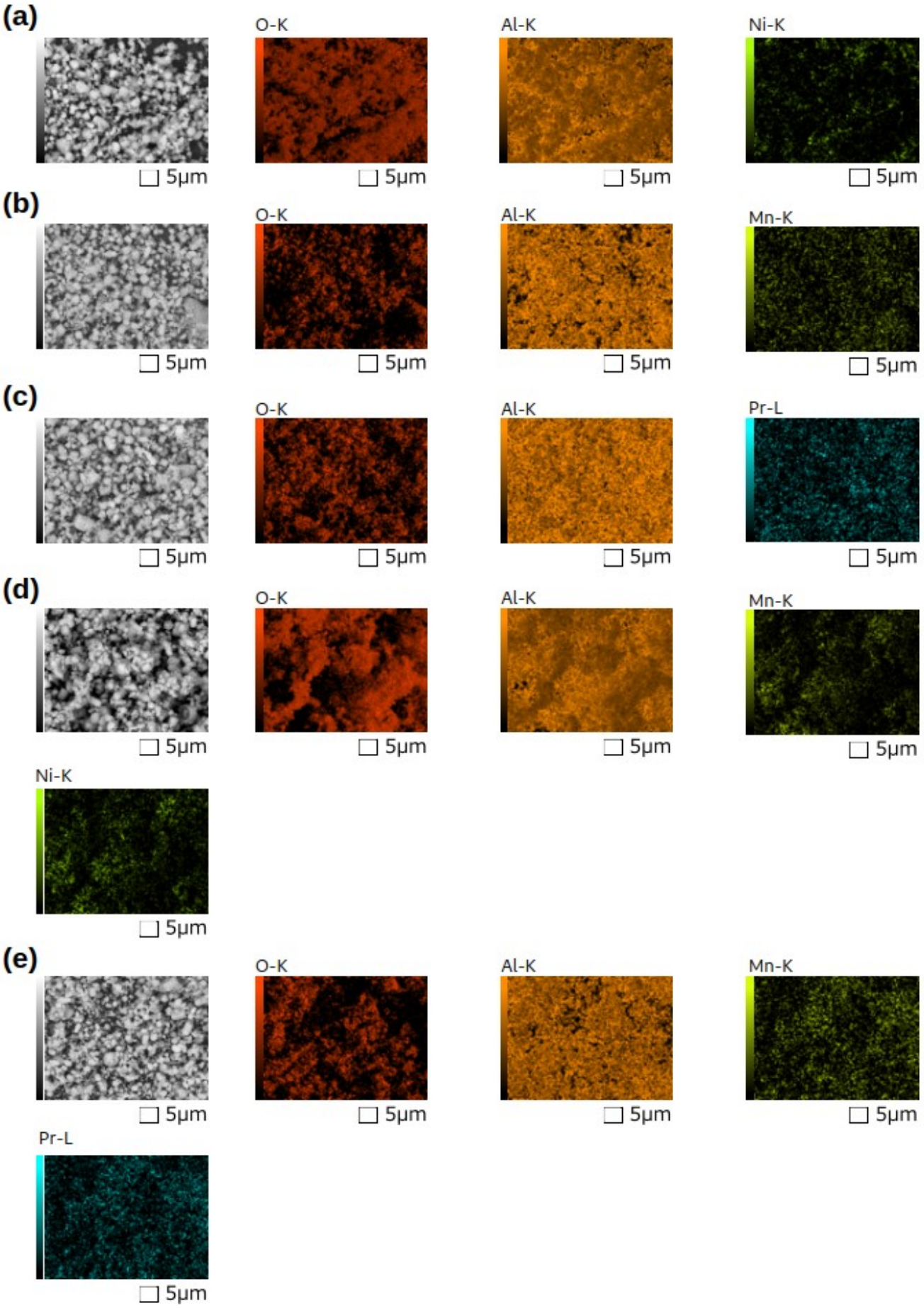


FigureS4 (a)CO_2 conversion, %, (b)CH_4 yield, %, (c)CO yield, %, (d)CH_4 selectivity, %, and (e)CO selectivity, % as a function of temperature for catalysts including Pr. Ternary, binary, and unary catalysts are represented by circles, squares, and triangles, respectively. Note that the plots and error bars at each temperature represent the average and standard deviation of two measurements to ensure reproducibility.

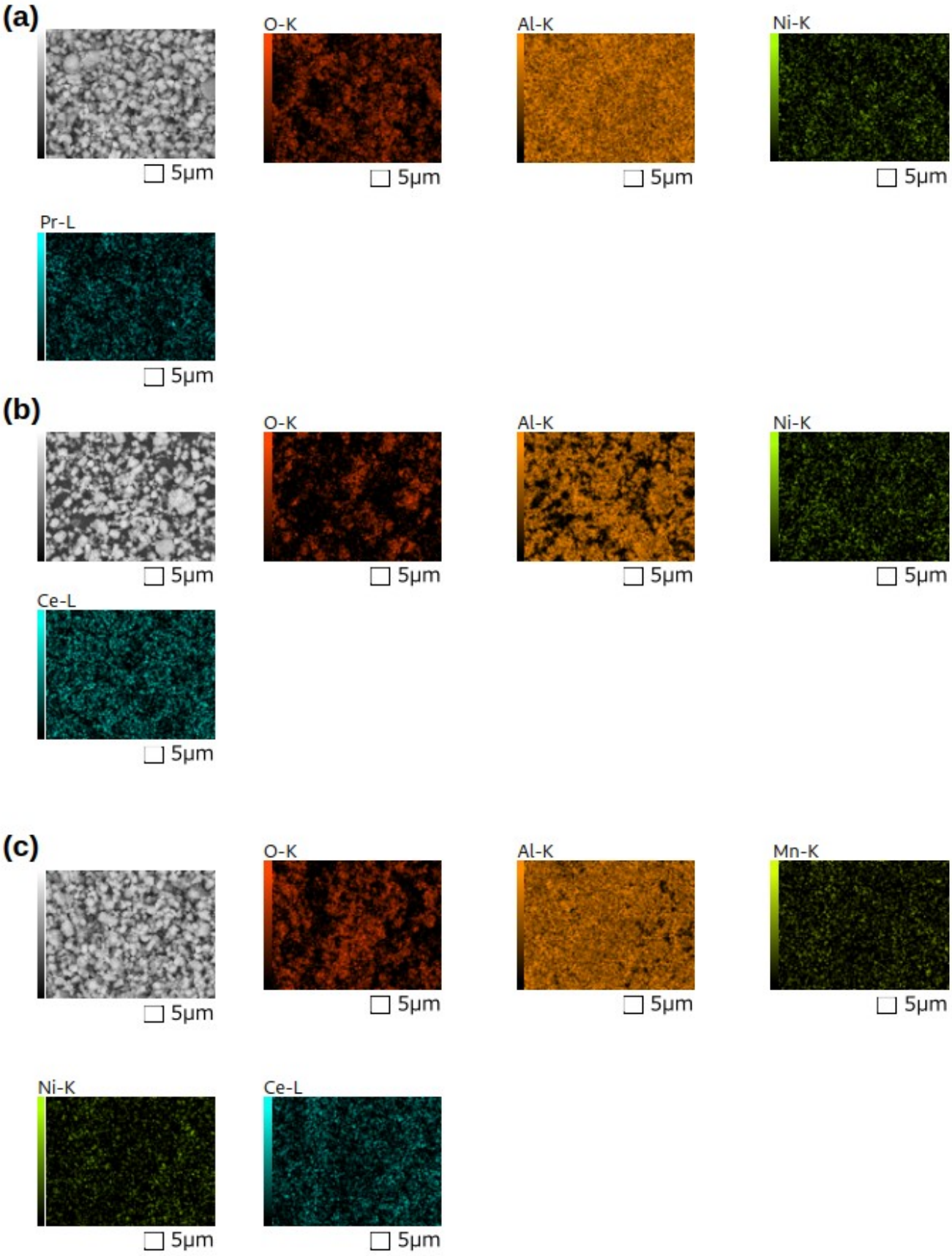


FigureS5 XRD for (a)NiMnCe/Al_2O_3, (b)NiCe/Al_2O_3, (c)NiMnBa/Al_2O_3, (d)NiMnCo/Al_2O_3, (e)NiMnNa/Al_2O_3, (f)NiMnFe/Al_2O_3. The reference patterns shown in red and green are obtained from the built-in database of the Rigaku Miniflex 600-C software.

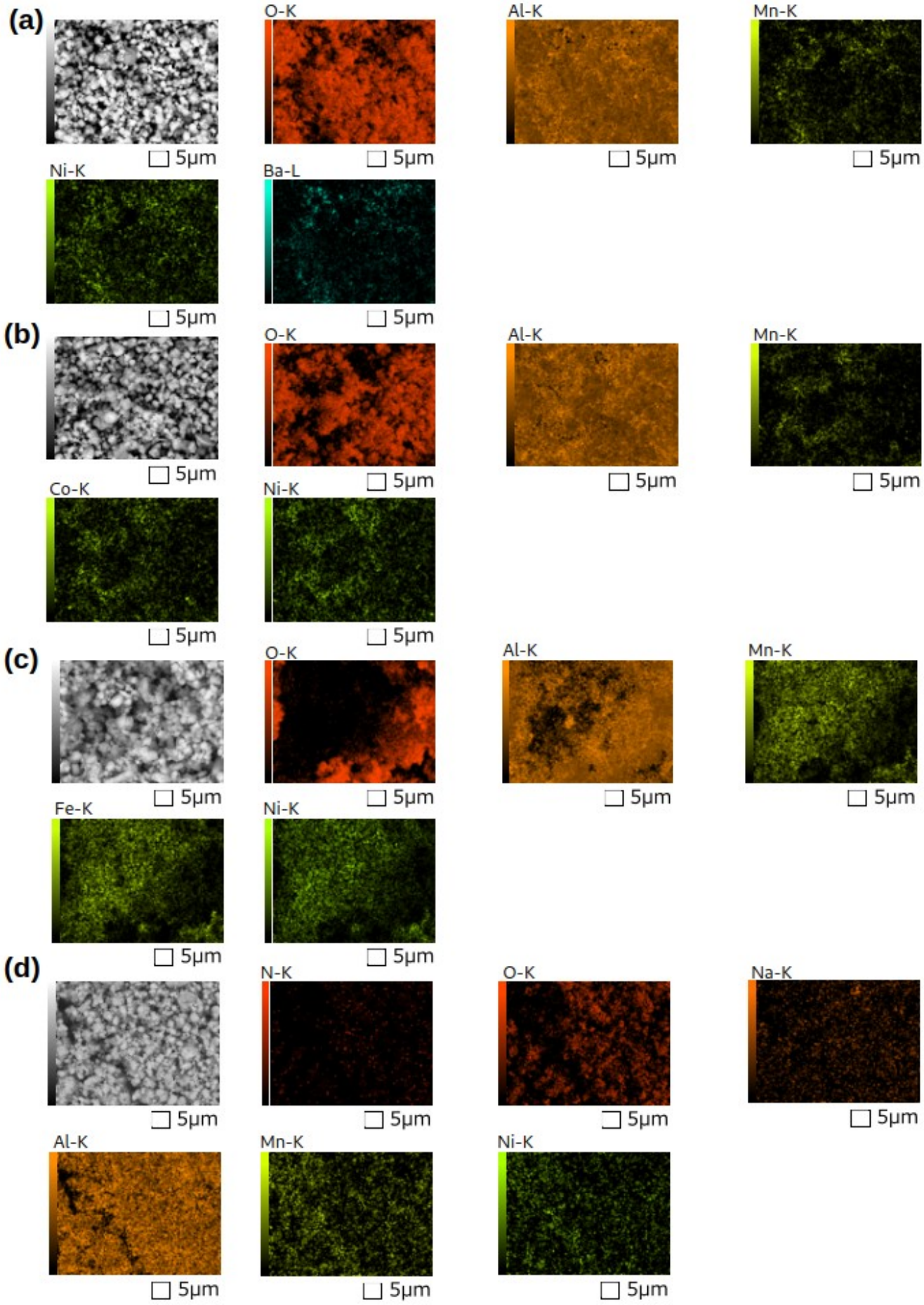
S5



FigureS6 SEM-EDS for (a)Ni/Al_2O_3, (b)Mn/Al_2O_3, (c)Pr/Al_2O_3, (d)NiMn/Al_2O_3, and (e)MnPr/Al_2O_3



FigureS7 SEM-EDS for (a)NiPr/Al_2O_3, (b)NiCe/Al_2O_3, and (c)NiMnCe/Al_2O_3.



FigureS8 SEM-EDS for (a)NiMnBa/Al_2O_3, (b)NiMnCo/Al_2O_3, (c)NiMnFe/Al_2O_3, and (d)NiMnNa/Al_2O_3.

Table S1 Comparison of CO_2 conversion activities reported in the literature at 450 °C with a Ni loading of 5 wt%.

M1	M2	M3	M2 wt%	Support	Preparation Method	Red Temp, ℃	Red Time, h	Inert Gas, %	H_2/CO_2 ratio	CO_2 Conv, %	Ref
Ni	Mn	Pr	5	Al_2O_3	CI	None	None	50	4	64.73	This Work
Ni	Mn	Ce	5	Al_2O_3	CI	None	None	50	4	65.85	This Work
Ni	Mn	Co	5	Al_2O_3	CI	None	None	50	4	51.62	This Work
Ni	Mn	Fe	5	Al_2O_3	CI	None	None	50	4	42.78	This Work
Ni	Mn	Na	5	Al_2O_3	CI	None	None	50	4	48.50	This Work
Ni	Mn	Ba	5	Al_2O_3	CI	None	None	50	4	61.93	This Work
Ni	None	None	5	Al_2O_3	MC	500	2	0	4	6.87	S1
Ni	None	None	10	Al_2O_3	MC	500	2	0	4	59.34	S1
Ni	None	None	10	Al_2O_3	MC	500	2	0	4	22.33	S1
Ni	None	None	0	Al_2O_3	MC	500	2	0	4	53.31	S1
Ni	None	None	0	Al_2O_3	WI	600	1	21	4	46.97	S2
Ni	Ce	None	15	Zeolite	IWI	470	1	18.2	4	60.50	S3
Ni	None	None	0	Al_2O_3	EISA	500	3	15	5	69.16	S4
Ni	None	None	0	Na-Zeolite	WI	470	1	18.2	4	36.3	S5
Ni	Ce	None	3	Na-Zeolite	WI	470	1	18.2	4	55.67	S5
Ni	Ce	None	3	Na-Zeolite	CI	470	1	18.2	4	52.67	S5
Ni	None	None	5	Al_2O_3	IWI	700	2	45.45	5	38.72	S6
Ni	None	None	0	Zeolite	IWI	500	2	16.7	4	57	S7
Ni	None	None	0	CeO_2	WI	550	1	25	4	74.26	S8
Ni	None	None	0	Y_2O_5	WI	550	1	25	4	73.35	S8

Note: Red Temp: Reduction Temperature, Red Time: Reduction Time, WI: Wet Impregnation, MC: Mechano Chemical, IWI:Incipient to wetness impregnation, CI: Co-impregnation

Reference

[S1]Shafiee, Parisa, Seyed Mehdi Alavi, and Mehran Rezaei. "Investigation of the effect of cobalt on the Ni–Al_2O_3 catalyst prepared by the mechanochemical method for CO_2 methanation: P. Shafiee et al." Research on Chemical Intermediates 48.5 (2022): 1923-1938.

[S2]Zhang, Zhanming, et al. "Impacts of nickel loading on properties, catalytic behaviors of Ni/γ–Al_2O_3 catalysts and the reaction intermediates formed in methanation of CO_2." International Journal of Hydrogen Energy 44.18 (2019): 9291-9306.

[S3]Graça, I., et al. "CO_2 hydrogenation into CH_4 on NiHNaUSY zeolites." Applied Catalysis B: Environmental 147 (2014): 101-110.

[S4]Aljishi, Ali, et al. "The effect of synthesis parameters on ordered mesoporous nickel alumina catalyst for CO_2 methanation." Applied Catalysis A: General 549 (2018): 263-272.

[S5]Bacariza, M. Carmen, et al. "Ni-Ce/Zeolites for CO_2 Hydrogenation to CH_4: Effect of the Metal Incorporation Order." ChemCatChem 10.13 (2018): 2773-2781.
[S6]Chein, Rei-Yu, and Chih-Chang Wang. "Experimental study on CO_2 methanation over Ni/Al_2O_3, and $RuNi/Al_2O_3$ catalysts." Catalysts 10.10 (2020): 1112.
[S7]Goodarzi, Farnoosh, et al. "Methanation of Carbon Dioxide over Zeolite-Encapsulated Nickel Nanoparticles." ChemCatChem 10.7 (2018): 1566-1570.
[S8]Sun, Chao, et al. "Ni/CeO2 nanoparticles promoted by yttrium doping as catalysts for CO_2 methanation." ACS Applied Nano Materials 3.12 (2020): 12355-12368.