Electronic Supplementary Material (ESI) for Catalysis Science & Technology.

Supplementary Information

Enhanced Ni-Ce interactions enable efficient low-temperature catalytic CO_2 methanation

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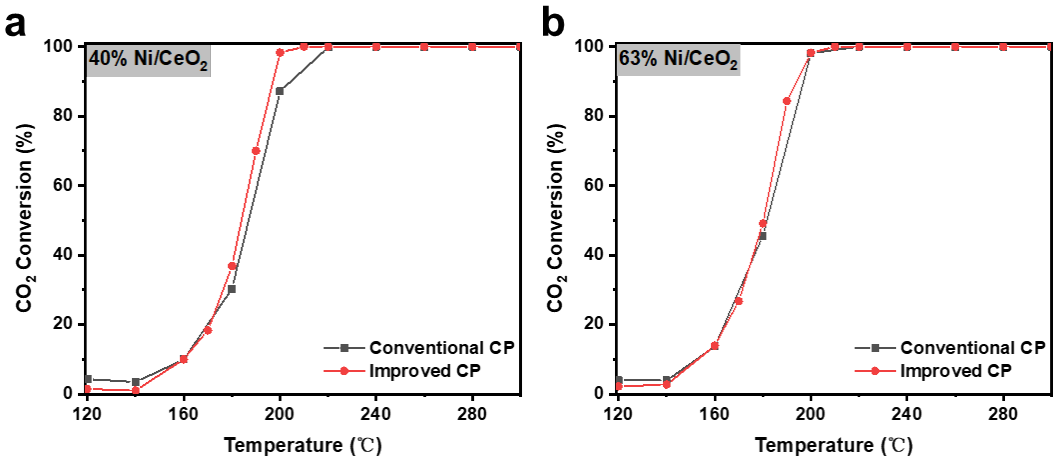

Figure S1: The activity comparison of Ni/CeO_2 catalysts synthesized by the improved and conventional co-precipitation methods a 40% Ni/CeO_2 b 63% Ni/CeO_2

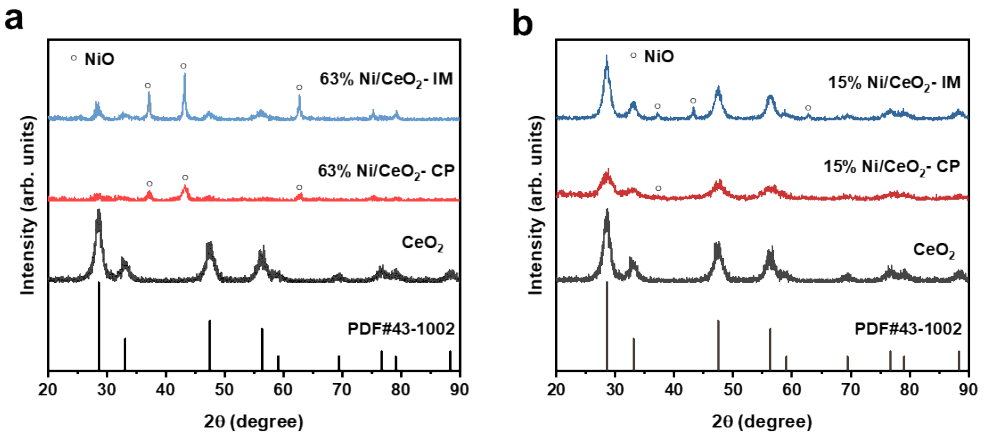

Fig S2: a XRD patterns of the calcined 63% Ni/CeO_2-CP and 63% Ni/CeO_2-IM catalysts, **b** XRD patterns of the calcined 15% Ni/CeO_2-CP and 15% Ni/CeO_2-IM catalysts.

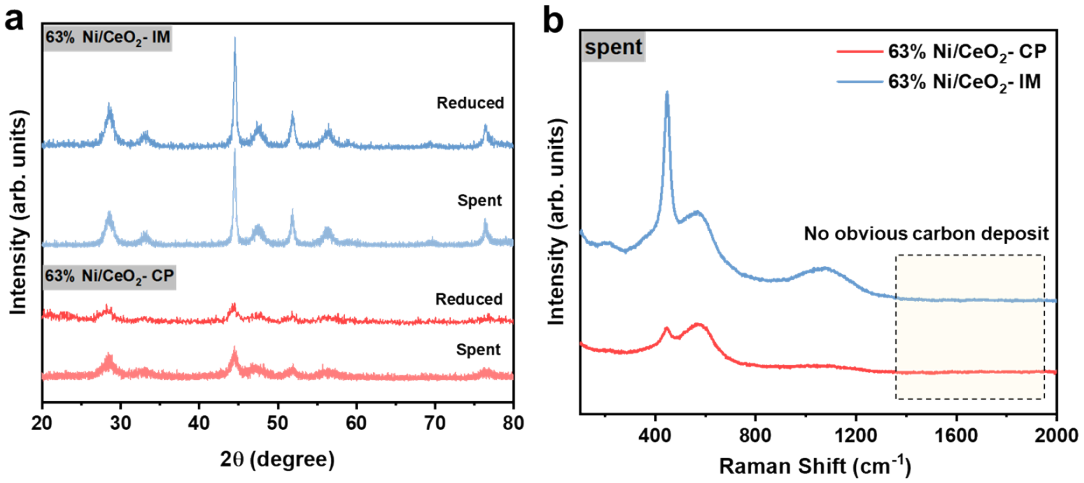

Fig S3: a XRD patterns of the reduced and spent 63% Ni/CeO_2-CP and 63% Ni/CeO_2-IM catalysts **b** Raman spectra of the spent 63% Ni/CeO_2-CP and 63% Ni/CeO_2-IM catalysts

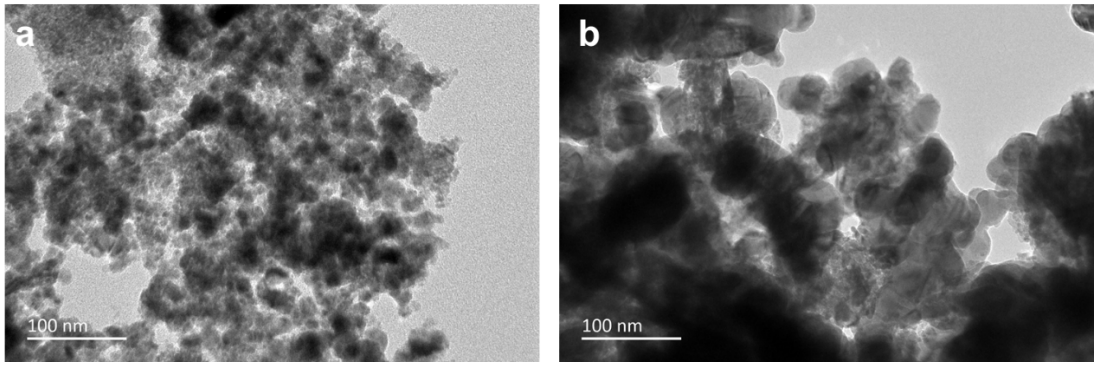

Fig S4: TEM images of the reduced **a** 63% Ni/CeO_2-CP and **b** 63% Ni/CeO_2-IM catalysts.

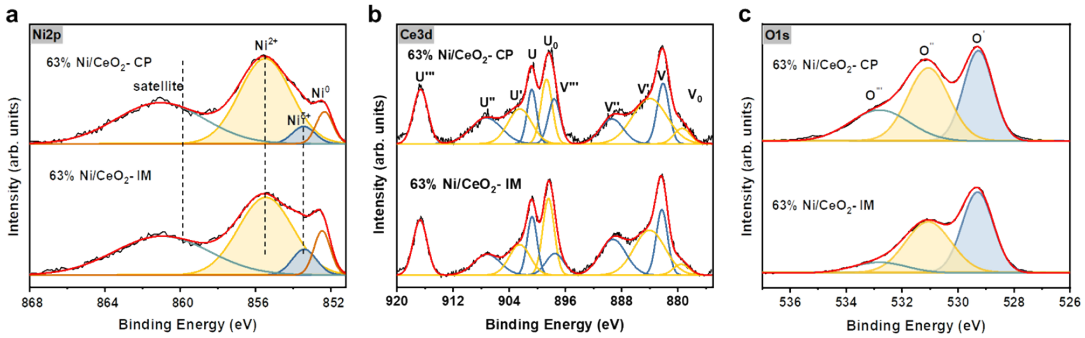

Fig S5: Fitting XPS spectra of **a** Ni2p, **b** Ce3d, and **c** O1s for the reduced 63% Ni/CeO_2-CP and 63% Ni/CeO_2-IM catalysts.

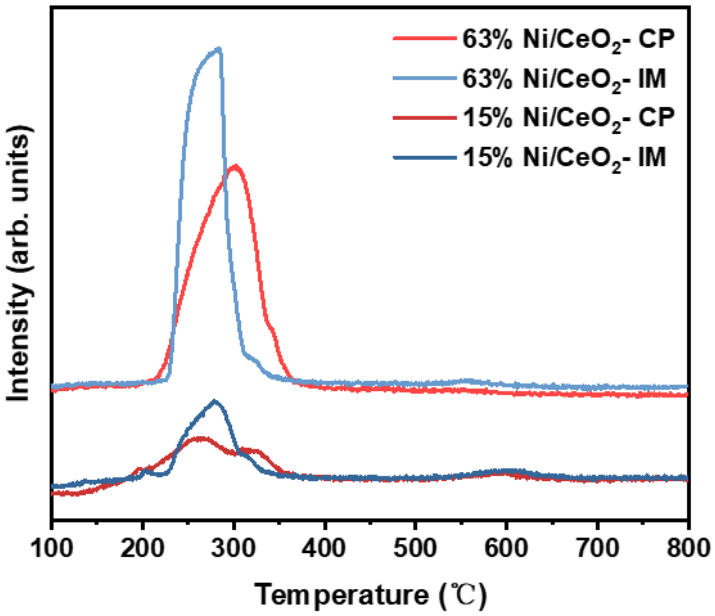

Fig S6: H_2-TPR results of 63% Ni/CeO_2-CP, 63% Ni/CeO_2-IM, 15% Ni/CeO_2-CP, and 15% Ni/CeO_2-IM.

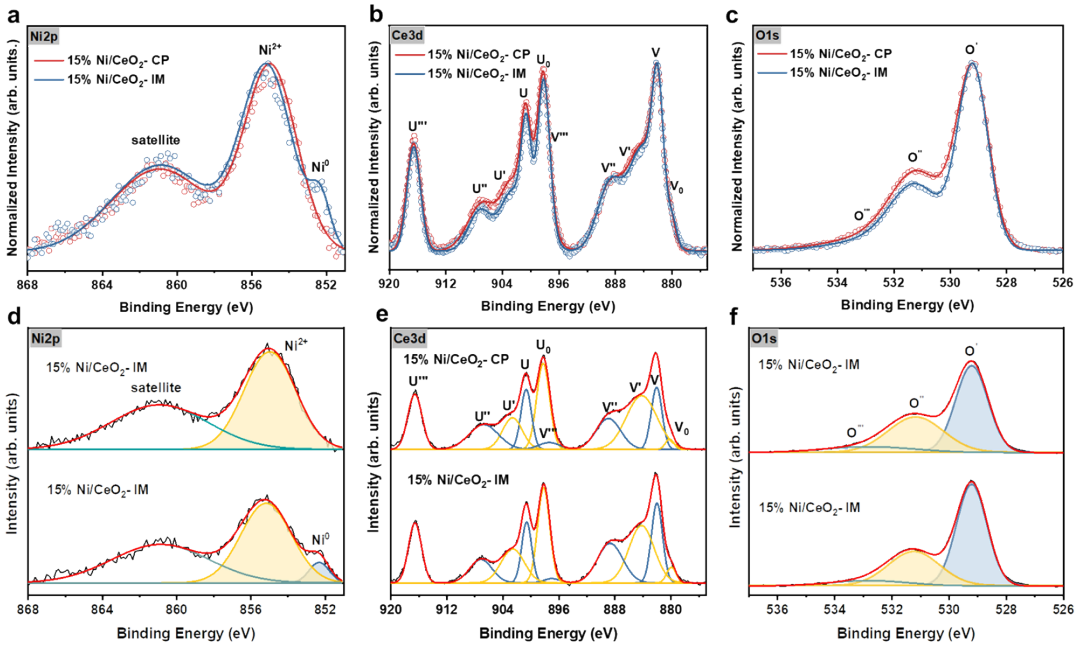

Fig S7: XPS spectra of **a** Ni2p, **b** Ce3d, and **c** O1s for the reduced 15% Ni/CeO_2-CP and 15% Ni/CeO_2-IM catalysts. Fitting XPS spectra of **d** Ni2p, **e** Ce3d, and **f** O1s for the reduced 15% Ni/CeO_2-CP and 15% Ni/CeO_2-IM catalysts.

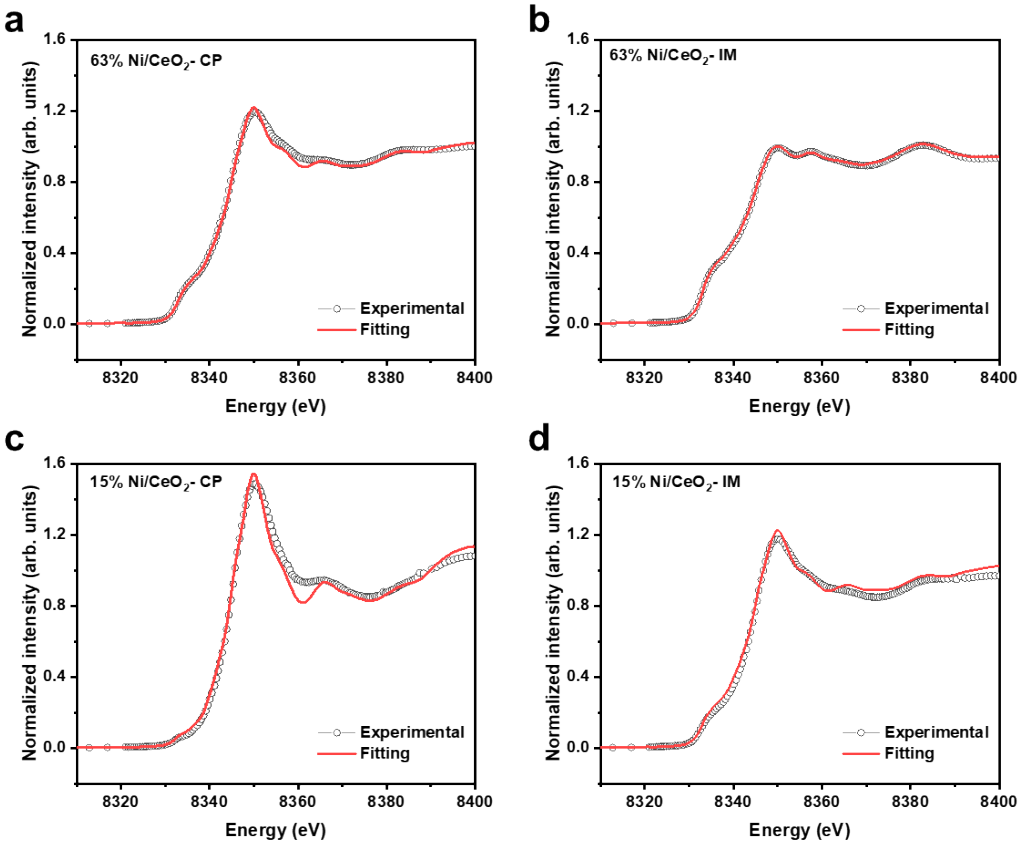

Fig S8: LCF of XANES spectra for the reduced **a** 63% Ni/CeO_2-CP, **b** 63% Ni/CeO_2-IM, **c** 15% Ni/CeO_2-CP, and **d** 15% Ni/CeO_2-IM catalysts.

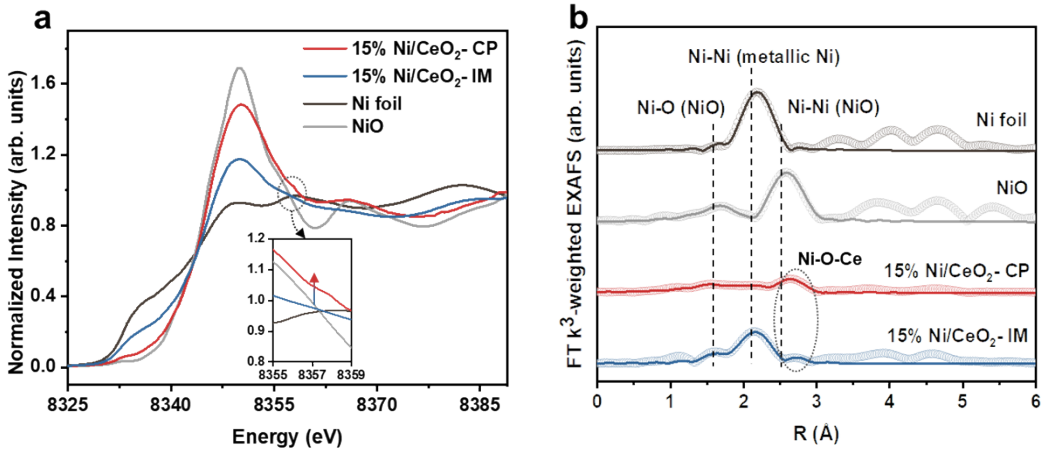

Fig S9: Comparisons between the reduced 15% Ni/CeO_2-CP and 15% Ni/CeO_2-IM catalysts **a** XANES spectra, **b** EXAFS spectra**.**

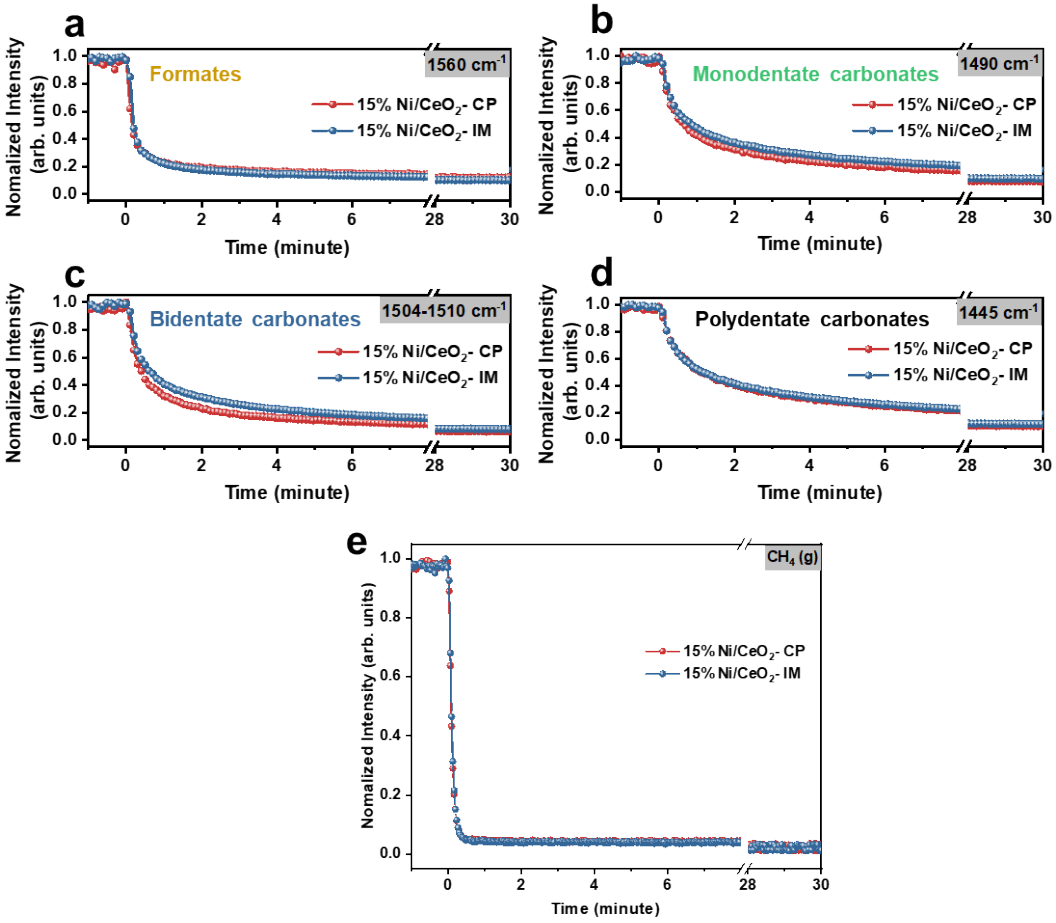

Fig S10: Intensity variation curves of **a** 1560 cm^{-1} (Formates), **b** 1490 cm^{-1} (Monodentate carbonates), **c** 1504~1510 cm^{-1} (Bidentate carbonates), **d** 1445 cm^{-1} (Polydentate carbonates), and **e** gaseous CH_4 upon switching from CO_2+H_2 ($n(H_2)$:$n(CO_2)$ = 80:1) to H_2 at 350 °C during 30 minutes.

Table S1: CO_2 methanation performance of the 63% Ni/CeO_2-CP catalyst at different temperatures (Calculated by averaging data points in 1 h on stream, 100 mg catalyst, $CO_2/H_2/Ar$ = 8/32/10 mL/min).

Temperature/°C	X_{CO2}/%	S_{CH4}/%	r(CH_4)/mmol·g_{cat}^{-1}·h^{-1}	**Carbon balance/%**
150	0.3	100	0.4	99.7
200	9.4	99.4	11.5	100.3
250	77.3	99.7	86.1	105.0
300	78.5	99.6	79.8	104.8
350	77.4	99.3	72.2	105.3
400	74.1	98.3	63.3	104.2

Table S2: Textural properties of the calcined catalysts.

Catalyst	Ni mass fraction (%)[a]	d_{NiO} (nm)[b]	d_{CeO2} (nm)[b]	Ni dispersion
63% Ni/CeO_2-CP	62.7	9.5	--	1.0
63% Ni/CeO_2-IM	63.0	22.6	7.3	0.02
15% Ni/CeO_2-CP	13.2	--	< 5.0	4.4
15% Ni/CeO_2-IM	15.3	12.6	10.5	0.55

[a] determined by ICP-OES

[b] determined by XRD

[c] determined by CO chemisorption

Table S3: Textural properties of the reduced 63% Ni/CeO_2-CP and 63% Ni/CeO_2-IM catalysts

Catalyst	d_{Ni} (nm)[a]	d_{CeO2} (nm)[a]	Surface Area (m^2/g)[b]
63% Ni/CeO_2-CP	7.8	< 5.0	68.6
63% Ni/CeO_2-IM	28.6	7.1	35.9

[a] determined by XRD

[b] determined by BET