Fabricating Stable CoNi Active Sites and Elucidating the Role of N in Enhancing Hydrogenation and Dehydrogenation of MgH_2

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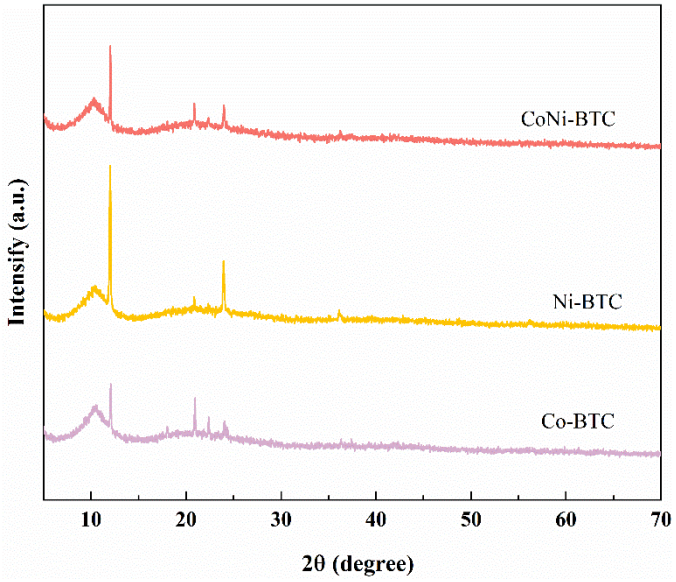

Fig. S1. XRD patterns of the as-prepared Co-MOF, Ni-MOF and CoNi-MOF.

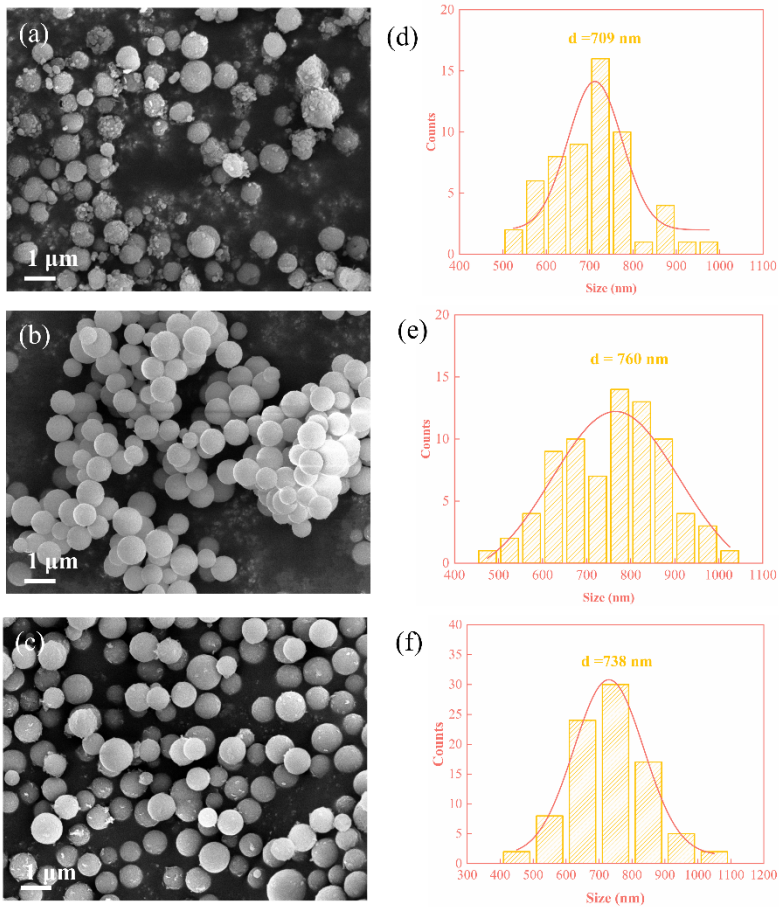

Fig. S2. SEM characterization (a, b ,c) and particle size distributions (d, e, f) of the as-prepared Co-MOF, Ni-MOF and CoNi-MOF.

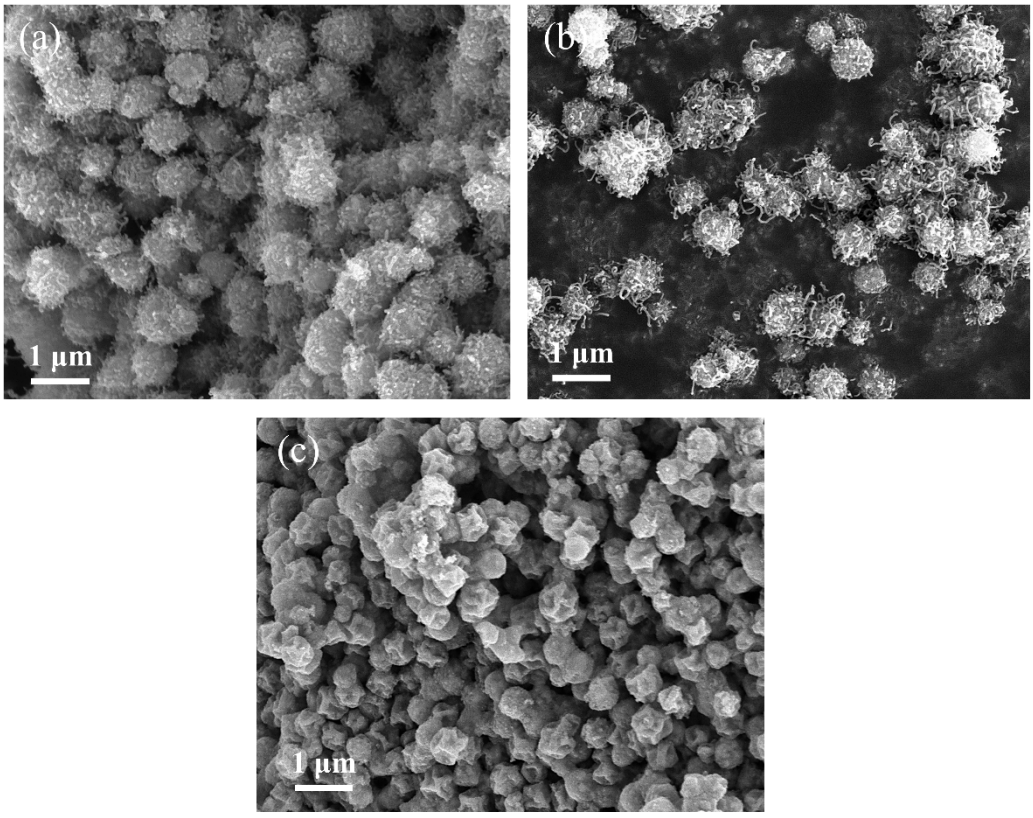

Fig. S3. SEM characterization for (a) Co@N-CNTs, (b) Ni@N-CNTs and (c) CoNi@C.

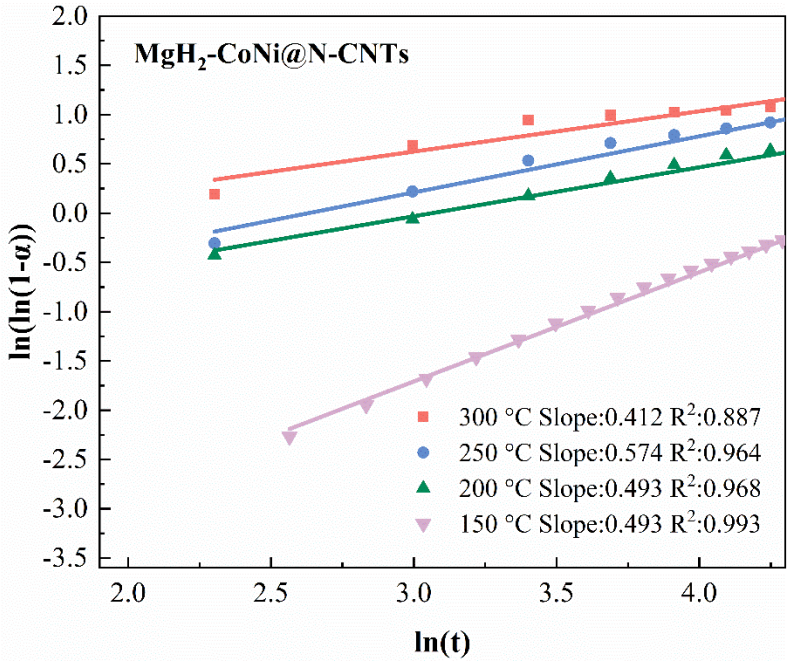

Fig. S4 Isothermal hydrogenation JMAK curve plots of MgH_2-CoNi@N-CNTs at different temperatures.

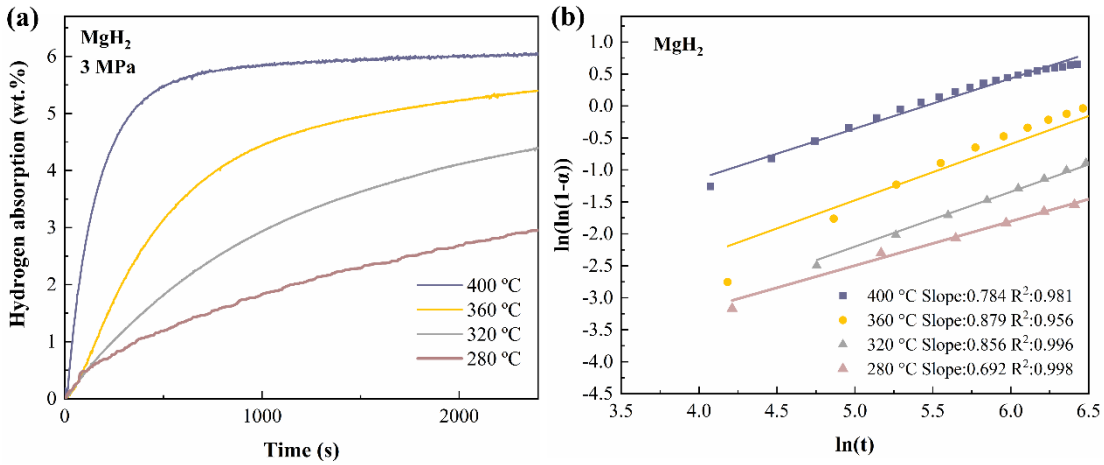

Fig. S5. (a) Isothermal adsorption curves of MgH_2 under different temperatures (b)Isothermal hydrogenation JMAK curve plots of pure MgH_2 at different temperatures.

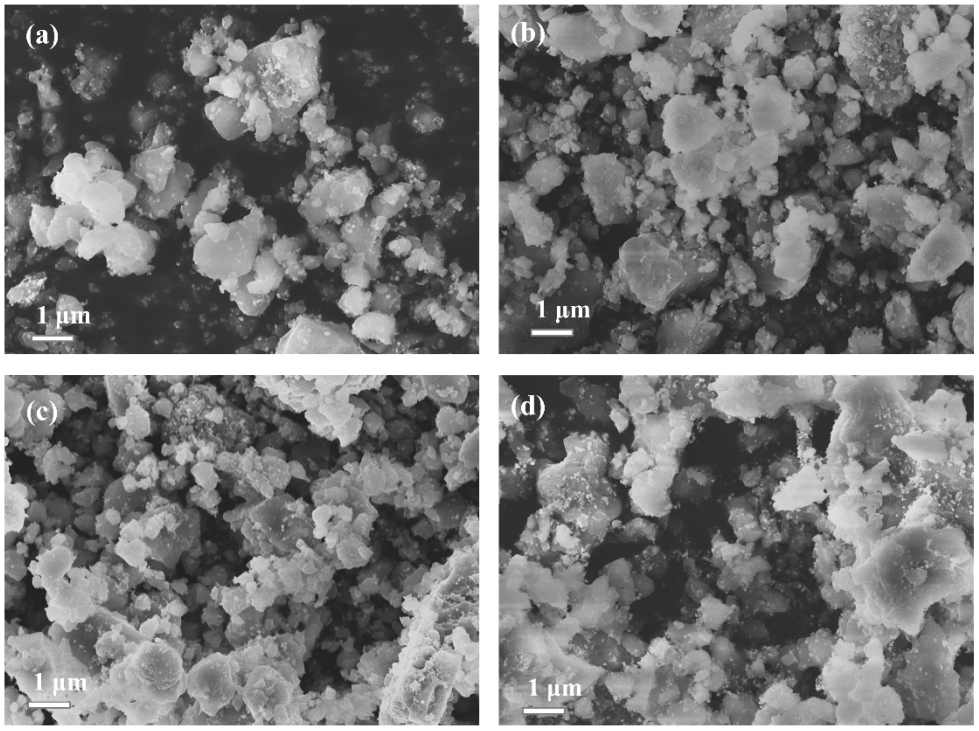

Fig. S6. The SEM images of the as-synthesized (a) MgH_2-Co@N-CNTs, (b) MgH_2-Ni@N-CNTs, (c) MgH_2-CoNi@N-CNTs and (d) MgH_2-CoNi@C.

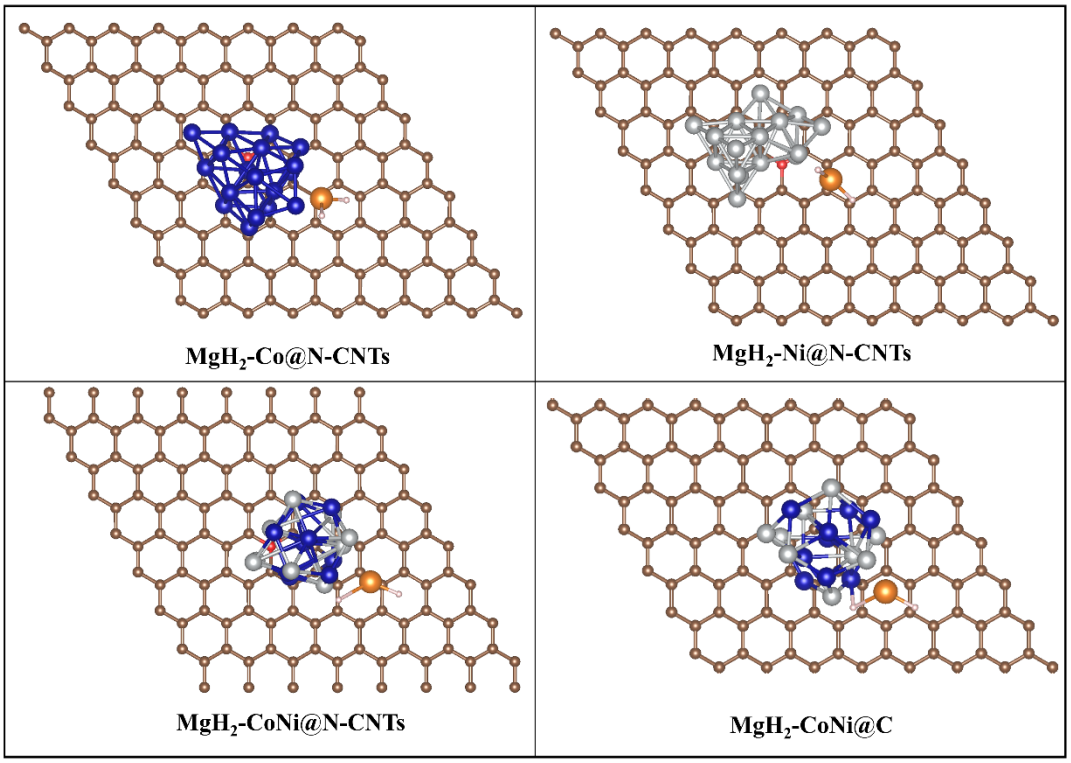

Fig. S7. Optimized structure of MgH_2-*X*@N-CNTs (*X* = Co, Ni, CoNi) and MgH_2-CoNi@C (Color identification: white is hydrogen, orange is Mg, red is N, brown is C, blue is Co and grey is Ni).

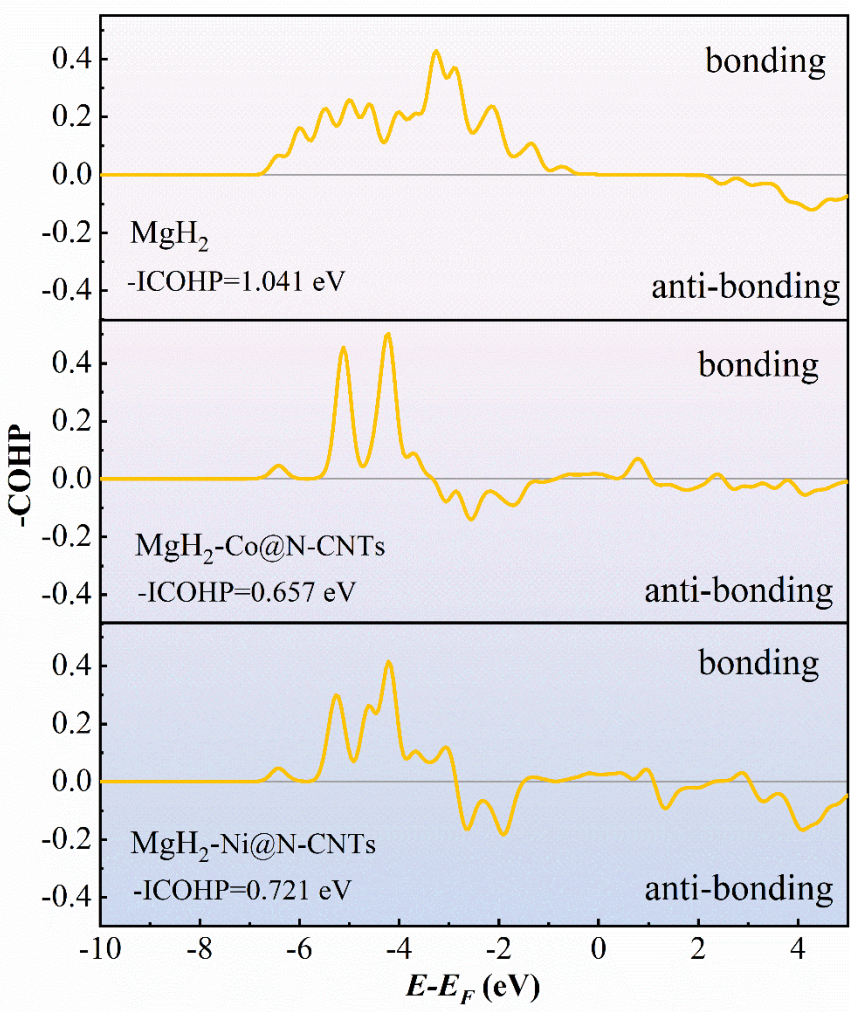

Fig. S8 Calculated -ICOHP of Mg-H for different samples.

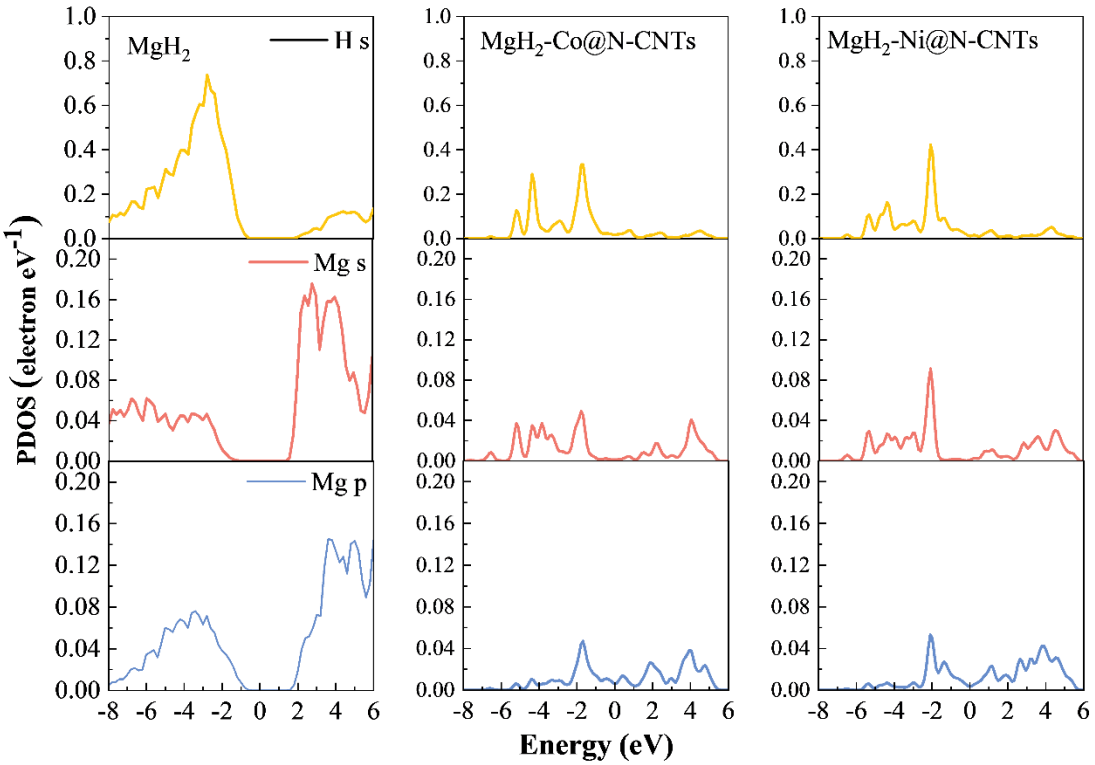

Fig. S9 PDOS of the Mg–H bond in different samples.

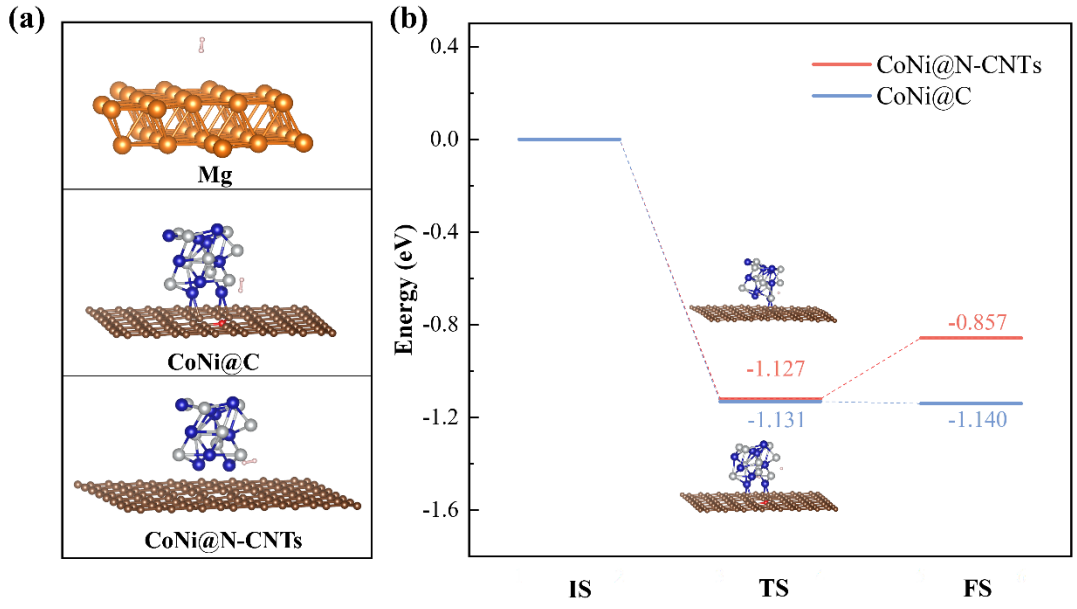

Fig. S10 (a) Optimized structure of Mg (0001), CoNi@C and CoNi@N-CNTs toward H_2; (b) The dissociation energy barriers for H_2 on the surfaces of CoNi@C and CoNi@N-CNTs (Color identification: white is hydrogen, orange is Mg, red is N, brown is C, blue is Co and grey is Ni).

Table S1 Comparison of the activation energy for dehydrogenation in the MgH_2 catalytic system.

Addictive	E_{de} (kJ/mol)	Refs.
VC	89.3	[1]
$HfCl_4$	102.0	[2]
MnVO	81.2	[3]
$BaCoF_4$	100.0	[4]
$BaMnO_3$	99.0	[5]
CoNi@N-CNTs	78.6	This work

Table S2 H_2 absorption enrgy and H-H bong length in different systems.

	Absorption energy (eV)	bong length (Å)
Mg	-0.122	0.753
CoNi@C	-0.562	0.847
CoNi@N-CNTs	-0.608	0.856

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