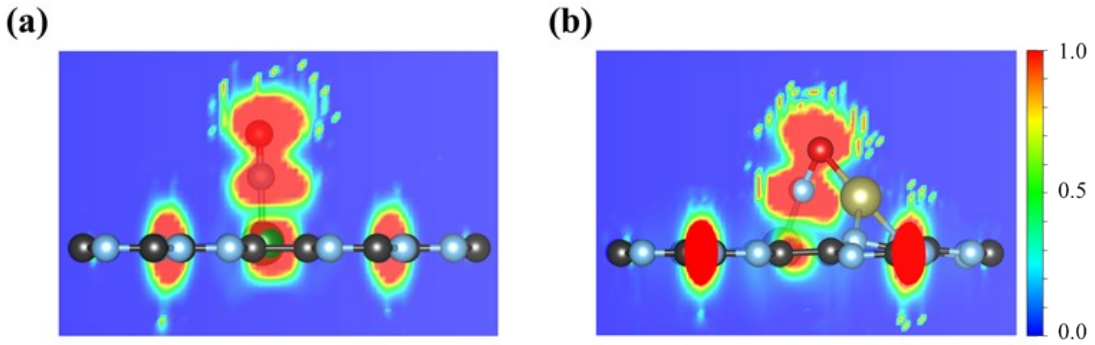
system	binding energy		cohesive energy
	Sa-CN	Da-CN	
Sc	-6.49	-9.04	-4.26
Ti	-6.00	-10.87	-5.54
V	-5.34	-10.23	-5.63
Cr	-3.53	-5.78	-4.067
Mn	-3.43	-6.31	-3.4
Fe	-3.79	-7.24	-4.79
Co	-3.92	-7.80	-5.11
Ni	-3.86	-10.02	-4.74
Cu	0.49	-5.31	-3.55
Zn	-0.51	-2.61	-1.28
Y	-7.20	-7.37	-4.32
Zr	-7.53	-11.00	-6.53
Nb	-6.10	-10.71	-7.25
Mo	-4.45	-9.68	-6.58
Ru	-5.68	-12.78	-6.99
Rh	-4.83	-10.29	-6.03
Pd	-2.58	-5.01	-4.02
Ag	-2.00	-1.90	-2.99
Cd	-0.33	0.42	-1.01
Hf	-7.11	-10.30	-6.57
Ta	-6.76	-12.01	-8.52
W	-5.17	-10.59	-8.73
Re	-3.96	-10.37	-8.08
Os	-4.89	-11.84	-8.1
Ir	-4.68	-11.10	-7.23
Pt	-3.39	-7.91	-5.65
Au	-1.30	-1.32	-3.47

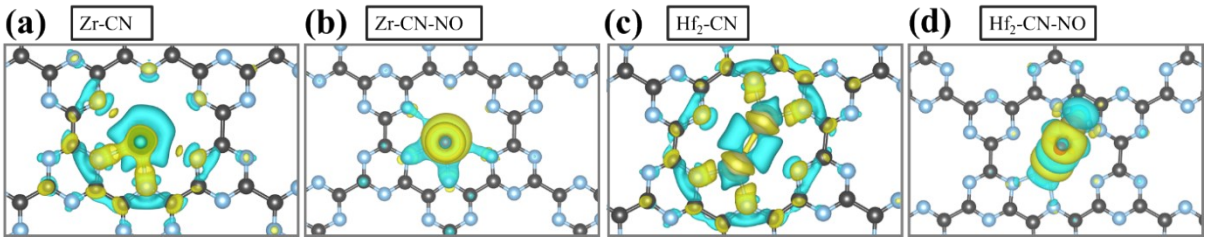
FigS1.Binding energy of SACs and DACs versus transition metal own cohesion energy values

SACs system	N-end	NO-side	O-end	Eads-H_2O
Sc	-2.12	-1.82	0.98	-0.78
Ti	-3.21	-2.79	-1.67	-0.84
Mn	-2.25	-1.68	-0.94	-1.07
Y	-1.99	-1.73	-1.13	-0.86
Zr	-3.16	-2.71	-1.58	-0.67
Hf	-3.07	-2.81	-0.43	-0.57
DACs system	Eads-end-no	Eads-side-no	Eads-O-end	Eads-H_2O
Sc	-6.28	-6.29	-5.17	-2.16
Ti	-5.33	-5.37	-5.72	-0.58
V	-3.02	-3.53	-2.89	-0.19
Cr	-3.91	-3.91	-0.36	-0.62
Mn	-2.70	-2.69	-0.45	-0.46
Fe	-3.46	-2.53	-1.27	-0.25
Co	-3.31	-3.44	-1.28	-0.09
Ni	-0.91	-3.43	-1.28	-1.71
Cu	-2.30	-1.25	-0.50	-0.44
Zn	-0.32	-0.45	0.07	-0.52
Y	-7.73	-8.10	-7.07	-3.06
Zr	-7.86	-7.04	-5.67	-1.35
Nb	-3.40	-5.42	-4.08	-1.38
Mo	-3.09	-2.62	-0.61	-0.30
Ru	-2.50	-1.89	-0.23	-0.35
Rh	-2.56	-1.34	-0.14	-1.29
Pd	-2.57	-1.65	0.12	-0.23
Hf	-7.32	-7.32	-5.77	-2.71
Ta	-5.41	-8.94	-6.98	-2.04
W	-5.26	-7.31	-2.82	-2.01
Re	-3.98	-3.49	-1.77	-0.95
Os	-2.74	-2.04	-0.43	-0.40
Ir	-2.64	-1.31	0.10	-0.28
Pt	-2.58	-1.94	0.41	-0.23

FigS2. The adsorption energy of NO at different sites and the adsorption energy of H_2O



FigS3. Electronic localization function (ELF) of NO adsorbed on Zr-CN and Hf_2-CN surfaces



FigS4. Top views of the differential charges of Zr-CN, Zr-CN-NO, Hf_2-CN, Hf_2-CN-No, with the yellow and cyan regions indicating charge accumulation and loss, respectively. The equivalent face value is set to 0.003 e $Å^{-3}$.

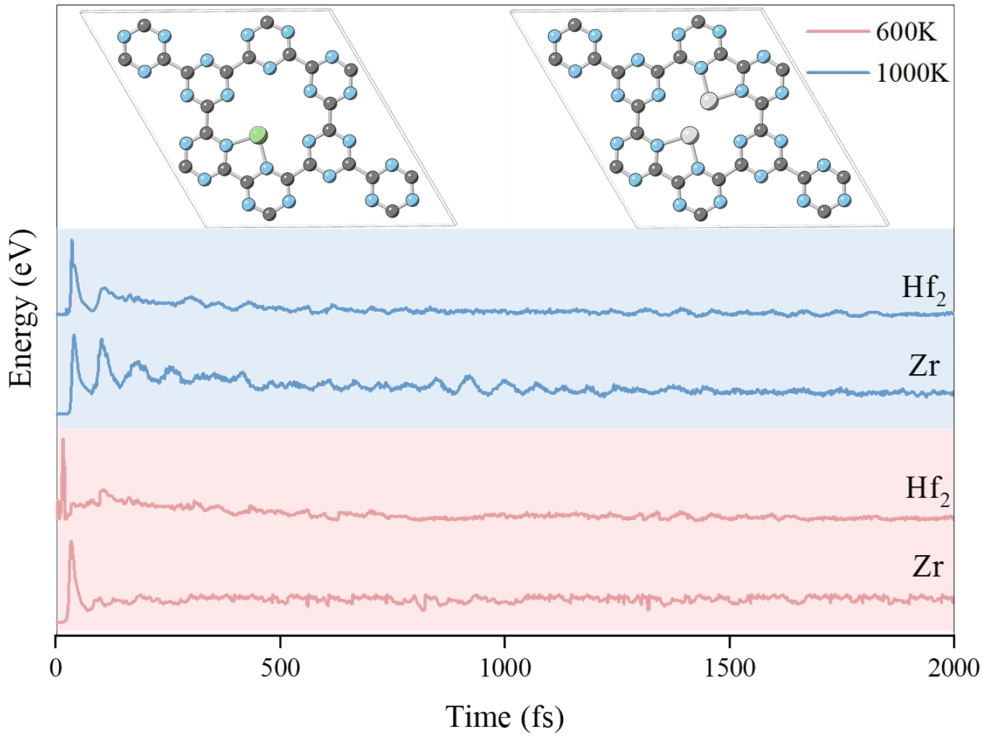


Fig.S5 The potential energy of Hf_2-CN and Zr-CN at 600 K and 1000 K for 2000 fs.

Table S1 Fukui function values of Zr-CN and Hf_2-CN

Hf_2-CN					Zr-CN				
	Fukui(+)	Fukui(0)	Fukui (-)	atomic charges		Fukui(+)	Fukui(0)	Fukui (-)	atomic charges
atom	Mulliken	Mulliken	Mulliken		atom	Mulliken	Mulliken	Mulliken	
N (1)	0.012	0.014	0.015	-0.141	N (1)	0.016	0.016	0.015	-0.222
N (2)	0.016	0.008	0	-0.748	N (2)	0.024	0.013	0.002	-0.74
N (3)	0.015	0.016	0.016	-0.154	N (3)	0.018	0.019	0.02	-0.224
N (4)	0.013	0.014	0.015	-0.142	N (4)	0.016	0.016	0.016	-0.222
N (5)	0.014	0.015	0.015	-0.155	N (5)	0.018	0.019	0.02	-0.224
N (6)	0.015	0.007	-0.001	-0.745	N (6)	0.024	0.013	0.003	-0.741
N (7)	0.012	0.012	0.012	-0.21	N (7)	0.018	0.021	0.023	-0.264
N (8)	0.013	0.012	0.012	-0.218	N (8)	0.015	0.014	0.013	-0.263
N (9)	0.014	0.013	0.012	-0.222	N (9)	0.017	0.019	0.02	-0.268
N (10)	0.014	0.009	0.004	-0.556	N (10)	0.017	0.016	0.014	-0.529
N (11)	0.011	0.012	0.012	-0.158	N (11)	0.013	0.013	0.013	-0.228
N (12)	0.015	0.015	0.014	-0.166	N (12)	0.017	0.017	0.017	-0.243
N (13)	0.015	0.009	0.002	-0.555	N (13)	0.018	0.016	0.013	-0.534
N (14)	0.013	0.013	0.013	-0.166	N (14)	0.017	0.017	0.017	-0.243
N (15)	0.012	0.012	0.012	-0.158	N (15)	0.013	0.013	0.013	-0.228
N (16)	0.012	0.012	0.012	-0.21	N (16)	0.018	0.021	0.023	-0.264
N (17)	0.014	0.013	0.013	-0.22	N (17)	0.017	0.019	0.02	-0.268
N (18)	0.013	0.013	0.013	-0.22	N (18)	0.015	0.014	0.013	-0.263
N (19)	0.014	0.015	0.015	-0.15	N (19)	0.009	0.009	0.009	-0.231
N (20)	0.013	0.014	0.014	-0.147	N (20)	0.012	0.012	0.012	-0.234
N (21)	0.016	0.007	-0.002	-0.743	N (21)	0.004	0.005	0.006	-0.378
N (22)	0.014	0.014	0.015	-0.15	N (22)	0.009	0.009	0.009	-0.231
N (23)	0.018	0.008	-0.002	-0.745	N (23)	0.004	0.005	0.006	-0.38
N (24)	0.013	0.014	0.014	-0.146	N (24)	0.013	0.013	0.013	-0.234
C (25)	0.018	0.02	0.023	0.227	C (25)	0.015	0.021	0.026	0.311
C (26)	0.016	0.013	0.01	0.237	C (26)	0.013	0.017	0.021	0.33
C (27)	0.028	0.027	0.026	0.12	C (27)	0.025	0.028	0.03	0.2
C (28)	0.026	0.026	0.025	0.121	C (28)	0.025	0.028	0.03	0.2
C (29)	0.016	0.013	0.01	0.237	C (29)	0.013	0.017	0.021	0.329
C (30)	0.017	0.019	0.021	0.229	C (30)	0.015	0.02	0.026	0.31
C (31)	0.017	0.016	0.016	0.179	C (31)	0.02	0.02	0.02	0.241
C (32)	0.015	0.016	0.017	0.163	C (32)	0.019	0.02	0.02	0.232
C (33)	0.016	0.016	0.016	0.179	C (33)	0.018	0.019	0.019	0.232
C (34)	0.026	0.021	0.017	0.211	C (34)	0.019	0.023	0.026	0.304
C (35)	0.037	0.029	0.022	0.164	C (35)	0.026	0.029	0.032	0.231
C (36)	0.02	0.018	0.016	0.213	C (36)	0.018	0.021	0.024	0.286
C (37)	0.021	0.018	0.015	0.214	C (37)	0.018	0.021	0.024	0.288
C (38)	0.037	0.029	0.021	0.164	C (38)	0.026	0.029	0.033	0.231
C (39)	0.024	0.02	0.016	0.211	C (39)	0.019	0.023	0.027	0.304
C (40)	0.016	0.016	0.016	0.178	C (40)	0.018	0.019	0.019	0.233
C (41)	0.015	0.016	0.017	0.163	C (41)	0.019	0.02	0.02	0.232
C (42)	0.016	0.016	0.016	0.18	C (42)	0.02	0.02	0.02	0.24
C (43)	0.026	0.026	0.025	0.123	C (43)	0.019	0.019	0.02	0.244
C (44)	0.019	0.015	0.01	0.238	C (44)	0.016	0.017	0.017	0.265
C (45)	0.015	0.018	0.02	0.235	C (45)	0.017	0.018	0.019	0.264
C (46)	0.015	0.017	0.02	0.233	C (46)	0.017	0.018	0.02	0.265
C (47)	0.02	0.015	0.01	0.236	C (47)	0.016	0.017	0.018	0.265
C (48)	0.029	0.027	0.024	0.124	C (48)	0.019	0.019	0.02	0.243
Hf(49)	0.078	0.119	0.161	1.278	Zr(49)	0.187	0.152	0.117	1.475
Hf(50)	0.086	0.123	0.16	1.269					

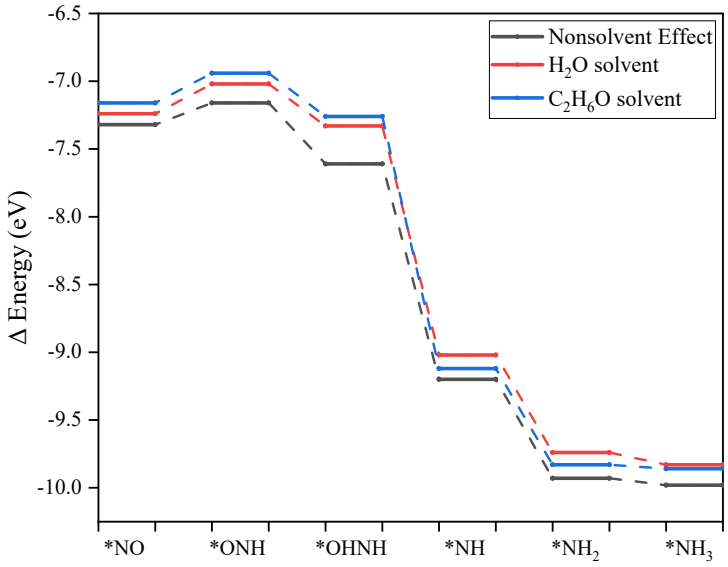
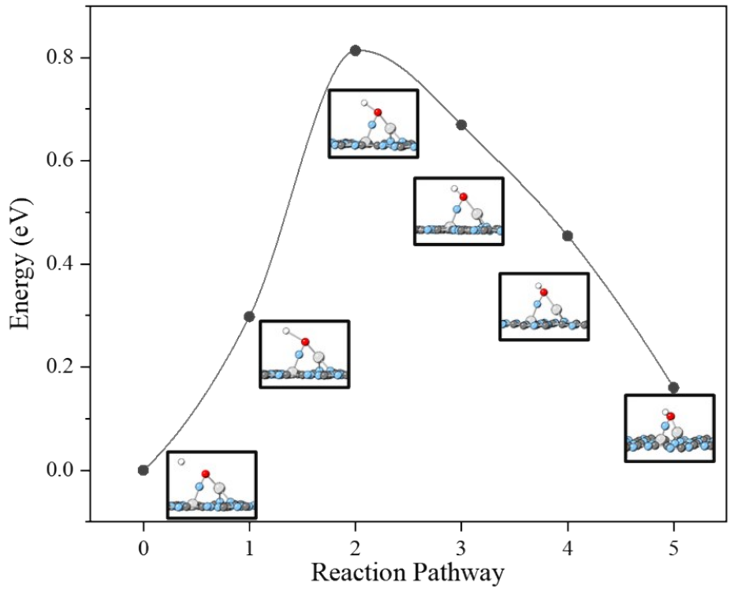


Fig.S6 Stepwise reduction of NO to NH_3 on Hf_2-CN without using solvent.



FigS7: Transition state diagram for the NO hydrogenation reaction on Hf_2-CN.

Table S2 Formation energy of Hf_2-CN in different solvents

	Hf_2-CN	CN	Gaverage	Formative energy
CH_3COOH	-427.45	-406.55	-10.30	-0.31
Ethanol	-427.20	-406.34	-10.30	-0.26
H_2O	-428.95	-407.77	-10.30	-0.59