Table S1. ΔG*H of all SACs

SACs	ΔG*H
MoS_2_N_3_Sc	0.086
Ti	-0.62
V	-0.87
Cr	-0.95
Mn	-0.41
Fe	-0.40
Co	-0.36
Ni	-0.19
Cu	-0.099
Zn	-0.77
Zr	-0.67
Nb	-1.06
Mo	-1.02
Tc	-0.88
Ru	-0.61
Rh	-0.25
Pd	-0.24
Ag	0.35
Cd	-0.63
Ta	-1.22
W	-1.20
Re	-1.04
Os	-0.75
Ir	-0.61
Pt	-0.43
Au	0.28

Table S2. ΔG*NOH, ΔG*NHO and ΔG^*NH_3 of all SACs

SACs	ΔG*NOH	ΔG*NHO	ΔG^*NH_3
MoS_2_N_3_Sc	1.04	-0.39	-0.95
Ti	0.53	-0.33	-0.62
V	0.43	0.16	0.08
Cr	0.72	0.81	-0.40
Mn	1.00	0.82	0.39
Fe	1.27	0.81	-0.88
Co	1.27	0.71	-0.91
Ni	1.32	0.44	-1.19
Cu	0.97	0.03	-1.18
Zn	0.84	-0.01	-1.06

Zr	0.63	-0.46	-0.04
Nb	0.36	-0.20	0.23
Mo	0.53	0.49	0.36
Tc	0.86	0.77	-0.52
Ru	0.79	0.62	-0.72
Rh	0.84	0.73	-1.14
Pd	0.95	0.02	-1.77
Ag	1.22	0.02	-1.61
Cd	1.01	-0.06	-1.05
Ta	0.32	-0.23	0.42
W	0.44	0.18	0.16
Re	0.92	0.90	-0.12
Os	0.76	0.75	-0.63
Ir	1.19	0.81	-1.34
Pt	1.10	0.32	-2.09
Au	1.43	0.21	-1.52

Table S3. Rate-determining steps and thermodynamic energy barriers(ΔG_{max}) of all SACs

SACs	Rate-determining step	ΔG_{max}
MoS_2_N_3_Sc	*NHOH	0.177
Ti	*NHOH	0.214
V	*NHO	0.162
Cr	*NHO	0.811
Mn	*NHO	0.818
Fe	*NHO	0.812
Co	*NHO	0.710
Ni	*NHO	0.436
Cu	*NHO	0.028
Zn	*NH_2O	-0.013
Zr	*NH_3	-0.037
Nb	*NHOH	0.236
Mo	*NHO	0.486
Tc	*NHO	0.773
Ru	*NHO	0.623
Rh	*NHO	0.735
Pd	*NH	0.273
Ag	*NH_2O	0.224
Cd	*NHO	-0.062
Ta	*NH_3	0.422
W	*NHOH	0.217

Re	*NHO	0.897
Os	*NHO	0.746
Ir	*NHO	0.811
Pt	*NHO	0.323
Au	*NHO	0.214

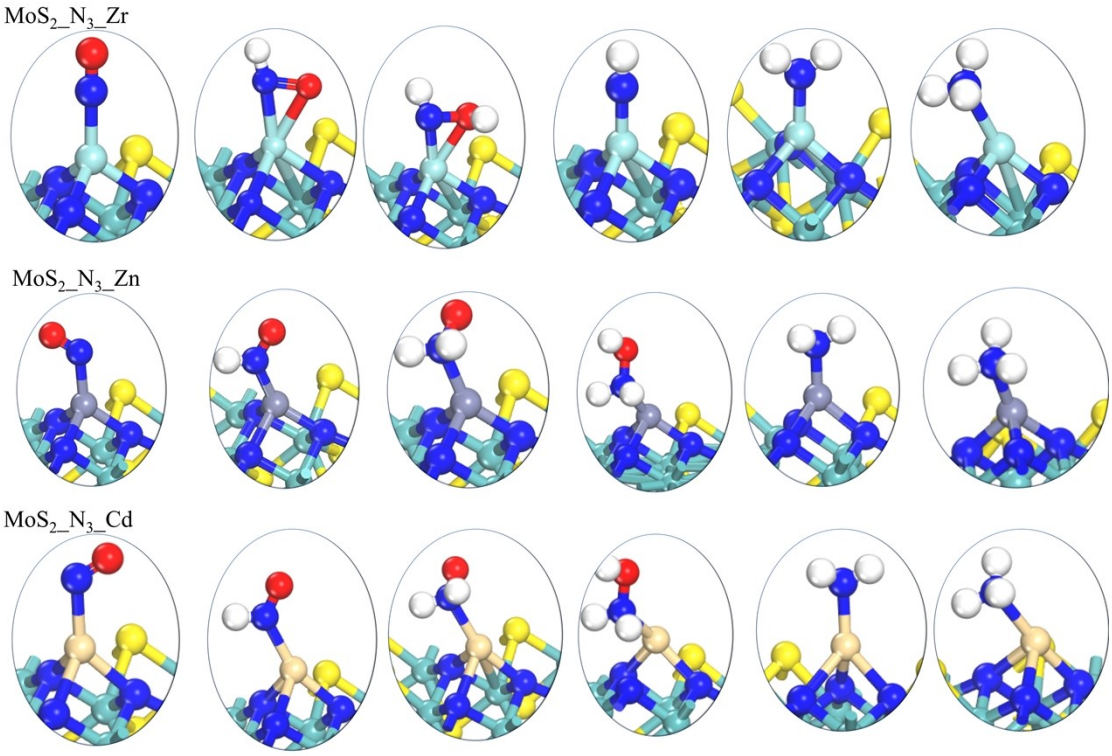

Figure S1. Structural evolution of NORR reaction processes

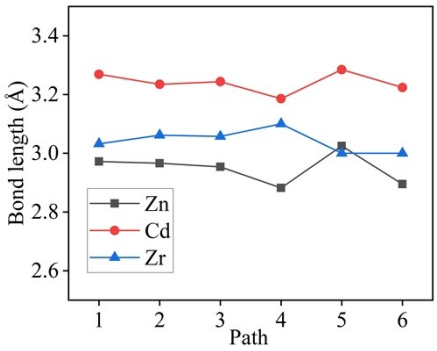

Figure S2.TM-Mo length evolution occurs in three catalyst reaction paths

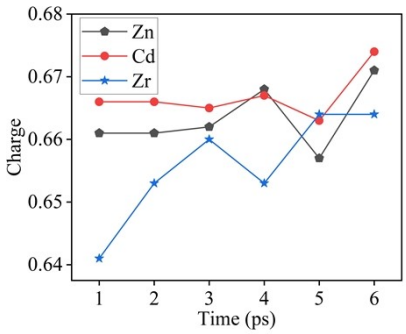

Figure S3. Charge evolution of Mo atom occurs in three catalyst reaction paths

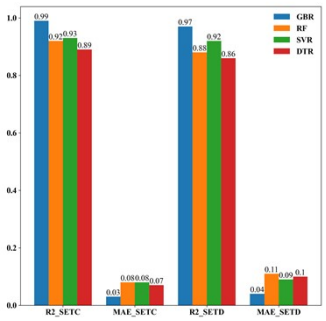

Figure S4. R^2 and MAE obtained by regression training of four machine learning models

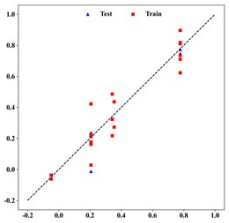

Figure S5. Regression line based on DTR method

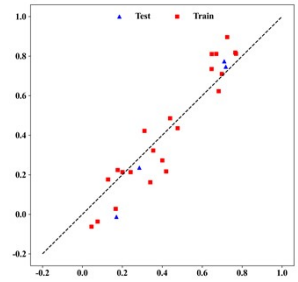

Figure S6. Regression line based on RF method

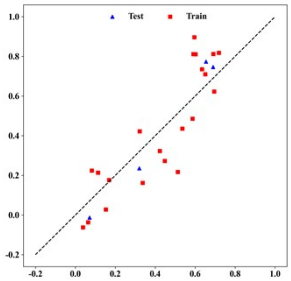

Figure S7. Regression line based on SVR method