Supporting Information for

Screening the activity of single-atom catalyst for the catalytic oxidation of sulfur dioxide with kinetic activity model

Weijie Yang[a], Hanyu Huang[a], Xiaoshuo Liu[a], Jianuo Ren[a], Kai Ma[a], Zhuohong Pan[b], Zhao Ding[c], Xunlei Ding*[d,e], Zhengyang Gao*[a]

[a] Department of Power Engineering, School of Energy, Power and Mechanical Engineering, North China Electric Power University, Baoding 071003, China
[b] Hebei Provincial Key Laboratory of Power Transmission Equipment Security Defense, North China Electric Power University, Hebei Baoding 071003, China
[c] Department of Mechanical, Materials and Aerospace Engineering Illinois Institute of Technology, Chicago, Illinois, USA, 60616
[d] School of Mathematics and Physics, North China Electric Power University, Beijing 102206, China
[e] Institute of Clusters and Low Dimensional Nanomaterials, School of Mathematics and Physics, North China Electric Power University, Beijing, People' s Republic of China

* Corresponding Author: dingxl@ncepu.edu.cn and gaozhyan@163.com

Calculation method

In detail, the convergence thresholds of the energy and force were selected as 10^{-5} eV and 0.02 eV/Å in the calculation of structural optimization. Furthermore, a K-point grid of 8×8×1 with zero-point energy (ZPE) correction was used to calculate the accurate ground-state energy.

A 5×5 graphene supercell containing 50 carbon atoms was selected to build the model of SACs. The vacuum layer of 15 Å was used to eliminate the periodic mirror error. The energy of gas molecule was calculated in a 12.3 Å × 12.3 Å × 15 Å large cell with a Γ cantered k-point grid. In detail, the energy of O atom was calculated by the half of oxygen molecule energy.

Difference between two models

To study the difference between activity model in thermodynamics and kinetics, the activity model based on reaction heat in thermodynamics was also developed (Fig. S7). Like the kinetic model, there are also significant linear relations between E_{ads}(O) and reaction heats of three reaction stages (Fig. S7(a)~(c)), which can guarantee the accuracy of activity model based on thermodynamics. Different from the kinetic model, Co/DV-N1234 shows the highest activity among the calculated six SACs, and the best adsorption energy of O is 0.84 eV (Fig. S7(d)). The lack of kinetic information makes the $\triangle E_{max}$ and the best E_{ads}(O) in thermodynamics model smaller, indicating that it is necessary to use the activity model of kinetic to predict catalytic activity.

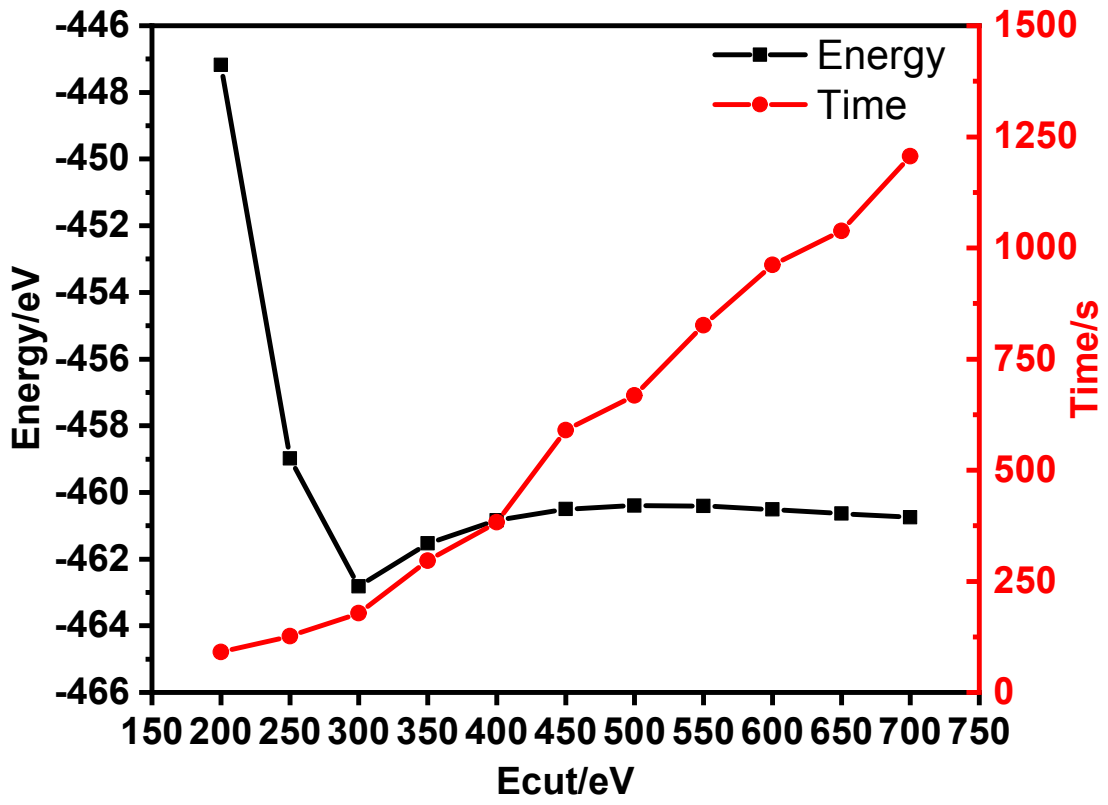

Fig. S1 Convergence test for kinetic energy cutoff.

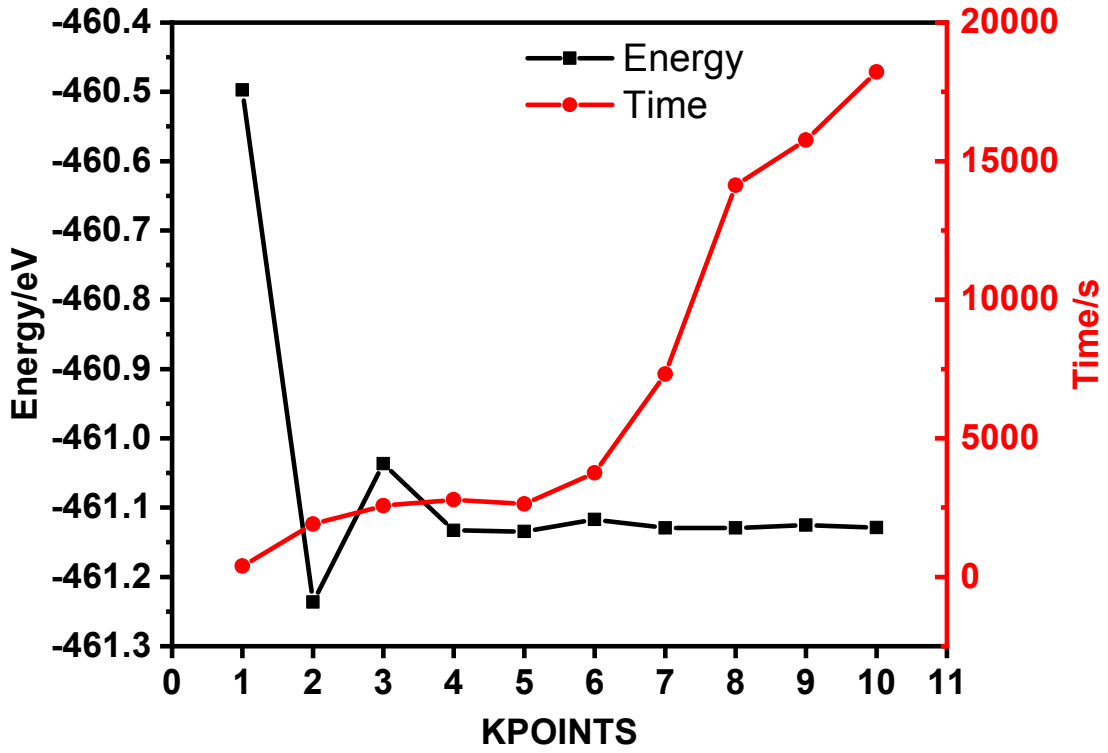

Fig. S2 Convergence test for KPOINTS.

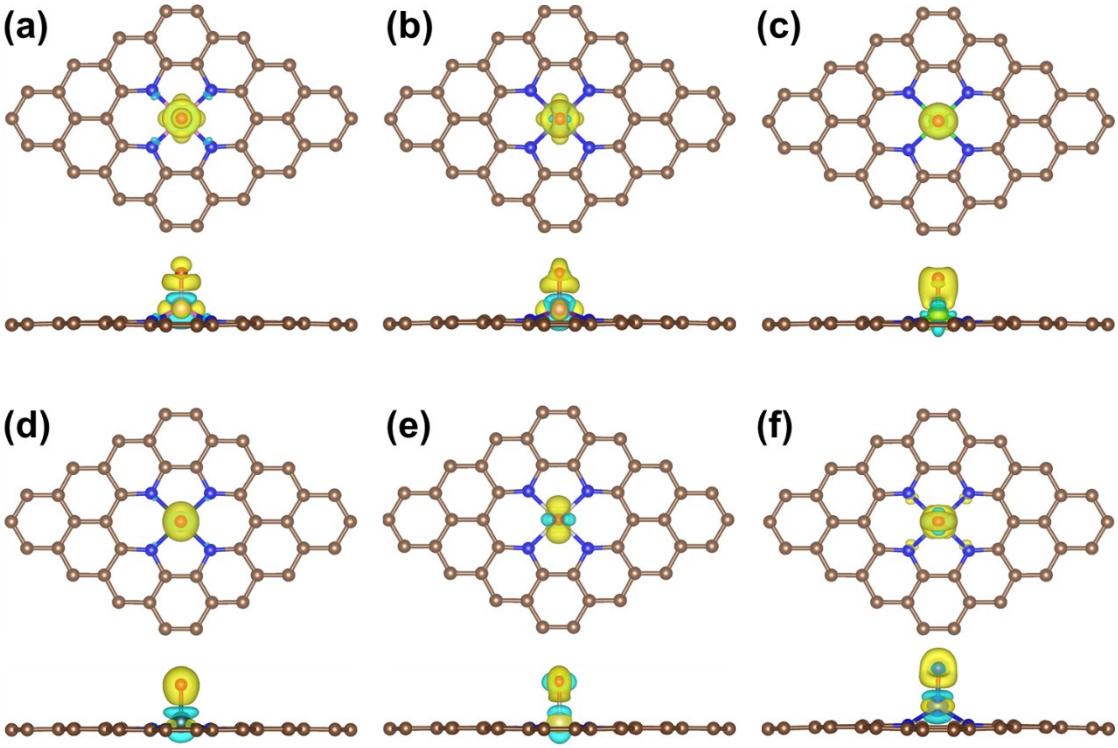

Fig. S3 Electron density difference plot of O adsorbed on six kinds of SACs (Yellow and cyan color denote gain and loss of the electron density, and the contour lines in plots are drawn at 0.08 e/Å^3 intervals).

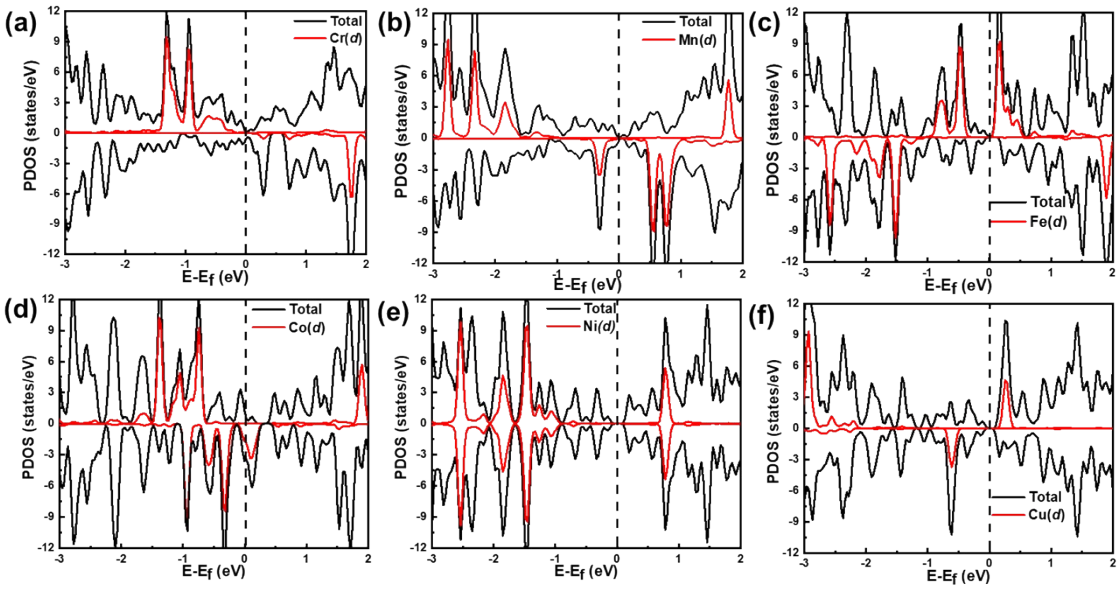

Fig. S4 Projected density of states for six SACs (a) Cr/DV-N1234 (b) Mn/DV-N1234 (c) Fe/DV-N1234 (d) Co/DV-N1234 (e) Ni/DV-N1234 (f) Cu/DV-N1234

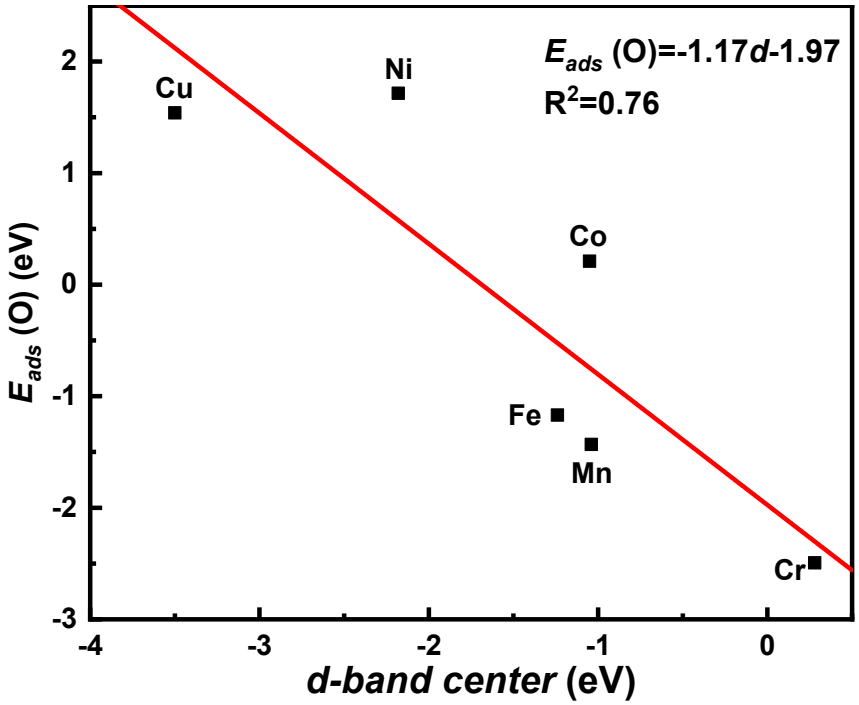

Fig. S5 Liner relationship between *d*-band center and adsorption energy of oxygen atom

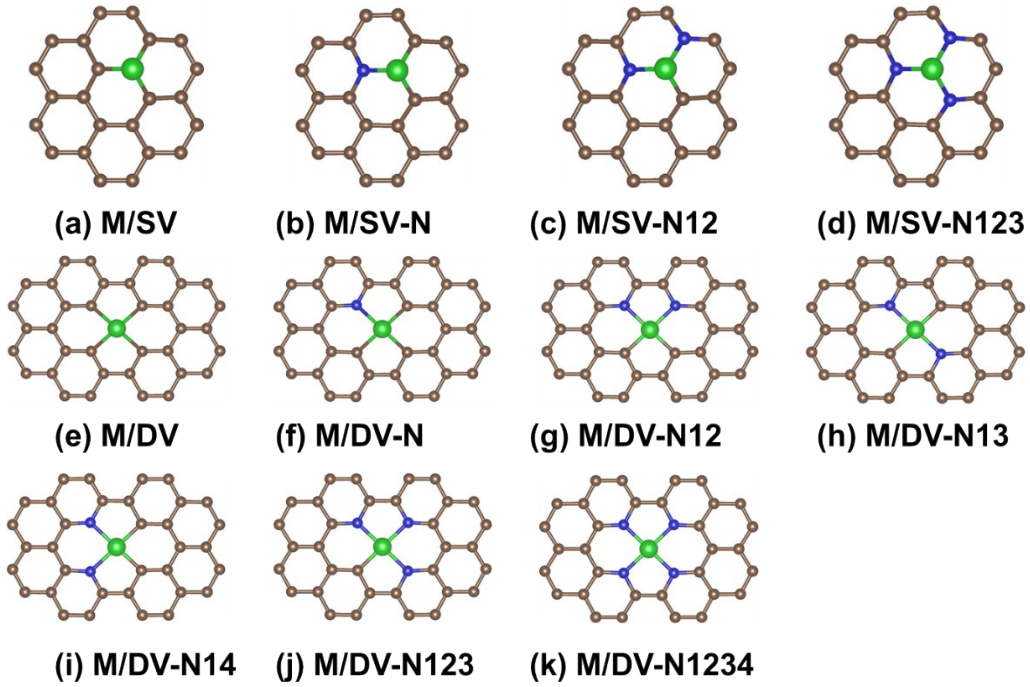

Fig. S6 Structures of active center for 132 SACs (M= Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Au, Ag, Pt, and Pd).

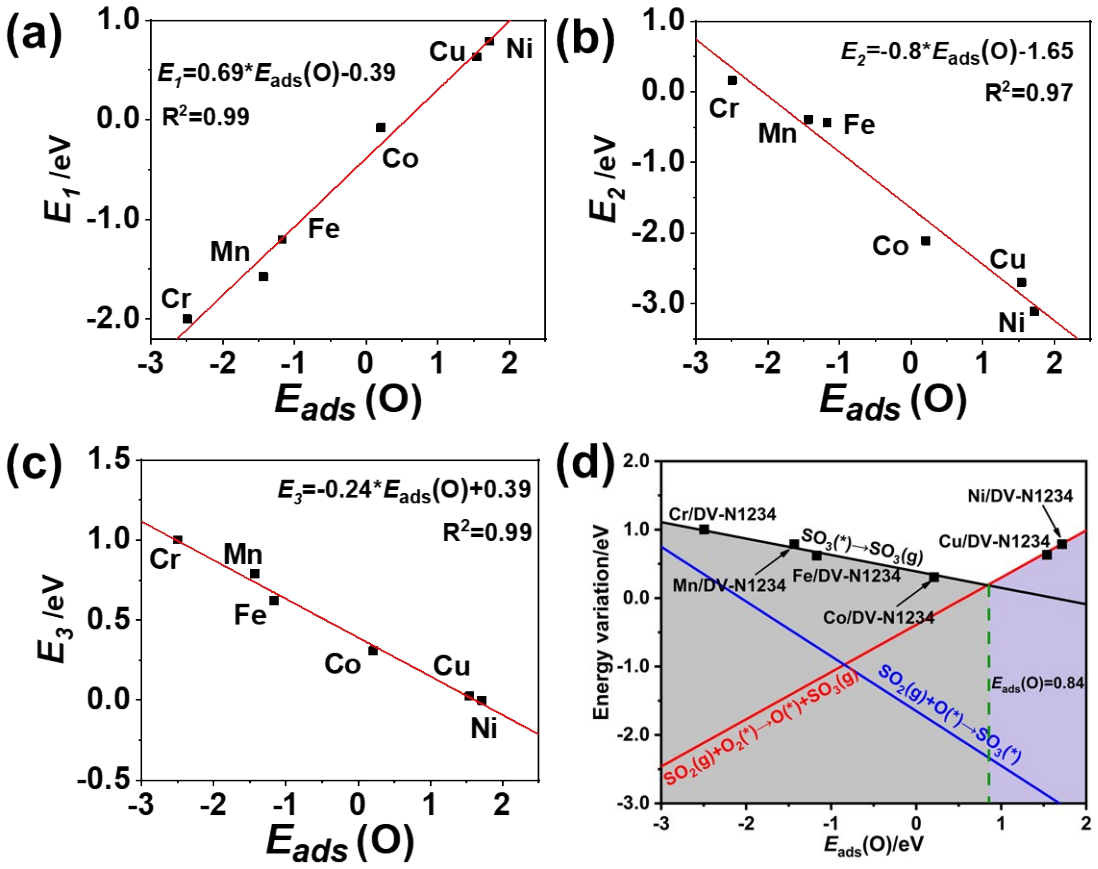

Fig. S7 (a)~(c) Liner relationship between E_{ads}(O) and other energy variation in thermodynamics (d) Energy barriers variation with different E_{ads}(O).

Table S1 Key parameters of the six SACs including bond length between metal atoms and nitrogen atoms (B_{TM-N}, Å), Bader charge of metal atoms (Q_{TM}, e), electronegativity of metal atoms (E_{TM}) in PubChem database, electronegativity of catalysts (X), absolute values of magnetic moment (M, □B), and relative binding energy of single metal atom on graphene supports (E_{rb}, eV).

Properties	Cr	Mn	Fe	Co	Ni	Cu
B_{TM-N} (Å)	1.94(1.94[1])	1.91(1.92[2])	1.89(1.90[3])	1.87(1.89[4])	1.88(1.82[5])	1.92(1.92[6])
Q_{TM} (e)	+1.26(1.25[1])	+1.29(1.36[2])	+1.10(1.08[7])	+0.88(0.87[1])	+0.85(0.77[5])	+0.93(0.92[6])
E_{TM}	1.66[8]	1.55[8]	1.83[8]	1.91[8]	1.88[8]	1.90[8]
X	5.25	5.31	6.20	7.18	8.22	10.26
M (□B)	4.00(3.97[6])	3.00(3.01[2])	2.00(2.00[7])	0.85(0.89[6])	0.00(0.00[9])	1.00(1.01[6])
E_{rb} (eV)	-3.08(-2.57[1])	-4.32(-3.68[2])	-3.53(-2.86[7])	-3.79(-3.69[1])	-3.82(-3.90[9])	-2.04(-1.60[6])

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