Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supporting Information

Rational design of transition metal single-atom electrocatalysts: A simulation-based, machine learning-accelerated study

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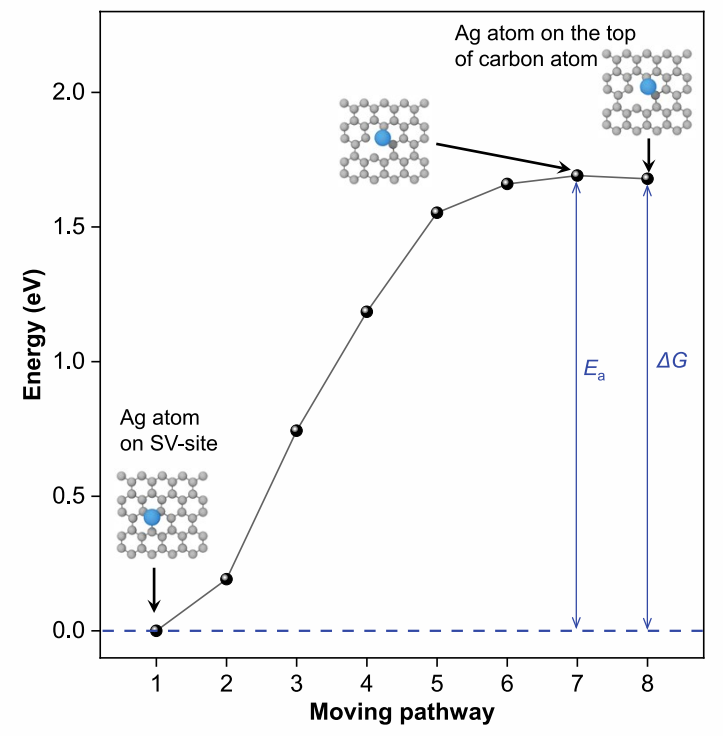

Fig. S1 Energy variation of the system during the process of a Ag SA detaches from a SV-site and then lands on the top of the closest neighboring carbon atom. The inset images are the detailed moving path of the Ag SA from the SV-site to the top of closest neighboring carbon atom. As the Ag SA starts to detach from the SV-site, the energy increases and reaches a peak value when the SA is fully detached. The peak value defines the energy barrier E_a that the Ag SA has to overcome to leave the SV-site. ΔG is the free-energy change for the Ag SA moving from the SV-site to the top of the closest neighboring carbon atom. The positive free-energy change indicates final state (Ag SA on the top of the closest neighboring carbon atom) is unstable comparing to the initial state (Ag SA on the SV-site). Thus, it is energetically unfavorable for the Ag SA to leave the SV-site.

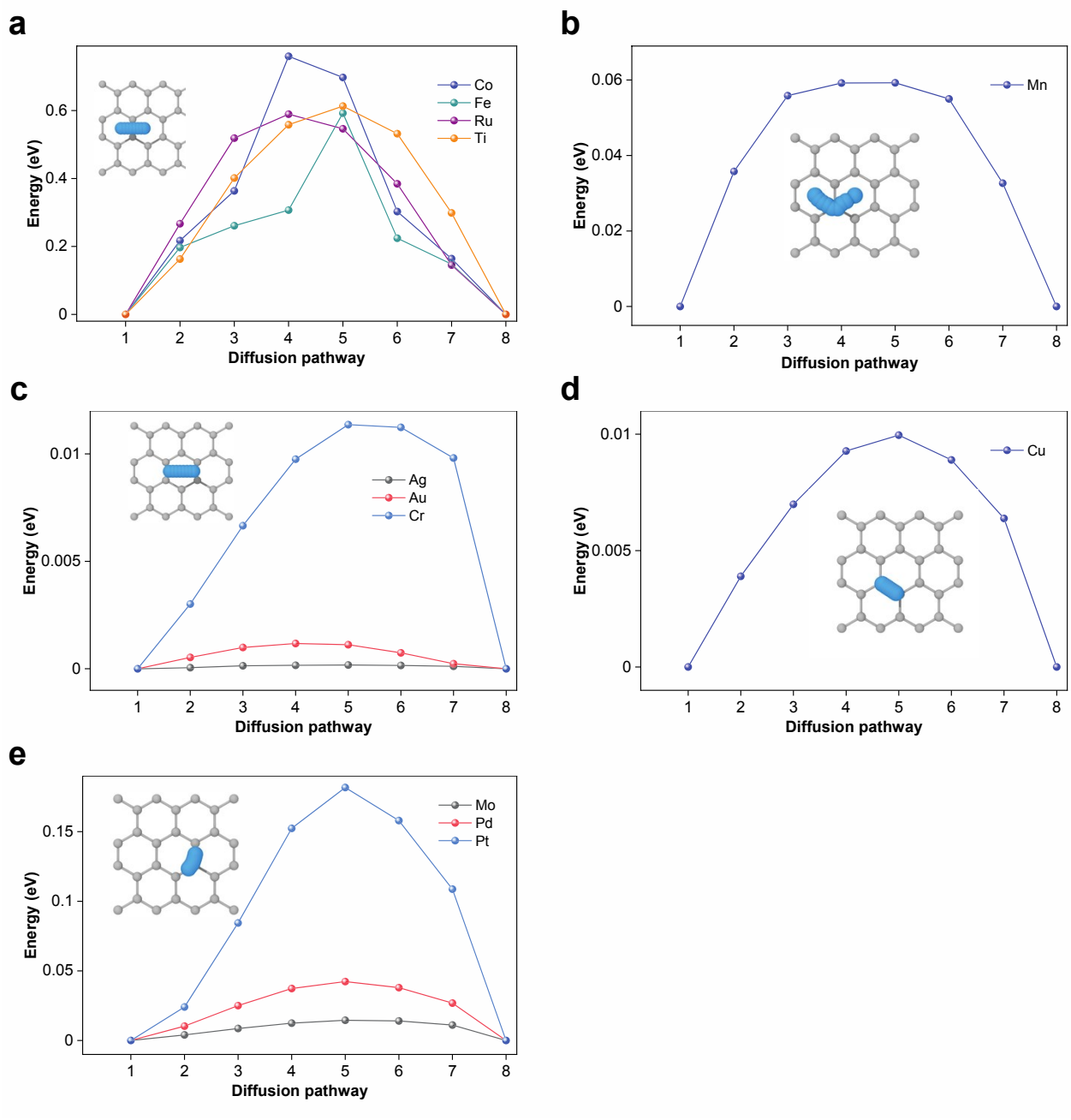

Fig. S2 Energy variation of the system during the process of a metal SA diffuses on a defect-free carbon surface. The inset image depicts the detailed diffusion path of the metal SA on the carbon surface. The peak value defines the diffusion energy barrier as shown in Figure. 1b.

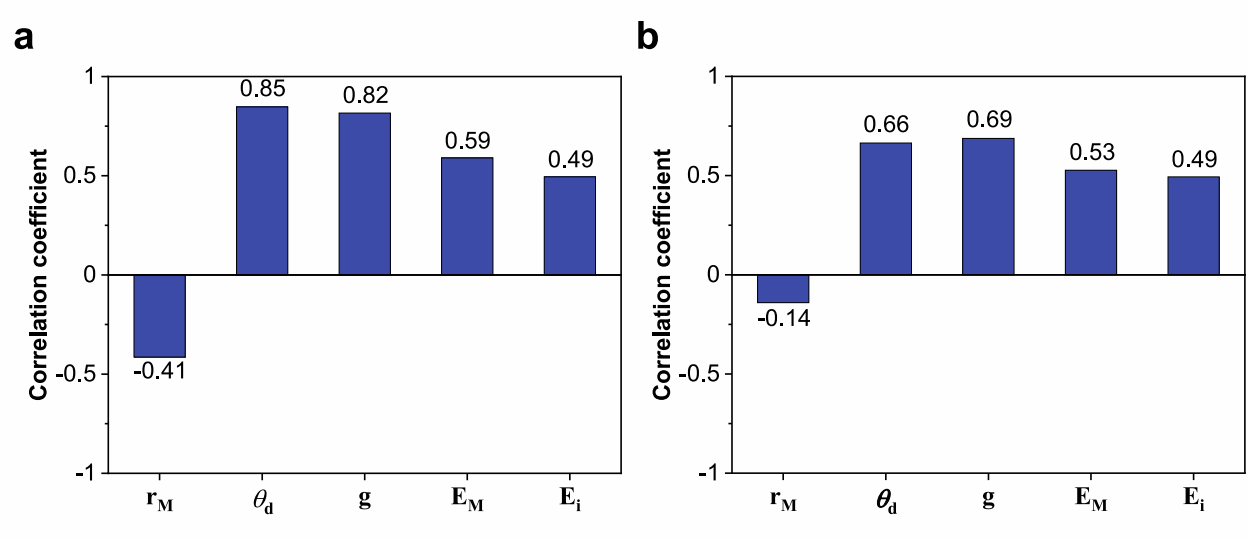

Fig. S3 Pearson Correlation coefficients of ORR activities corresponding to atomic properties of elements. a, Correlation coefficient of the five atomic properties of a SAC on a SV-site. **b**, Correlation coefficient of the five atomic properties of a SAC on a DV-site. The Pearson correlation coefficient is used to measure the strength of the correlation between two variables (i.e. the atomic parameters and the η^{ORR} in this work), where a correlation coefficient of a value of 1.0 means a perfect positive correlation, and a correlation coefficient of a value of -1.0 means a perfect negative correlation. [1] The Pearson correlation coefficient, $p_{X,Y}$, can be calculated by $p_{X,Y} = \frac{\sum(X_i - \bar{X})(Y_i - \bar{Y})}{\left[\sum(X_i - \bar{X})^2 \sum(Y_i - \bar{Y})^2\right]^{1/2}}$,where X_i and $\bar{X}$ are one of the atomic parameters and their mean value, respectively, and Y_i and $\bar{Y}$ are one of the η^{ORR} values and their mean value, respectively.

Supplementary Table

Table S1 Atomic parameters as inputs in the ML model. The first 14 elements (shaded in blue) are selected as the training/testing dataset for ML model, and η^{ORR} values of the last 15 elements (shaded in orange) are predicted by the best performing ML model.

Element	Radius [pm]	*d* electron count	Group	Electronegativity	First ionization energy [eV]
Ti	187	2	4	1.54	6.578
V	179	3	5	1.63	6.501
Cr	189	5	6	1.66	6.524
Mn	197	5	7	1.55	7.168
Fe	194	6	8	1.83	7.591
Co	192	7	9	1.88	7.581
Ni	163	8	10	1.91	7.362
Cu	140	10	11	1.90	7.450
Mo	209	5	6	2.16	6.848
Ru	207	7	8	2.20	7.103
Pd	202	8	10	2.20	8.035
Ag	172	10	11	1.93	7.305
Pt	209	9	10	2.28	8.681
Au	166	10	11	2.54	8.893
Sc	211	1	3	1.22	6.861
Zn	139	10	12	1.65	9.128
Y	219	1	3	1.22	6.517
Zr	186	2	4	1.33	6.597
Nb	207	4	5	1.60	6.636
Tc	209	5	7	1.90	7.020
Rh	195	8	9	2.28	7.195
Cd	158	10	12	1.69	8.728
Hf	212	2	4	1.30	5.752
Ta	217	3	5	1.50	7.601
W	210	4	6	2.36	7.697
Re	2.17	5	7	1.90	7.591
Os	216	6	8	2.20	8.198
Ir	202	7	9	2.20	8.681
Hg	209	10	12	2.00	10.172
C	170	2	6	2.55	11.560

Supplementary References

1. J. Lee Rodgers and W. A. Nicewander, *The American Statistician*, 1988, **42**, 59-66.