**Stability Universal Assessment for Heterogeneous Single-Atom Catalysts**

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**Abstract**: **Single-atom catalysts (SACs) generally provide high reactivity and maximize atom utilization of noble metals. How to appraise the stability of SAs dispersed on supports is a critical issue in heterogeneous catalysis. Density functional theory calculations were performed to investigate the binding and diffusion a series of metal SAs on oxide and non-oxide supports. We identify for the first time that the diffusion energy barrier of metal SAs on supports is determined with binding strength and cohesive energy of metal bulk. Feature extraction further verifies our findings based on machine learning approaches. We provided a general kinetic descriptor (Ebind)2/Ec to screen the overall stability of SACs which is significant for better catalysts design.**

Single-atom catalysts (SACs) only contain isolated individual atoms dispersed on the support, which not only maximize atom efficiency of precise metals but also offer high reactivity and selectivity.[1](#_gjdgxs), [2](#_30j0zll), [3](#_1fob9te), [4](#_3znysh7) However, single atoms on the support is generally lack of stability and can be mobile and agglomerate into clusters or particles.[4](#_3znysh7), [5](#_2et92p0), [6](#_tyjcwt) Sintering of metal particles is typically governed by particle migration and Ostwald ripening mechansims[7](#_3dy6vkm), [8](#_1t3h5sf), [9](#_4d34og8) and the interaction strength between single atoms and supports determines their propensity for sintering of single atom catalysts.[10](#_2s8eyo1), [11](#_17dp8vu), [12](#_3rdcrjn) Many previous studies suggest that the binding strength of single adatoms on a support can be used to assess the stability of SACs.[11](#_17dp8vu), [13](#_26in1rg), [14](#_lnxbz9) Ripening process always involves the diffusion and coalescence of metal adatoms. Undoubtedly, the binding strength is an important factor for descripting the thermodynamic stability of metal adatoms on a support, but the strong binding does not signify the low mobility of metal adatoms. The stability of SACs not only depends on both the thermodynamically binding strength between metal adatoms and supports, but also the activation energy barrier required for their kinetic diffusion. The proper modification of single atom adsorption strength and diffusion barrier of metal adatoms is essential to inhibit sintering.

The diffusion activation energy (Ea) can be used to evaluate the kinetic stability of a species on a support surface. Once the diffusion is difficult, we can suggest that the SACs is kinetically stable.[15](#_35nkun2), [16](#_1ksv4uv) While the calculation of Ea generally requires abundant of computational resource. For magnetic materials, such as LaFeO3 and cobalt oxides, the accurate energy is so difficult to be obtained owing to the convergence difficulty of magnetic moments which generally cause huge energy fluctuation for the same geometry. Therefore, it is significant to develop the physical descriptors for the prediction of Ea. In the present study, we developed a universal descriptor for the kinetic stability of metal adatoms on a support using the binding energy of metal adatoms and the cohesive energy (Ec) of metal bulk. Consequently, the stability of single atom catalysts can be obtained by evaluating the predicted Ea without substantial DFT calculations. As shown in Fig. 1, we have considered seven different types of supports, including reducible metal oxide (e.g. CeO2), irreducible metal oxide (e.g. MgO), perovskite (e.g. SrTiO3), two-dimensional materials (e.g. MoS2 and graphene). We investigated a great deal of transition metals on these several supports. Based on our density functional theory (DFT) calculations, we investigate the thermodynamic binding strength and kinetic diffusion of metal adatoms on supports, and find a universal correlation between them. The developed universal correlation can provide a comparable precision to the models obtained from the machine learning approaches. Finally, we provide the simple and accurate description of the binding energy of metal adatoms on a support to predict the interaction strength using the simple descriptors.

A picture containing abacus, object

Description automatically generated

**Fig. 1 Models of several catalyst supports.** Color code: red, O; white, Ce; light green, Mg; dark green, Sr; light gray, Ti; dark gray, C; light blue, Mo; yellow, S.

Previously, it is generally suggested that the binding energy can be used to evaluate the stability of metal atoms on a support.[1](#_ENREF_1), [2](#_ENREF_2) For a given metal atom, the stronger metal-support interaction indicates the lower mobility of it on the support. Fig. 2a shows the strong correlation of the required activation energy for diffusion with the binding energy of a metal on various supports. For a given support which weakly interacts with metal atoms, the interaction strength is positively correlated to the activation of metal adatoms, shown in Figure 2b, such as those on graphene and MoS2 surfaces. While for the supports which strongly interplay with metal atoms, the linear dependence between interaction strength and diffusion barrier is generally imperfect. To make certain the reason, we further consider an physical quantity, cohesive energy of metal bulk, which represents the metal-metal interaction strength. Figure 2c shows a weak correlation between Ebind and Ec, although partially a large Ec brings about a strong binding. The atom with high cohesive energy generally indicates its high reactivity and propensity to interact with the supports. Besides, σ = Ebind/Ec can be used to gauge the dispersion or agglomeration propensity of metal atoms on a support. The smaller σ, the larger clustering propensity in thermodynamics. While, the correlation between Ea and σ is also weak, shown in Fig. 2d. It means that the diffusion barrier does not solely depend on the ratio σ.

A close up of a map

Description generated with high confidence

**Fig. 2 Diffusion of single metal atom on supports. a**, Quadratic scaling relation between diffusion activation energy Ea and binding energy Ebind. **b**, Scaling relation between diffusion activation energy Ea and binding energy Ebind. **c**, Correlation between binding energy Ebind and cohesive energy Ec of metal bulk. **d**, Scaling relation between diffusion activation energy Ea and binding strength factor Ebind/Ec.

A screenshot of a cell phone

Description generated with high confidence

**Figure 3. Predictive models for diffusion activation energy Ea. a,** the weight map showing the values of normalized regression coefficients across models(The gray boxes indicate the coefficient value equals to 0. In the cases of the LASSO and elastic net, the corresponding descriptors are not selected by the model).

To obtain the appropriate description of the diffusion barrier of metal adatoms, we performed diverse machine learning (ML) studies that are commonly used for regularization and descriptor selection, including ridge regression, the LASSO and the elastic net.[22](#_4i7ojhp) The primary descriptors used here are Ebind, Ec, σ, 1/σ. Since figure 2 suggests weak linear correlations between these primary descriptors and the diffusion activation energy Ea, we include the second order terms and interactions between primary descriptor pairs as the secondary descriptors. The full mathematical form of the model contains 11 unique descriptors and is:

(1)

Our objective is to build a model with general applicability for the diffusion barrier to any metal-support systems. The quality of a predictive model is usually evaluated through two aspects, namely, the accuracy of prediction on the future data and the interpretability of the model. As linear regression models through an ordinary least square (OLS) fit often does poorly in both criteria, penalization techniques have been proposed to improve their performance.{Zou, 2005 #13} Ridge regression, for example, penalizes the magnitude of regression coefficients subject to a bound of the L2-norm.[24](#_1ci93xb) The LASSO does both continuous shrinkage and automatic feature selection by imposing an L1-penalty on the regression coefficients, and therefore, produces a simpler and more interpretable model with a fewer number of descriptors.[25](#_3whwml4) Both methods have seen increasing applications in catalysis.[26](#_2bn6wsx), [27](#_qsh70q) In addition, the elastic net is an alternative shrinkage and selection method which incorporates both L1 and L2 penalty and often shows superior prediction accuracy compared to the LASSO.[23](#_2xcytpi) One should note that kernel-based nonlinear ML methods and deep learning methods could yield lower prediction error;[28](#_3as4poj) however, given the poor interpretability and the difficulty to draw generalization, they were not considered in this work.

We first divided the randomly shuffled data containing 63 samples into the training and testing sets, with the testing set comprising 1/10 of the total data. The dataset is standardized to have a zero-mean and unit-variance so that each dimension is on the same scale. We then ran 10-fold cross-validation with 10 repeats on the training set to obtain the optimized hyperparameters for the three ML methods and an OLS mode. The relative magnitude of the normalized regression coefficients values across models are visualized in the heap map shown in Figure 3 and the detailed values are tabulated in the supplementary materials. In agreement with the USM, both the LASSO and the elastic net models select (Ebind)2/Ec as the most weighted descriptor, indicating that it is the most informative descriptor given by the models.

A screenshot of a cell phone

Description generated with very high confidence

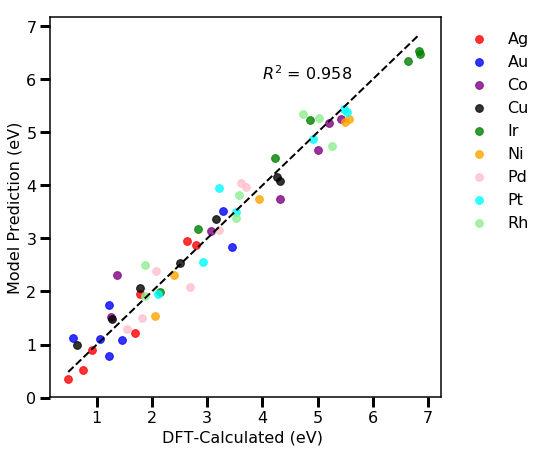
**Figure 4. Predictive models for diffusion activation energy Ea. a,** Scaling relation between diffusion activation energy Ea and binding strength factor Ebind/Ec. **b,** the prediction performance across models. **c,** the parity plot showing the DFT-calculated diffusion barrier against the model prediction.

Next, we examine the performance of (Ebind)2/Ec to describe the diffusion barrier of metal adatoms. Figure 4a shows a strong correlation between (Ebind)2/Ec and Ea. The developed relation is described as

Ea = 0.6185(Ebind)2/Ec – 0.1477 (2)

Equation (2) implies that Ea depends on both Ebind and σ, and in turn, Ea is proportional to (Ebind)2 for a given metal. The diffusion behavior of a metal atom on a support is intrinsically related to the interaction strength between them and the agglomeration/dispersion propensity in thermodynamics. We name this general relationship as “*Universal Scaling Model*” (USM).

Figure 4b shows the performance across the ML models. The prediction accuracy is evaluated through the root mean square error (RMSE) of the testing set. The LASSO, elastic net with a L1 ratio of 0.1, ridge, OLS have the RMSE of 0.186 eV, 0.166 eV, 0.155 eV and 0.170 eV, respectively. The universal scaling model has the RMSE of 0.204 eV. R2values of the training set describe the general goodness-of-fit for the model and do not vary much across different models. A comparison between the elastic net model with the lowest RMSE and the USM is presented in Figure 4c. Both models give satisfactory performance in predicting the diffusion barrier Ea with the majority of points scattered near the ideal prediction line. Therefore, we suggest the simple form of USM is sufficient and also universally applicable in describing the diffusion behavior of metal atoms on a support. Overall, the machine-learning statistical analysis further validates the intrinsic correlation between Ea and (Ebind)2/Ec for the heterogeneous SACs.



**Fig. 5 Comparison of DFT-calculated binding energy and the model prediction for single metal atoms on all considered supports. RMSE = 0.409, R2 = 0.958.**

As we show above, the binding energy is required to predict the diffusion properties for metal atoms on a support. Next, we explore some simple descriptors to describe the binding energies. The primary feature space contains four common descriptors: 1) the formation energy of vacancy Evac to evaluate the activity of surface lattice atoms (e.g. O, S and C) on a support; 2) the cohesive energy of metal bulk Ec; 3) the electronegativity difference Δχ between metal atom and surface lattice atoms to evaluate the electron transfer ability between them; 4) the coordination number CN to describe how many surface lattice atoms can interact with the metal single atom. Some other descriptors are also generally adopted to describe the metal-metal or metal-oxide interactions, such as, (*n* –1)th and *n*th ionization energies (IE*n*) of the bulk metal in the *n*+ oxidation state,[29](#_1pxezwc) electron affinity (EA),[29](#_1pxezwc) highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) of single metal atoms relative to vacuum from DFT calculations,[30](#_49x2ik5), [31](#_2p2csry) chemical potential of the electrons in the metal (ϕ) and the discontinuity in electron density between the two binding metals (*η*1/3).[32](#_147n2zr) While these descriptors generally requires a lot of complicated calculations, and consequently is inapplicable to predict the binding energy of metal atoms on a support. Moreover, some of these descriptors are correlated to each other, and generally reflect the same information. Therefore, we should adopt the independent descriptors to avoid the overlap pollution of information.

In this section, the adopted four descriptors are independent between each other, and also can be easily obtained or have already been known, such as the electronegativity of metal and surface lattice atoms. Similarly, the primary descriptors can be combined to generate the secondary descriptors containing their second order terms and pairwise interactions. The performance of an OLS model obtained from the same test/train split and 10 fold cross-validation scheme is shown in Figure 5 and the model formula is shown in the supplementary materials. The testing RMSE is 0.409 eV, indicating of a good fit. Other regularization and selection method do not show significant improvement on the fit. Recently, Senftle and co-workers described the binding energy of metal atoms on a support using the descriptors such as HOMO and LUMO energies, IEn, EA, *η*1/3 and etc, and the lowest RMSE is 0.41 eV in their trained models.[26](#_2bn6wsx) Therefore, we suggest that our developed model can be employed to universally and precisely predict the binding energy of metal atoms on a support.

In conclusion, we discover an intrinsic correlation of diffusion energy barrier Ea of metal atoms on a support with the corresponding binding energy Ebind and cohesive energy of metal bulk Ec. It opens a new insight on the understanding of the diffusion behavior and the resulting kinetic stability of single metal atoms on a support. Once the binding energy is obtained, the diffusion energy barrier can be easily determined by our developed universal scaling model. We further describe the binding energy of metal atoms on a support using some common descriptors in conjunction with machine learning tools, and then the binding energy can be easily and accurately screened. The stability of heterogeneous SACs can be evaluated from thermodynamic and kinetic properties.

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**Acknowledgements**

The authors acknowledge financial support for the research from The Netherlands Organization for Scientific Research (NWO) through a Vici grant and Nuffic funding. Supercomputing facilities were provided by NWO. This work has received funding from the European Union’s Horizon 2020 research and innovation programme under grant No 686086 (Partial-PGMs). Y.W. and D.G.V. acknowledge support for the machine-learning studies from the Catalysis Center for Energy Innovation, an Energy Frontier Research Center funded by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences under Award No. DE-SC0001004. Y.W. acknowledges also support from the Ferguson fellowship.

**Author contributions**

Y.Q.S. conceived this idea and developed the theoretical model and methodology. L.Z. participated in the DFT calculations. Y.W. participated in the machine-learning study and the data analysis. E.J.M.H. and D.G.V. provided supervision. All authors contributed to analyzing the results and writing the manuscript.

**Competing interests**

The authors declare no competing financial interest.

**Additional information**

Supplementary information is available for this paper here.

**Methods:**

**Density functional theory (DFT)**. The spin-polarized DFT calculations were carried out by the Vienna *ab initio* simulation package (VASP). The ion-electron interactions are represented by the projector-augmented wave (PAW) method and the electron exchange-correlation by the generalized gradient approximation (GGA) with the Perdew-Burke-Ernzerhof (PBE) exchange-correlation functional.

For ceria systems, the DFT+U approach was used and the U = 4.5 eV was adopted for Ce atoms. A (4×4) periodic expansion of ceria surface unit cell was used. For Brillouin zone integration, a 1×1×1 Monkhorst-Pack mesh was used. The ceria slab models consist of three Ce-O-Ce layers. The atoms in the bottom layer were frozen to their bulk positions and only the top two Ce-O-Ce layers were relaxed. For MgO(100), a (3×3) periodic expansion of surface unit cell was used with four Mg-O layers. For Brillouin zone integration, a 3×3×1 Monkhorst-Pack mesh was used. The bottom two layers were frozen to their bulk positions and only the top two layers were relaxed. For perovskite SrTiO3, a (3×3) periodic expansion of surface unit cell was used with eight atomic layers. For Brillouin zone integration, a 3×3×1 Monkhorst-Pack mesh was used. The bottom four atomic layers were frozen to their bulk positions and only the top four atomic layers were relaxed. For graphene and MoS2, a (4×4) periodic expansion of surface unit cell was used with one layer. For Brillouin zone integration, a 3×3×1 Monkhorst-Pack mesh was used. All atoms were relaxed. For all models, a vacuum gap of 15 Å was used.

The climbing image nudged-elastic band (CI-NEB) algorithm was used to identify the transition states of the metal atom diffusion on a support. The total energy difference was less than 10-4 eV and the relaxation convergence criterion was set at 0.05 eV/Å.

**Machine learning models**. We consider the usual linear regression model with n observations and p descriptors: **x1**, …, **xp**, where **xj** = (x1j, …xnj)T for j = 1,…,p. The response **y =** (y1, …,yn)T is predicted by

(2)

A model training procedure produces a vector of coefficients by minimizing a loss function. For the ordinary least squares (OLS), the loss function is the residual sum of squares.

Standardization of a dataset which rescales all descriptor values to a centered mean of 0 and have variance of 1 is a requirement for many learning algorithms. It ensures all descriptors vary within the same order of magnitude so that the selection algorithms can identify the dominate descriptors correctly. For descriptor **pi**, the scaled values **zi** are

(3)

where is the mean of training samples and is the standard deviation. The normalized form of model can be written as

(4)

The coefficients in the standardized form can be obtained from the original coefficients by,

; and , for j = 1,…,p. (5)

One should note that only reflects the weightage of each descriptor in producing the responses. Here we introduce three regularization techniques including the elastic net, the least absolute shrinkage and selection operator (LASSO) and ridge regression with the aim to select physically significant descriptors and to prevent overfitting in OLS. The coefficients are obtained through minimizing the loss function containing the residual sum of square, L2 and L1 norm of the coefficients:

 (6)

The above equation is the general formula of the elastic net, which contains both L1 and L2 penalty functions. Rewriting equation (6) gives,

, where and (7)

The LASSO and ridge regression are the special cases of the elastic net, where or , respectively. The training of machine learning models requires the tuning of hyperparameters including the degree of shrinkage and the amount of L1 penalty r1. We perform the training in Scikit-learn module for Python programming language. The dataset is first randomly shuttled and split into the training (90% of the dataset) and testing set (10% of the dataset). We then performed 10-fold cross validation on the training set. The loss function was evaluated by leaving 10% of the training set out and using the rest to fit the coefficients. This process was repeated for 10 times. The best set of coefficients giving the minimal value of the loss function during the entire training procedure were selected. The final prediction errors were determined by the RMSE of the testing set.

**Code and data availability.** The Python code written to perform the machine learning analysis together with the data supporting the plots in this paper is available on Github (the link needs to be updated).

**SI**

1. The regression formula:

The regular form:

The standardized form:

Where , j = 1, …, 11. is the mean of training samples and is the standard deviation.

Table 1. The regression coefficients in the standardized form and and in the regular form across models

|  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Notation | Descriptors | The elastic net (L1 ratio = 0.1) | | The LASSO | | Ridge | | OLS | |
|  |  |  |  |  |  |  |  |
| Intercept | | 1.240 | -0.222 | 1.234 | -0.285 | 1.246 | -0.217 | 1.248 | -2.469 |
| X1 |  | -0.019 | -0.012 | -0.025 | -0.016 | -0.077 | -0.048 | 1.229 | 0.764 |
| X2 |  | 0.000 | 0.000 | 0.000 | 0.000 | -0.033 | -0.020 | -3.862 | -2.327 |
| X3 |  | 0.000 | 0.000 | 0.000 | 0.000 | -0.170 | -0.637 | 1.540 | 5.785 |
| X4 |  | 0.050 | 0.038 | 0.078 | 0.060 | 0.234 | 0.181 | 0.545 | 0.423 |
| X5 |  | 0.000 | 0.000 | -0.045 | -0.002 | -0.002 | 0.000 | -0.513 | -0.028 |
| X6 |  | 0.000 | 0.000 | 0.000 | 0.000 | -0.034 | -0.003 | 1.551 | 0.116 |
| X7 |  | 0.000 | 0.000 | 0.042 | 0.007 | 0.042 | 0.007 | -0.448 | -0.070 |
| X8 |  | 0.207 | 0.017 | 0.370 | 0.031 | 0.410 | 0.035 | -0.748 | -0.063 |
| X9 |  | 0.722 | 0.407 | 0.522 | 0.295 | 0.616 | 0.348 | 3.592 | 2.027 |
| X10 |  | 0.248 | 0.747 | 0.349 | 1.052 | 0.465 | 1.402 | -1.075 | -3.240 |
| X11 |  | 0.000 | 0.000 | -0.034 | -0.004 | -0.206 | -0.024 | -0.201 | -0.023 |

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