Topological intelligence transfer to yield environmental catalysts' knowledge

Jie Liu[a], **Jiali Li** [a,*], **Chengzhi Hu** [a,*], **Jiuhui Qu** [a,*]

[a] *State Key Laboratory of Environmental Aquatic Chemistry, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing, China 100085* jlli@rcees.ac.cn, czhu@rcees.ac.cn, jhqu@rcees.ac.cn

1. Introduction

The integration of density functional theory (DFT) with dynamic modeling has led to a framework where the adsorption energies of intermediates in complex catalytic reactions are interconnected via scaling relations[1–4]. When the microscopic kinetic equilibrium of the reaction is taken into account, these scaling relations emerge as volcano plots[1,5,6]. Since the introduction of volcano plots, the rational design of catalysts has primarily focused on identifying the peak of the volcano, which corresponds to optimal catalytic activity, and understanding how this activity is influenced by the adsorption energies of intermediates[2–4,7]. As a result, the ideal characteristic for optimal catalytic materials has been defined as the adsorption strength of key intermediates, which should be neither too strong nor too weak[8,9]. However, this paradigm has imposed limitations on the discovery of catalysts with superior activity and selectivity[4,10–15]. Consequently, overcoming the constraints of traditional empirical design strategies to develop catalysts with enhanced efficiency and selectivity has become essential for advancing catalysis and addressing critical energy and environmental challenges[16–18].

A promising approach to overcoming these limitations involves increasing the configurational complexity of catalyst surfaces, such as through the development of high-entropy materials, which create diverse active sites that facilitate the rapid migration and transformation of intermediates, thus breaking linear scaling relations[3,4]. However, increasing configurational complexity leads to exponential growth in both the design space and the computational complexity of first-principles calculations, which significantly hinders rational design efforts. To address this challenge, we propose a novel artificial intelligence (AI) framework based on topological intelligence, which aims to alleviate the computational bottlenecks inherent in first-principles methods. This framework offers intelligent support for the rational development and optimization of high-dimensional, complex catalytic systems, while also facilitating the mechanistic interpretation of catalytic reactions. Ultimately, this approach holds promise for advancing the application of high-entropy alloys in environmental remediation.

2. Substantial section

The proposed framework utilizes three equivariant encoders[19–22], which are pre-trained through self-supervised learning[23–26], to capture the geometric structural information of the adsorbates, the bulk catalyst, and the conjugated catalyst-adsorbate surface, respectively. These three encodings interact through a ternary attention block, simulating the adsorbate-catalyst binding process, and ultimately produce a fused encoding that combines weighted contributions from different components of the catalytic system. This fused encoding is then fed into a feedforward neural network, where the model is trained using the adsorption energies derived from first-principles calculations as the training labels. Through this approach, the model effectively learns the structure-activity relationships between the microscopic structure of the catalytic system and its adsorption energies.

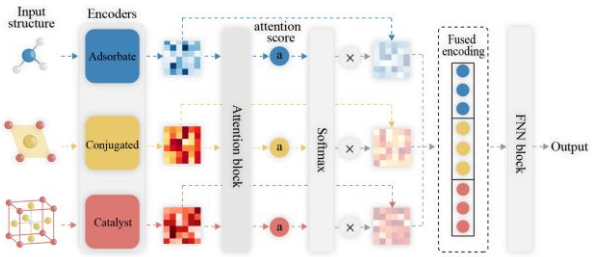

Fig. 1: Illustration of the Topological Intelligence Network

The modular design of the framework is particularly innovative, as it allows for controlled fine-tuning of the three pre-trained encoders, facilitating effective knowledge transfer from established material databases to data-scarce scenarios, such as high-entropy material systems. This strategy mitigates the risk of catastrophic forgetting and improves the model's predictive accuracy in low-data environments. Although certain aspects of the research are still ongoing, we have already optimized the screening process for several potential catalytic materials using this AI framework. For instance, the framework achieves a mean absolute error (MAE) of 0.06 eV when predicting the adsorption energies of oxygen species (O, OH, OOH) on high-entropy alloy surfaces.

We anticipate that this framework will not only accelerate the screening of heterogeneous environmental catalysts but will also leverage the attention mechanisms to analyze the interactions between catalyst surface sites and reactants. This capability will enable automated mechanistic interpretation across various catalytic systems, providing valuable insights for the rational design of catalysts. In the future, we plan to synthesize and experimentally validate the high-entropy alloy candidates identified by this framework to assess their performance in real-world environmental remediation applications.

Acknowledgments

This work was supported by the National Natural Science Foundation of China (52388101).

References

[1] Nørskov J K, Kulkarni A, Siahrostami S, Patel A. Understanding Catalytic Activity Trends in the Oxygen Reduction Reaction[J]. Chemical

Topological intelligence transfer to yield environmental catalysts' knowledge

Jie Liu[a], **Jiali Li** [a,*], **Chengzhi Hu** [a,*] , **Jiuhui Qu** [a,*]

[a] *State Key Laboratory of Environmental Aquatic Chemistry, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing, China 100085* jlli@rcees.ac.cn, czhu@rcees.ac.cn, jhqu@rcees.ac.cn

Reviews, 2018, 118(5): 2302-2312.

[2] Abild-Pedersen F, Greeley J, Studt F, Rossmeisl J, Munter T R, Moses P G, Skúlason E, Bligaard T, Nørskov J K. Scaling Properties of Adsorption Energies for Hydrogen-Containing Molecules on Transition-Metal Surfaces[J]. Physical Review Letters, 2007, 99(1): 016105.

[3] Pedersen J K, Batchelor T A A, Yan D, Skjegstad L E J, Rossmeisl J. Surface electrocatalysis on high-entropy alloys[J]. Current Opinion in Electrochemistry, 2021, 26: 100651.

[4] Pérez-Ramírez J, López N. Strategies to break linear scaling relationships[J]. Nature Catalysis, 2019, 2(11): 971-976.

[5] Seh Z W, Kibsgaard J, Dickens C F, Chorkendorff I, Nørskov J K, Jaramillo T F. Combining theory and experiment in electrocatalysis: Insights into materials design[J]. Science, 2017, 355(6321): eaad4998.

[6] Nørskov J K, Rossmeisl J, Logadottir A, Lindqvist L, Kitchin J R, Bligaard T, Jónsson H. Origin of the Overpotential for Oxygen Reduction at a Fuel-Cell Cathode[J]. The Journal of Physical Chemistry B, 2004, 108(46): 17886-17892.

[7] Nørskov J K, Wang S, Temel B, Shen J, Jones G, Grabow L C, Studt F, Bligaard T, Abild-Pedersen F, Christensen C H. Universal Brønsted-Evans-Polanyi Relations for C–C, C–O, C–N, N–O, N–N, and O–O Dissociation Reactions[J]. Catalysis Letters, 2011, 141(3): 370-373.

[8] Nørskov J K, Bligaard T, Logadottir A, Bahn S, Hansen L B, Bollinger M, Bengaard H, Hammer B, Sljivancanin Z, Mavrikakis M, Xu Y, Dahl S, Jacobsen C J H. Universality in Heterogeneous Catalysis[J]. Journal of Catalysis, 2002, 209(2): 275-278.

[9] Kuo D-Y, Paik H, Kloppenburg J, Faeth B, Shen K M, Schlom D G, Hautier G, Suntivich J. Measurements of Oxygen Electroadsorption Energies and Oxygen Evolution Reaction on RuO2(110): A Discussion of the Sabatier Principle and Its Role in Electrocatalysis[J]. Journal of the American Chemical Society, 2018, 140(50): 17597-17605.

[10] Wang P, Khan A M, Zhao C, Zhu Z, Li C, Wang F, Niu C, Jia Y. Breaking linear scaling relationships in NRR with hydrogen bonding in confined space: A case study of g-C3N4 covered Ru(001) system[J]. Surfaces and Interfaces, 2024, 47: 104219.

[11] Lai Y, Watkins N B, Rosas-Hernández A, Thevenon A, Heim G P, Zhou L, Wu Y, Peters J C, Gregoire J M, Agapie T. Breaking Scaling Relationships in CO2 Reduction on Copper Alloys with Organic Additives[J]. ACS Central Science, 2021, 7(10): 1756-1762.

[12] Zhao Z, Lu G. Circumventing the scaling relationship on bimetallic monolayer electrocatalysts for selective CO2 reduction[J]. Chemical Science, 2022, 13(13): 3880-3887.

[13] Gani T Z H, Kulik H J. Understanding and Breaking Scaling Relations in Single-Site Catalysis: Methane to Methanol Conversion by FeIV═O[J]. ACS Catalysis, 2018, 8(2): 975-986.

[14] Sun G, Zhao Z-J, Mu R, Zha S, Li L, Chen S, Zang K, Luo J, Li Z, Purdy S C, Kropf A J, Miller J T, Zeng L, Gong J. Breaking the scaling relationship via thermally stable Pt/Cu single atom alloys for catalytic dehydrogenation[J]. Nature Communications, 2018, 9(1): 4454.

[15] Jin H, Zhao R, Cui P, Liu X, Yan J, Yu X, Ma D, Song W, Cao C. Sabatier Phenomenon in Hydrogenation Reactions Induced by Single-Atom Density[J]. Journal of the American Chemical Society, 2023, 145(22): 12023-12032.

[16] Wu W, Niu C, Yan P, Shi F, Ma C, Yang X, Jia Y, Chen J, Ahmed M I, Zhao C, Xu Q. Building of sub-monolayer MoS2-x structure to circumvent the scaling relations in N2-to-NH3 electrocatalysis[J]. Applied Catalysis B: Environmental, 2021, 298: 120615.

[17] Wang Z, Hu N, Wang L, Zhao H, Zhao G. In Situ Production of Hydroxyl Radicals via Three-Electron Oxygen Reduction: Opportunities for Water Treatment[J]. Angewandte Chemie International Edition, 2024: e202407628.

[18] Saidi W A. Optimizing the Catalytic Activity of Pd-Based Multinary Alloys toward Oxygen Reduction Reaction[J]. The Journal of Physical Chemistry Letters, 2022, 13(4): 1042-1048.

[19] Gasteiger J, Groß J, Günnemann S. Directional Message Passing for Molecular Graphs[M]. arXiv, 2020[2023-09-13]. http://arxiv.org/abs/2003.03123.

[20] Gasteiger J, Giri S, Margraf J T, Günnemann S. Fast and Uncertainty-Aware Directional Message Passing for Non-Equilibrium Molecules[J]. 2020.

[21] Gasteiger J, Becker F, Günnemann S. GemNet: Universal Directional Graph Neural Networks

Topological intelligence transfer to yield environmental catalysts' knowledge

Jie Liu[a], **Jiali Li** [a,*], **Chengzhi Hu** [a,*] , **Jiuhui Qu** [a,*]

[a] *State Key Laboratory of Environmental Aquatic Chemistry, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing, China 100085* jlli@rcees.ac.cn, czhu@rcees.ac.cn, jhqu@rcees.ac.cn

for Molecules[M]. arXiv, 2022[2023-09-14]. http://arxiv.org/abs/2106.08903.

[22] Zitnick C L, Das A, Kolluru A, Lan J, Shuaibi M, Sriram A, Ulissi Z, Wood B. Spherical Channels for Modeling Atomic Interactions[M]. arXiv, 2022[2023-09-14]. http://arxiv.org/abs/2206.14331.

[23] Wang Y, Wang J, Cao Z, Barati Farimani A. Molecular contrastive learning of representations via graph neural networks[J]. Nature Machine Intelligence, 2022, 4(3): 279-287.

[24] Ni Y, Feng S, Hong X, Sun Y, Ma W-Y, Ma Z-M, Ye Q, Lan Y. Pre-training with fractional denoising to enhance molecular property prediction[J]. Nature Machine Intelligence, 2024.

[25] Yang Z, Zhao Y-M, Wang X, Liu X, Zhang X, Li Y, Lv Q, Chen C Y-C, Shen L. Scalable crystal structure relaxation using an iteration-free deep generative model with uncertainty quantification[J]. Nature Communications, 2024, 15(1): 8148.

[26] Cui T, Tang C, Su M, Zhang S, Li Y, Bai L, Dong Y, Gong X, Ouyang W. Geometry-enhanced pretraining on interatomic potentials[J]. Nature Machine Intelligence, 2024, 6(4): 428-436.