

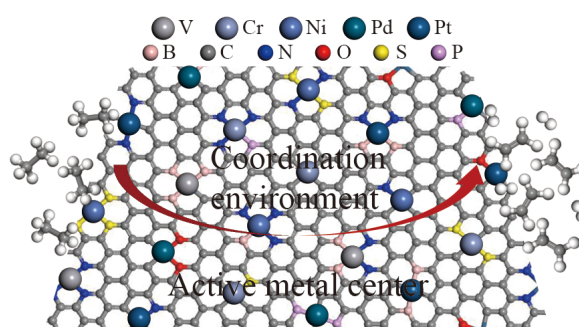
# A database for ethane dehydrogenation over heteroatom-doped graphene-supported single-atom catalysts

CAO Yajie, WANG Baojun, ZHANG Riguang\*

State Key Laboratory of Clean and Efficient Coal Utilization, Taiyuan University of Technology, Taiyuan 030024, China

**Abstract:** Single-atom catalysts (SACs) have shown great promise for ethane dehydrogenation (EDH), owing mainly to their near-100% atomic utilization, precisely tunable active sites, and superior catalytic performance. Studies reveal that the nature of active metals, the properties of support and the coordination environments are critical factors affecting EDH performance. Among various supports, graphene has emerged as an ideal support material due to its excellent thermal stability and flexible, tunable coordination structure. While previous research has explored the effects of different active metals and coordination environments, a systematic understanding of their underlying principles is still lacking due to fragmented data. To bridge this gap, this work constructed a systematic, comprehensive database of heteroatom-doped graphene-supported SACs, covering five representative metal single atoms and 51 distinct coordination environments grouped into six major categories. Through high-throughput calculations, multi-dimensional data were systematically obtained, including elementary reaction energies, vibrational frequencies, density of states and Bader charges. A rigorous quality control system was implemented at both the parameter-setting and computational result levels. The database also provides complete raw calculation files, offering reliable data support for in-depth analysis of the catalytic performance, structure-performance relationship and reaction mechanisms of heteroatom-doped graphene-supported SACs in EDH.

**Key words:** single-atom catalysts; graphene; heteroatom; ethane dehydrogenation; database; first-principles calculation



Single-atom catalysts (SACs) offer distinct advantages for the direct dehydrogenation of ethane (EDH), primarily attributed to their atomically dispersed active sites, near-perfect atomic utilization, uniform active centers and superior catalytic performance<sup>[1]</sup>. While Pt-based catalysts are still predominant in this area<sup>[2]</sup>, recent research has revealed promising light-alkane dehydrogenation activity for non-noble metal systems such as V<sup>[3]</sup>, Cr<sup>[4]</sup> and Ni<sup>[5]</sup>, expanding the pool of potential catalytic materials and enabling greater flexibility in the design of active metal centers. Notably, a key challenge for EDH, which typically operates under harsh high-temperature and strongly reducing conditions, is preserving the structural stability of isolated metal atoms.

Studies have shown that both optimizing support properties<sup>[6–8]</sup> and fine-tuning the local coordination environment of the active sites<sup>[9]</sup> can substantially improve the stability and service life of SACs. Graphene, as a typical two-dimensional support, presents a challenge for anchoring metal atoms due to its intrinsic chemical inertness. However, its tunable electronic structure and well-defined geometry make it an excellent model system for investigating structure-performance relationships, offering unique value for theoretical studies and rational catalyst design.

Current research on graphene-supported SACs has largely focused on symmetric M-N<sub>4</sub> coordination structures. Yet, their uniform electron distribution often limits charge-transfer efficiency and the tunable adsorption of reaction

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\* Corresponding author. E-mail: zhangriguang@tyut.edu.cn.

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intermediates, leading to relatively high energy barriers and compromised catalytic performance<sup>[10–11]</sup>. To address this, researchers have introduced heteroatoms like B, O, S and P to construct asymmetric M-N<sub>3</sub>Y coordination environments. This strategy effectively modulates the electronic properties of the active center, optimizes intermediate adsorption, and thereby significantly enhances both catalytic activity and stability<sup>[12–17]</sup>. Nevertheless, systematic studies on how different coordination structures and active metal centers synergistically promote the EDH reaction remain scarce. This gap hinders a deeper understanding of the structure-performance relationship for graphene-supported SACs in EDH. Therefore, conducting systematic theoretical investigations covering a wide range of coordination environments paired with various active metals is of great importance for the rational design of high-performance EDH catalysts.

The ongoing advancement of computer hardware capabilities and the widespread adoption of high-throughput computational techniques have collectively driven the rapid development of computational materials science. With the sustained promotion of the “Materials Genome Initiative”, a series of efficient high-throughput computing frameworks and material screening workflows have been successfully established<sup>[18–20]</sup>, leading to the emergence of numerous material databases. However, there is no systematic database specifically dedicated to heteroatom-doped graphene-supported SACs for the EDH reaction.

Constructing a SACs database that covers diverse heteroatom-doped graphene supports and different metal

single atoms holds dual significance. On the one hand, this database will provide critical data to uncover novel material properties and to rationalize the design of new catalysts. On the other hand, databases integrating comprehensive information, including atomic structures, electronic properties and computational parameters, will greatly facilitate data sharing and reuse within the scientific community. This would effectively minimize redundant computational efforts and significantly conserve valuable computational resources.

Therefore, establishing a specialized database for heteroatom-doped graphene-supported SACs will provide a solid theoretical foundation for the screening and preparation of high-efficiency EDH catalysts. Furthermore, this database can also serve as a representative model of doped material databases. The open sharing of its data is expected to advance research in EDH catalysis and related scientific fields significantly.

## 1 Data source

### 1.1 Data profile

The database profile of ethane dehydrogenation over heteroatom-doped graphene-supported single-atom catalysts mainly includes: Title, Data corresponding author, Data authors, Time range, Data volume, Data format, Data service system, Source of funding, Database composition and Citation format of data (see Table 1 for details).

Table 1 Database profile of ethane dehydrogenation over heteroatom-doped graphene-supported single-atom catalysts

| Title                     | A database for ethane dehydrogenation over heteroatom-doped graphene-supported single-atom catalysts                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                          |
|---------------------------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| Data corresponding author | ZHANG Riguang                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                 |
| Data authors              | CAO Yajie, WANG Baojun, ZHANG Riguang                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                         |
| Time range                | 2021–2025                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                     |
| Data volume               | 288.63 GB, 43900 entries                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                      |
| Data format               | *.zip                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                         |
| Data service system       | <a href="https://doi.org/10.57760/sciencedb.32498">https://doi.org/10.57760/sciencedb.32498</a>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                               |
| Source of funding         | Advanced Materials-National Science and Technology Major Project (2024ZD0606500)                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                              |
| Database composition      | This database systematically integrates complete structural data, along with input and output files for computational simulations, for 255 heteroatom-doped graphene-supported single-atom catalysts (SACs), generated by combining 51 distinct coordination structures with 5 metal single atoms. It is primarily organized into the following three components. DFT calculation data of SACs by Materials Studio.zip contains structural data both before and after geometry optimization, optimized adsorption structures for reactants, intermediates, and products, input and output data from transition-state searches, as well as input and output data for density of states (DOS) calculations with a data volume of 254.5 GB. Frequency analysis data of SACs by VASP.zip contains the input and output data from frequency analysis calculations performed using the Vienna Ab-initio Simulation Package (VASP), totaling 4.43 GB. The Bader charge data for SACs by VASP.zip contains the input and output files from Bader charge analysis calculations performed using VASP, totaling 29.7 GB. |
| Citation format of data   | CAO Yajie, WANG Baojun, ZHANG Riguang. A database for ethane dehydrogenation over heteroatom-doped graphene-supported single-atom catalysts[J]. <i>J Fuel Chem Technol</i> , 2026, 54(7): 20250394.<br>CAO Yajie, WANG Baojun, ZHANG Riguang. A database for ethane dehydrogenation over heteroatom-doped graphene-supported single-atom catalysts[DS/OL]. Science Data Bank, 2026. <a href="https://doi.org/10.57760/sciencedb.32498">https://doi.org/10.57760/sciencedb.32498</a> . DOI: 10.57760/sciencedb.32498.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                          |

## 1.2 Data generation method

The construction of this database is based on a monolayer graphene support, whose crystal structure is derived from the Materials Project database. A  $p(6\times6)$  monolayer graphene supercell containing 72 carbon atoms was used as support. Two adjacent carbon atoms were removed to generate a double vacancy (double C-vacancy), and a single metal atom M (V, Cr, Ni, Pd and Pt) was placed at the center of the vacancy to form a four-coordinated structure. By introducing heteroatoms such as B, N, O, P and S to modify the coordination environment, with C or N serving as the primary coordinating atoms, 51 distinct coordination structures were created. These structures are categorized into six types based on their coordination ratios (Figure 1):  $A_3B$ ,  $A_2B_2$ cis-a,  $A_2B_2$ cis-b,  $A_2B_2$ trans,  $AB_3$  and  $A_4$ , including two cis configurations ( $A_2B_2$ cis-a and  $A_2B_2$ cis-b, abbreviated as  $A_2B_2$ c-a and  $A_2B_2$ c-b) and one trans configuration ( $A_2B_2$ trans, abbreviated as  $A_2B_2$ t). All structures were fully optimized to obtain stable SAC models.

To eliminate interlayer interactions, a vacuum layer of 20 Å was applied along the  $z$ -direction to ensure isolation of the model in the non-periodic direction. DFT calculations were performed using the Dmol<sup>3</sup> module within Materials Studio (MS) 8.0. All calculations considered spin polarization<sup>[21]</sup> and utilized the GGA-PBE functional to describe electron exchange-correlation effects<sup>[22–23]</sup>. An all-electron basis set was applied for non-metal elements, while effective core potentials (ECP) were applied for metal elements<sup>[24–25]</sup>. Valence electron wavefunctions were expanded using a double-numeric plus polarization (DNP) basis set<sup>[26]</sup>. The Brillouin zone was sampled using a  $3\times3\times1$   $k$ -point mesh, and van der Waals interactions were corrected using the DFT-D3 method<sup>[27–28]</sup>.

Geometry optimizations were considered converged when the following criteria were met: energy ( $2\times10^{-5}$  Ha),

maximum atomic displacement ( $5\times10^{-3}$  Å), and maximum force ( $4\times10^{-3}$  Ha/Å). Transition states were located using the complete linear synchronous transit/quadratic synchronous transit (LST/QST) method and subsequently validated by transition-state confirmation and vibrational frequency analysis<sup>[29–30]</sup>. Free energy corrections at 873.15 K and 1 atm were implemented using the VASPKIT toolkit<sup>[31–32]</sup>.

Using high-throughput computational technology, all input and output files have been systematically stored in a structured file system. The resulting database supports convenient retrieval, querying and utilization.

## 1.3 Data content overview

This database systematically integrates complete structural data along with the input and output files for computational simulations of 255 heteroatom-doped graphene-supported SACs, generated by combining 51 distinct coordination structures with 5 metal single atoms. It is primarily organized into the following three components:

(1) SACs calculation data package based on MS

The file “DFT calculation data of SACs by Materials Studio.zip” contains structural data both before and after geometry optimization, optimized adsorption structures for reactants, intermediates, and products, input and output data from transition-state searches, as well as input and output data for density of states (DOS) calculations, all obtained using MS. The dataset is 254.5 GB and comprises approximately 11400 individual data entries.

(2) Frequency analysis data package for stable SACs based on VASP

The file “Frequency analysis data of SACs by VASP.zip” contains the input and output data from frequency analysis calculations performed using the Vienna Ab-initio Simulation Package (VASP). The dataset is 4.43 GB and comprises approximately 11500 individual data entries.

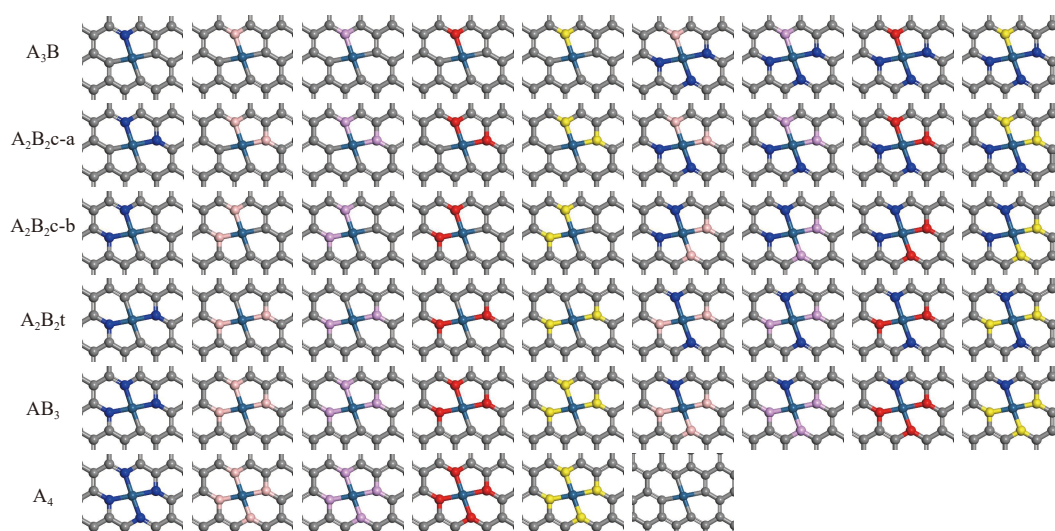


Fig. 1 51 coordination structures of heteroatom-doped graphene supported SACs with six groups of coordination environments  $A_3B$ ,  $A_2B_2$ c-a,  $A_2B_2$ c-b,  $A_2B_2$ t,  $AB_3$  and  $A_4$ , respectively; Grey, blue, pink, violet, red and yellow balls represent C, N, B, P, O and S atoms, respectively

(3) Bader charge analysis data package for stable SACs based on VASP

The file “Bader charge data of SACs by VASP.zip” contains the input and output data from Bader charge analysis calculations, also performed with VASP. The total size of this dataset is 29.7 GB, encompassing approximately 21000 data entries.

All data are organized and managed according to unified standards and naming conventions, providing comprehensive and reliable support for in-depth studies into the performance and mechanistic understanding of SACs in EDH.

## 2 Data sample description

This database is centered on the study of heteroatom-doped graphene-supported SACs for the EDH reaction. It systematically integrates parameter data, structural information, software environment details and physical property data. All calculation files are organized by computation type and structured within a unified, standardized directory hierarchy. Using Pt SACs supported on graphene with the  $-N_3B$  coordination structure (Pt- $N_3B$  SAC) as an example, the file storage structure is illustrated (see Figures 2–4 for details). All data files follow a consistent two-level naming convention: Level-1 folders are named by metal type (Cr SACs, Ni SACs, Pd SACs, Pt SACs and V SACs), and Level-2 folders are named by coordination structure type ( $A_3B$ ,  $A_2B_2c-a$ ,  $A_2B_2c-b$ ,  $A_2B_2t$ ,  $AB_3$  and  $A_4$ ). The specific data contents are as follows:

(1) MS Calculation Data (Figure 2)

The file “**DFT calculation data of SACs by Materials Studio**” contains computational files for 162 stable SACs (across different combinations of coordination structures and metal single-atom), covering: Geometry optimization (slab), Electronic energy (DMol<sup>3</sup> Energy) and Density of States (DOS), Structures of adsorbed reactants,

intermediates, and products (adsorption and co-adsorption), Transition-state search (TS) results. Additionally, it includes files for 93 unstable SACs (across different combinations of coordination structures and metal single-atom), covering: Geometry optimization (slab), Electronic energy (DMol<sup>3</sup> Energy) results. Here, unstable SACs refer to systems where the binding energy of the metal single-atom ( $E_b = E_{M/sub} - (E_M + E_{sub})$ ) is lower (more negative) than the corresponding metal’s cohesive energy ( $E_{coh-M}$ ).  $E_{M/sub}$  is the total energy of the metal-single-atom-on-graphene system;  $E_M$  is the energy of an isolated metal single-atom, and  $E_{sub}$  is the energy of the graphene support.

(2) VASP frequency analysis data (Figure 3)

The file “**Frequency analysis data of SACs by VASP**” contains frequency analysis results for six elementary reactions on the 162 stable SAC structures:  $C_2H_6 \rightarrow C_2H_5 + H$ ,  $C_2H_5 \rightarrow CHCH_3 + H$ ,  $C_2H_5 \rightarrow CH_3 + CH_2$ ,  $C_2H_5 \rightarrow C_2H_4 + H$ ,  $C_2H_4 \rightarrow CHCH_2 + H$  and  $C_2H_4 \rightarrow CH_2 + CH_2$ . The data for each reaction step includes the structures for the initial state (IS), transition state (TS) and final state (FS).

(3) VASP Bader charge data (Figure 4)

The file “**Bader charge data of SACs by VASP**” contains the Bader charge calculation results, along with the corresponding processed data, for all 162 stable SAC structures.

## 3 Data quality control and evaluation

Throughout the development of this database, rigorous convergence tests were conducted on the key parameters that influence computational outcomes—total energy, maximum atomic displacement and maximum force—ensuring that all computational parameters meet strict convergence criteria. These tests ensured that all computational settings met stringent convergence criteria, thereby guaranteeing the stability and reliability of the results, with a typical energy error of  $\pm 0.15$  eV.

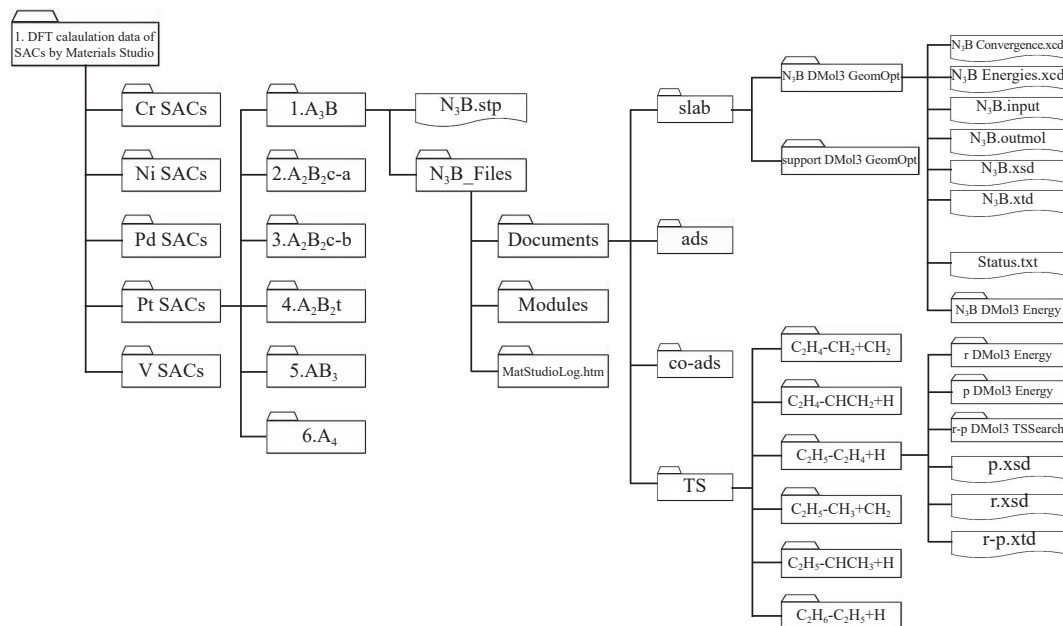


Fig. 2 The file storage directory structure of the Pt- $N_3B$  SAC in DFT calculation data of SACs by Materials Studio.zip

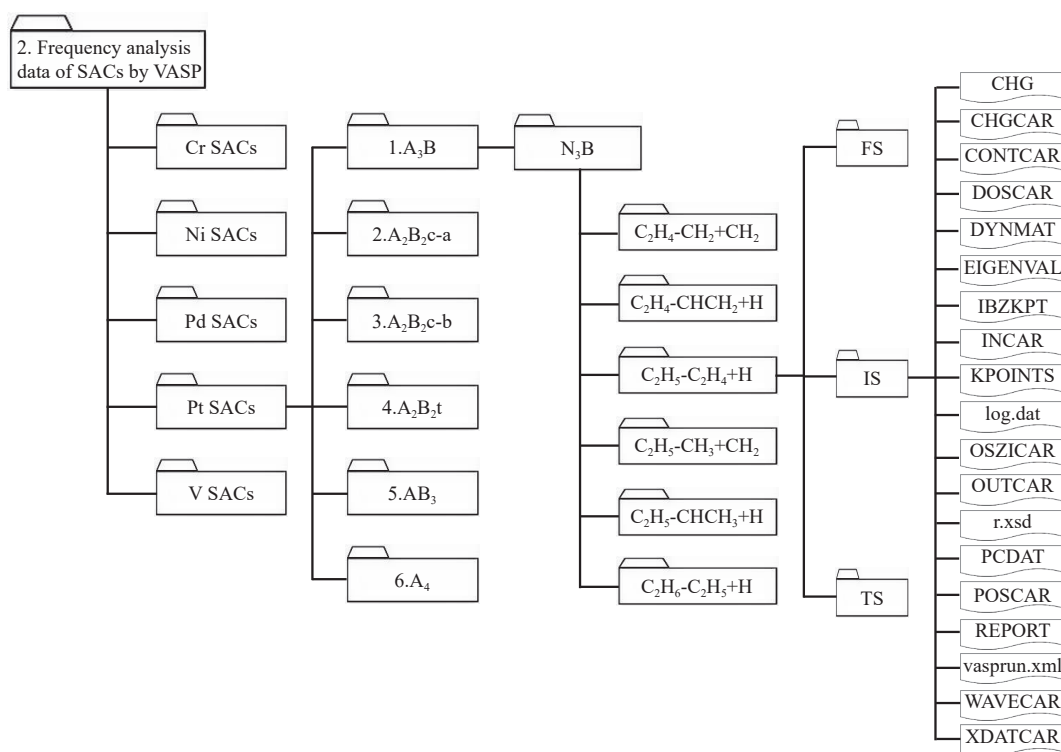


Fig. 3 The file storage directory structure of the Pt-N<sub>3</sub>B SAC in Frequency analysis data of SACs by VASP.zip

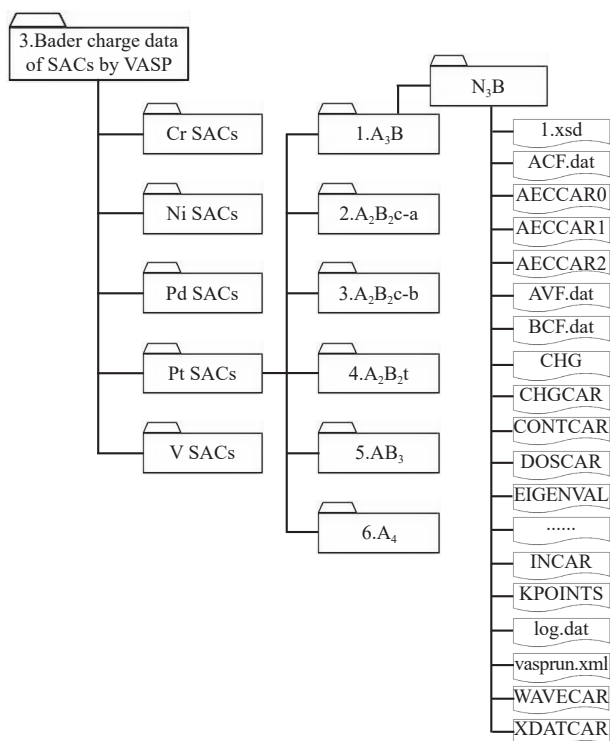


Fig. 4 The file storage directory structure of the Pt-N<sub>3</sub>B SAC in Bader charge data of SACs by VASP.zip

All calculations were performed using first-principles methods within the DFT framework. This theoretical approach offers significant advantages for analyzing material structural stability, evaluating catalytic performance, and interpreting electronic structure. It can accurately describe experimental phenomena and provide

profound theoretical insights into reaction mechanisms.

## 4 Data value

The complete computational source files provided in this database hold significant value for in-depth structural analysis and subsequent research. Researchers can directly access the original files for data interpretation or secondary processing to validate existing conclusions or extract specific physicochemical parameters. Moreover, the standardized and comprehensive calculation files can serve as a basis for further studies, facilitating tasks such as transition-state searches and electronic structure analysis.

Data users can also directly use the pre-optimized structure files in the database to quickly initiate simulations of related properties, substantially improving research efficiency. This database systematically integrates 255 heteroatom-doped graphene-supported SACs, comprising 51 distinct coordination environments with five different metal single atoms. This well-structured dataset can provide reliable structural references and a solid theoretical foundation for future theoretical calculations and experimental explorations.

## 5 Data usage guidelines and availability statement

The files in this database can be directly downloaded and are formatted for compatibility with both MS and VASP software. Due to copyright restrictions on pseudopotential files, they are not included in the distributed package.

All first-principles calculation data presented in this paper—including optimized structures, electronic energies, DOS, Bader charges and frequency analysis results—are stored in the Science Data Bank (SDB) hosted by the Computer Network Information Center, Chinese Academy of Sciences. The dataset is titled “A dataset of heteroatom-doped graphene-supported SACs for EDH” and is assigned the following persistent identifiers: Database DOI:<https://doi.org/10.57760/sciencedb.32498>; Database CSTR:<https://cstr.cn/31253.11.sciencedb.32498>. The dataset is available for access and download upon request via the SDB platform, and may be used for subsequent non-commercial scientific research.

**Example 1 (Exploration of Structure-Performance Relationships):** Researchers can use this database to systematically extract Bader charges or *d*-band centers (which can be computed from the provided DOS files) for the active metal centers across all catalyst systems. These electronic structure descriptors can then be correlated with the overall energy barriers for EDH reaction, helping to reveal universal structure-performance relationships within heteroatom-doped catalytic systems.

**Example 2 (Screening of High-Performance Catalysts):** By screening the database for systems that exhibit both low overall energy barriers for the EDH reaction and a thermodynamic preference for ethylene formation, researchers can rapidly identify several potential high catalytic activity, ethylene selectivity candidate catalysts. For instance, the Pt-N<sub>3</sub>B SAC emerges from such an analysis, providing a clear and promising target for subsequent experimental synthesis and validation.

## 6 Conclusions

Through systematic first-principles calculations, this study constructed a comprehensive dataset of heteroatom-doped graphene-supported SACs for the EDH reaction. It systematically reveals variations in catalytic performance among different heteroatom-doped structures across individual elementary reaction steps. Based on electronic structure analyses involving DOS and Bader charge calculations, the physical mechanisms underlying these performance differences were elucidated at the electronic level, providing a theoretical foundation and screening criteria for the rational design of efficient EDH catalysts. This database not only presents trends in reaction energies but also establishes a structure-performance relationship characterized by “dopant type → electronic state of active metal center → catalytic activity” through its electronic structure data. This deepens the atomic-scale understanding of the EDH reaction mechanism on SACs. The research demonstrates how high-throughput calculations can generate well-structured, information-rich, and mechanistically explicit domain-specific data. To advance data-driven research paradigms, we plan to compile lightweight feature extraction code in future work, enabling the direct export of feature-value tables in .csv format to support the training of various machine-learning models.

Furthermore, we will carry out experimental validation to continuously enrich and enhance the content and reliability of this scientific database. We firmly believe that the value of data lies in its circulation and sharing. Therefore, we fully embrace data openness principles to promote the development of a new data-driven paradigm in the EDH research field.

## Declaration of competing interests

The authors declare no conflict of interest.

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## 杂原子掺杂石墨烯负载金属单原子催化剂上乙烷直接脱氢反应的数据库

曹雅洁，王宝俊，章日光\*

(太原理工大学 省部共建煤基能源清洁高效利用国家重点实验室, 山西 太原 030024)

**摘要:** 单原子催化剂(SACs)在乙烷直接脱氢(EDH)反应中展现出巨大潜力, 这主要得益于接近 100% 的原子利用率、可精准调控的活性位点及优异的催化性能。研究表明, 活性金属的种类、载体特性及配位环境是影响其 EDH 性能的关键因素。其中, 石墨烯因其优异的热稳定性和灵活可调的配位结构, 成为理想的载体材料。虽然已有文献对不同金属类型和配位环境的影响进行了探索, 但由于数据来源分散, 尚未形成系统性的认识规律。针对这一现状, 本研究构建了一个系统全面的石墨烯负载 SACs 数据库, 涵盖 5 种典型金属单原子和 6 大类 51 种不同的配位环境类型。通过高通量计算, 我们系统获取了包括基元反应能量、振动频率、电子态密度和 Bader 电荷在内的多维度数据, 并在参数设置和计算结果两个

层面建立了严格的质量控制体系。该数据库同时提供了完整的原始计算文件,为深入解析杂原子掺杂石墨烯负载 SACs 在 EDH 反应中的催化性能、构效关系和反应机理提供了可靠的数据支撑。

**关键词:** 单原子催化剂; 石墨烯; 杂原子; 乙烷脱氢; 数据库; 第一性原理计算

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#### 作者简介:



曹雅洁(1994), 硕士研究生, 乙烷脱氢反应的多相催化理论研究, caoyajie0168@link.tyut.edu.cn。



章日光(1981), 博士研究生, 教授, 碳基分子转化的多相催化基础理论研究, zhangriguang@tyut.edu.cn。