

A dataset for CO₂ cycloaddition with ethylene oxide over metal oxide catalysts

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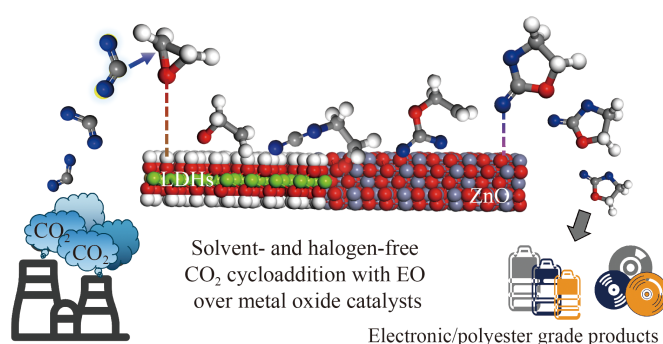
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Abstract: Metal oxide catalysts have emerged as highly promising materials for the CO₂ cycloaddition reaction, owing to their tunable composition, facile separation, reusability and low cost. Previous studies have identified that the type and ratio of metal dopants, surface defect characteristics and crystal plane orientation are critical factors affecting catalytic performance. Despite this potential, systematic investigations into metal oxide catalysts for CO₂ cycloaddition remain limited and a comprehensive understanding

of the underlying reaction mechanisms is hindered by the lack of extensive, well-curated datasets. To address this gap, this study establishes a systematic and comprehensive dataset of metal oxides including layered double hydroxide (LDH) and ZnO catalysts, encompassing variations in metal dopant type and ratio, defect characteristic and crystal plane orientation. Through high-throughput calculations, we have generated a robust multi-dimensional dataset containing elementary reaction energies, vibrational frequencies, Bader charges and density of states. A rigorous two-tiered quality control protocol is applied to both computational parameter settings and output results, ensuring the integrity and reliability of the data. This dataset, providing complete raw calculation files, offers a reliable foundation for exploring catalytic performance, structure-performance relationships and reaction mechanisms of metal oxide catalysts in CO₂ cycloaddition.

Key words: metal oxide; layered double hydroxide; ZnO; CO₂ cycloaddition; dataset; first-principles calculations



CO₂ cycloaddition with ethylene oxide (EO) to produce ethylene carbonate (EC) is a synthetic route of considerable economic and environmental importance^[1]. Currently, homogeneous catalysts such as KI/polyethylene glycol^[2] and quaternary ammonium salts^[3] are widely employed in industrial cyclic carbonate synthesis. However, these homogeneous systems suffer from inherent limitations, including complicated product separation and the presence of residual halogens. Therefore, developing efficient, halogen-free and solvent-free heterogeneous catalysts is critical to overcoming the challenges related to product purification and halogen contamination.

Metal oxides, such as layered double hydroxide (LDH) and ZnO, have attracted extensive attention in CO₂

cycloaddition reactions due to their tunable composition, facile separation, reusability and low cost. However, their relatively low catalytic activity is still challenging^[4–5]. Recent studies have indicated that catalytic performance can be enhanced through strategies such as introducing secondary metal dopants, adjusting dopant types^[6–10] and ratios^[11–12], engineering surface defects^[13–14] and controlling crystal plane orientations^[15–16]. Despite these advances, a systematic and comprehensive theoretical understanding of how these factors—namely dopant types and ratios, defect characteristics and exposed facets—affect the CO₂ cycloaddition mechanism remain limited. This gap hinders the in-depth elucidation of structure-performance relationships in metal oxide catalysts. Thus, systematic

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theoretical investigations encompassing these variables are urgently needed to guide the rational design of high-performance catalysts for CO₂ cycloaddition.

The rapid growth in computing power, coupled with the widespread implementation of high-throughput screening methodologies, has significantly accelerated the development of computational materials science. In particular, high-throughput methods have revolutionized the research paradigm by enabling capabilities analogous to experimental screening and discovery, leading to the establishment of major material databases such as the Materials Genome Project, the Open Quantum Materials Database, and the Materials Project^[17–19]. Nevertheless, a dedicated dataset focused on metal oxide catalysts for CO₂ cycloaddition is currently unavailable. The construction of such a database would not only provide essential data for exploring novel material properties and rationally designing advanced catalysts, but also promote efficient data sharing and reuse within the scientific community. By integrating multidimensional information including atomic structures, electronic properties and computational parameters, researchers can directly validate existing results, apply new analytical methods and perform cross-dataset comparisons, thereby reducing both time and economic costs. Moreover, minimizing redundant calculations at the source would contribute to considerable savings in computational

resources. Standardized and transparent data descriptions would further lower access barriers, facilitating understanding and reuse by scientists from diverse backgrounds and even the broader public, ultimately promoting dual advances in knowledge accumulation and research efficiency.

In this context, establishing a specialized dataset for metal oxide catalysts will provide a data-driven foundation for the rational screening and design of efficient CO₂ cycloaddition catalysts. Such a platform can also serve as a representative model for doped material dataset, and the open sharing of its data is expected to substantially advance research in CO₂ cycloaddition catalysis and related fields.

1 Data source

1.1 Dataset profile

This dataset, entitled “Dataset of CO₂ cycloaddition reactions over metal oxide catalysts” (version V1.0; see Table 1), focuses on the performance and mechanisms of CO₂ cycloaddition reactions over LDH and ZnO systems and integrates comprehensive structural data and computational input/output files for 102 metal oxide catalysts. It is structured into three main components.

Table 1 Basic information of the dataset

Data title	Dataset of CO ₂ cycloaddition reactions over metal oxide catalysts
Data version	V1.0 (released in March 2026)
Software version	VASP5.4.4 for ZnO system, Materials Studio DMol ³ version 8.0 for LDH system
Corresponding author	ZHANG Riguang
Data authors	XI Jianying, WANG Baojun, ZHANG Riguang
Time range	2023–2026
Data volume	264.71 GB, 102264 entries
Data format	*.zip
Data service system	CSTR: 31253.11.sciencedb.30198. https://doi.org/10.57760/sciencedb.30198
Database composition	This database integrates comprehensive structural data and computational input/output files for 102 metal oxide catalysts. It is structured into three main components: DFT calculation data of metal oxide catalysts.zip contains catalyst structures before and after optimization, adsorption configurations of reactants, intermediates, and products, along with input and output files for transition state searches with a data volume of 60.45 GB; Frequency analysis data of metal oxide catalysts.zip contains input and output data from frequency analysis calculations performed using the VASP with a data volume of 82.45 GB; Electronic Properties data of metal oxide catalysts.zip contains input and output data from Bader charge analysis and density of states (DOS) calculations performed using the VASP with a data volume of 121.81 GB.
Citation format of data	XI J Y, WANG B J, ZHANG R G. A dataset for CO₂ cycloaddition with ethylene oxide over metal oxide catalysts[J]. J Fuel Chem Technol, 2026, 54(7): 20260098. When citing this dataset, the associated article(s) must also be cited. XI J, HAN B, WANG B, et al. Halogen-free CO₂ cycloaddition with ethylene oxide on the Modified Zn_xM-LDH Catalysts: The Influences of M Type, Zn/M ratio and defect type on catalytic performance[J]. Sep Purif Technol, 2025, 374: 133676. XI J, GUAN Z, WANG B, et al. Halogen-free CO₂ cycloaddition with ethylene oxide over ZnO and M–ZnO catalysts: Roles of metal dopant and crystal facet in Tuning catalytic performance[J]. Chem Eng Sci, 2025: 122818. XI J Y, WANG B J, ZHANG R G. Database of CO₂ cycloaddition reactions over metal oxide catalysts[DS/OL]. V1. Science Data Bank, 2026[2026-06-10]. https://doi.org/10.57760/sciencedb.30198. DOI:10.57760/sciencedb.30198.

1.1.1 DFT calculation data package

The file “DFT calculation data of metal oxide catalysts.zip” contains catalyst structures before and after optimization, adsorption configurations of reactants, intermediates, and products, along with input and output files for transition state searches. The total size of this dataset is 60.45 GB, comprising approximately 73356 data entries.

1.1.2 Frequency analysis data package

The file “Frequency analysis data of metal oxide catalysts.zip” contains input and output data from frequency analysis calculations performed with VASP. This dataset has a total size of 82.45 GB and consists of approximately 18583 data entries.

1.1.3 Electronic properties analysis data package

The file “Electronic properties data of metal oxide catalysts.zip” contains input and output data from Bader charge analysis and density of states (DOS) calculations performed with VASP. The total size of this dataset is 121.81 GB, with approximately 10325 data entries.

All data are organized and managed according to unified standards within a hierarchical directory structure that follows a clear data architecture; file names reflect the data type, reaction system, and pathway, making the dataset easy to identify and retrieve.

1.2 Structural models

The dataset of metal oxide catalysts is categorized into two primary categories: LDH and ZnO systems. The structural models were constructed using Mg(OH)₂ and ZnO as parent frameworks, respectively, with initial crystallographic parameters obtained from the Materials Project database.

For the LDHs (Figure 1(a)), individual model configurations were generated by systematically varying four key structural descriptors: (i) divalent cation species (M^{2+} =Zn, Mg, Ca), (ii) trivalent cation species (M^{3+} =Fe, Co, Ni), (iii) M^{2+}/M^{3+} molar ratios ranging from 2 to 5 and (iv) surface defect types including oxygen vacancy (O_v) and hydrogen vacancy (H_v). Specifically, $M_x^{2+}M^{3+}$ -LDH ($x=2, 3, 5$) models were constructed by substituting 4, 3, or 2 M^{2+} cations with M^{3+} cations within a single-layer $p(3\times 4)$ supercell, respectively.

For models corresponding to an M^{2+}/M^{3+} ratio of 4 (i.e., $M_4^{2+}M^{3+}$ -LDH), three M^{2+} sites were replaced by M^{3+} in a single-layer $p(3\times 5)$ supercell. Pristine models without surface defects were designated as $M_x^{2+}M^{3+}$. To prevent spurious interactions between periodic slabs along the c-axis, a vacuum layer of 15 Å thickness was introduced

perpendicular to the layer plane.

Furthermore, defective LDH surfaces were generated by introducing specific vacancy defects into the pristine $M_x^{2+}M^{3+}$ models. Hydrogen vacancy models (denoted as $M_x^{2+}M^{3+}-H_v$) were created by removing a single hydrogen atom, while oxygen vacancy models (denoted as $M_x^{2+}M^{3+}-O_v$) were obtained by removing an entire hydroxyl group ($-OH$) from the surface layer. These systematic structural variations enable comprehensive investigation of compositional and defect-dependent catalytic properties.

For ZnO systems, two low-index surfaces were investigated: the nonpolar (10-10) and (11-20) facets. The (10-10) surface was modeled using a three-layer $p(3\times 2)$ supercell, while the (11-20) surface was represented by a four-layer $p(3\times 2)$ supercell. A 15 Å vacuum layer was introduced perpendicular to the surface to eliminate spurious interactions between periodic slabs. Doping was achieved by substituting a single surface Zn atom with a metal dopant M (M=Mg, Ca, Al, Ga, Fe, Co, Ni or Cu) (Figure 1(b)). The pristine and doped models were denoted as PZ1 (ZnO(10-10)), PZ2 (ZnO(11-20)), MZ1 (M-ZnO(10-10)) and MZ2 (M-ZnO(11-20)), respectively.

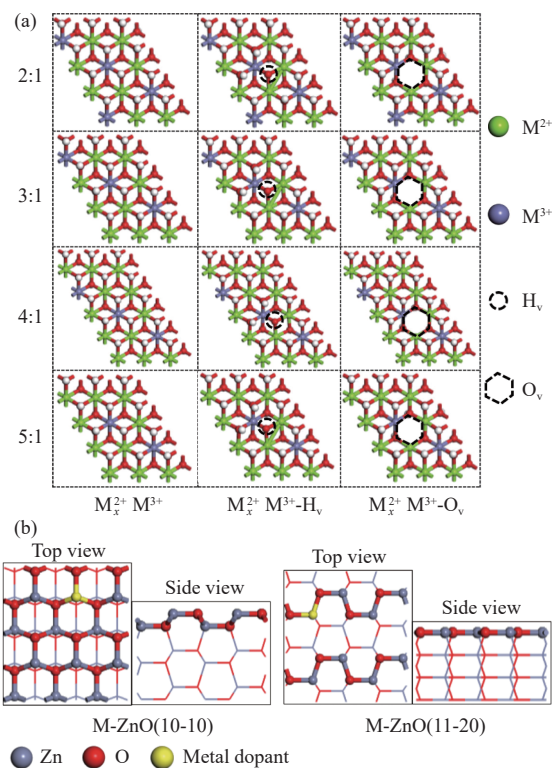


Fig. 1 (a) Top view of $M_x^{2+}M^{3+}$, $M_x^{2+}M^{3+}-H_v$ and $M_x^{2+}M^{3+}-O_v$ models, the green and gray balls represent M^{2+} (Mg, Ca, Zn) and the M^{3+} (Fe, Co, Ni) ions, respectively, (b) Top and side views of M-ZnO(10-10) and M-ZnO(11-20) surfaces, the light, gray and red balls represent the metal dopant M (M=Mg, Ca, Al, Ga, Fe, Co, Ni and Cu), Zn and O atoms, respectively

1.3 Computational details

1.3.1 LDH systems

For LDH systems, Dmol³ code in Materials Studio 8.0^[20] was selected to carry out the high-throughput density functional theory (DFT) calculations based on three considerations: **First**, the Hubbard *U* correction was not applied to the local *d* electrons of Zn in the previous works on similar systems (Zn₃M-LDHs)^[21]. **Second**, LDH catalyst types were numerous and a large number of transition state searches were required in the study of the CO₂ cycloaddition reaction catalyzed by LDHs. **Third**, the Dmol³ module exhibited high computational efficiency and could efficiently search for transition states. The electron exchange-correlation interaction was described using the GGA-PBE functional^[22–23]. Core-electron interaction was described using DFT semi-core pseudopotential^[24]. The inner electrons of Zn, Fe, Co and Ni atoms were frozen and replaced by effective core potential (ECP)^[25], while valence electron wavefunctions were expanded using the double-numeric polarized (DNP) basis set^[26–27]. The van der Waals interaction was corrected *via* the DFT-D method^[28], and a global orbital cutoff was assigned as medium. The Brillouin zone was sampled using a (3×2×1) *k*-point mesh based on the Monkhorst–Pack scheme^[29]. The convergence criteria for the energy, force and displacement were set to 2.0×10^{−5} Ha, 4.0×10^{−3} Ha/Å and 5.0×10^{−3} Å, respectively.

Transition state searches were conducted using the complete LST/QST method^[30–31]. All identified transition states were validated through transition state (TS) confirmation and frequency analysis to ensure the presence of a single imaginary frequency corresponding to the desired reaction coordinate. Free energy corrections were applied using the VASPKit software package^[32] under typical experimental conditions for CO₂ cycloaddition (423.15 K, 1 bar)^[4,33].

1.3.2 ZnO systems

For ZnO systems, previous studies considered *U*-correction^[34–35], while the Dmol³ module lacked this function. Thus, the Vienna Ab initio Simulation Package (VASP) software^[36–37] was employed to ensure calculation accuracy. The GGA-PBE functional was used to describe the electron exchange-correlation^[23]. Given that standard GGA functional inadequately describes the localized *d*-electrons of Zn, a Hubbard *U* correction was applied using the DFT+*U* formalism. The effective *U*-*J* value for Zn was set to 4.7 eV based on previous benchmarks^[38]. The plane-wave basis set cutoff energy was 400 eV. Dispersion interactions were corrected using Grimme's DFT-D3 method^[39]. Brillouin zone sampling was performed with Monkhorst-Pack *k*-point meshes^[29] of 3×2×1 and 2×2×1 for ZnO(10-10) and (11-20) surfaces, respectively. Geometry optimizations were converged until the maximum force was below 0.05 eV/Å. The transition states searches were

carried out using the climbing-image nudged elastic band (CI-NEB) method^[40–41] and the dimer method^[42–43]. All transition states were verified by vibrational frequency analysis to confirm the presence of exactly one imaginary mode along the reaction coordinate. Free energy corrections were similarly applied using VASPKit^[32] at 423.15 K and 1 bar to maintain consistency with the LDH system calculations^[4,33].

Although DMol³ and VASP are selected for DFT calculation on LDHs and ZnO systems, respectively, these two software packages exhibit good comparability in describing material geometric structures. They are consistent in qualitative analysis of energy and electronic properties, but they show significant differences in quantitative results, application scenarios and computational efficiency. In terms of energy, due to the subtle differences in basis sets, pseudopotentials and treatment methods, directly comparing the absolute values of the total energy calculated by DMol³ and VASP is meaningless, but the energy differences (such as reaction energy, adsorption energy) and relative energy trends are highly comparable. In terms of geometric structure, the differences in lattice parameters, atomic positions, bond lengths and bond angles calculated by these two software packages are typically within 1%, due to the use of the same functional (GGA-PBE), thus they are highly comparable. Electronic properties are also highly comparable, as both the LDH and ZnO systems were calculated using VASP.

1.4 High-throughput data management

In the high-throughput computing architecture, all input files, output files and associated metadata were systematically organized and stored within a hierarchical file system. This structured dataset architecture facilitates efficient data retrieval, querying, and reuse, thereby enabling comprehensive analysis and comparison across the entire catalyst space investigated in this work.

2 Data sample description

This dataset systematically encompasses parameter data, structural information, software environment details, and electronic property data for metal oxide catalysts applied in the CO₂ cycloaddition reaction. All calculation files are organized by catalyst type (LDHs and ZnO) within a unified, standardized directory hierarchy.

The data files are structured in a consistent two-level folder system. For LDHs, Level-1 folders are designated by the divalent cation type (e.g., Zn-LDHs, Ca-LDHs and Mg-LDHs), while Level-2 folders are categorized by the trivalent cation type (e.g., Fe, Co and Ni). The M²⁺/M³⁺ ratio and defect type can be identified from the folder naming. For instance, “2Zn-Fe” represents a M²⁺/M³⁺ ratio of 2 and no defects; “2Zn-Fe-H” represents a M²⁺/M³⁺ ratio of 2 with an H_v defect (i.e., one hydrogen atom is removed from the catalyst surface); “2Zn-Fe-OH” represents a M²⁺/M³⁺ of 2

with an O_v defect (i.e., one –OH group is removed from the catalyst surface). A specific example of the search process is presented as **Example 3 (Search Methods)** in Section 5. Taking Zn-LDHs containing Fe with a Zn/Fe ratio of 2 as an example, the file storage structure is illustrated in Figures 2–4. For ZnO systems, Level-1 folders correspond to the crystal facet type—specifically (10-10) and (11-20)—and Level-2 folders to the metal dopant M type (including Mg, Ca, Al, Ga, Fe, Co, Ni and Cu). Using Fe-doped ZnO(11-20) (FeZ2) as a representative case, the corresponding file storage structure is also shown in Figures 2–4. The detailed contents of each data category are as follows:

(1) DFT Calculation Data (Figure 2): The folder “**DFT calculation data of metal oxide catalysts**” provides computational files including geometry optimization of catalyst structures and adsorption configurations (reactants, intermediates and products), as well as transition state search results. This includes data for 84 LDH systems with various divalent (M²⁺) and trivalent (M³⁺) cations, M²⁺/M³⁺ ratios, and defect types, as well as 18 ZnO systems with different crystal plane orientations and metal dopants.

(2) Frequency Analysis Data (Figure 3): The folder “**Frequency analysis data of metal oxide catalysts**” contains frequency analysis results for all 102 metal oxide catalysts. These cover the most stable adsorption configurations of reactants and intermediates, as well as the initial state (IS), transition state (TS) and final state (FS) structures for each elementary reaction step.

(3) Electronic Properties Data (Figure 4): The folder “**Electronic properties data of metal oxide catalysts**” contains the Bader charge analysis and DOS calculation

results for the full set of 102 metal oxide catalysts.

3 Data quality control and evaluation

3.1 Convergence test

To ensure the accuracy and reliability of the computational results, rigorous convergence tests were first conducted on critical parameters affecting the calculation results such as the plane wave cut-off energy (ENCUT) for VASP, global orbital cutoff for Dmol³ and KPOINTS. Generally, the convergence testing serves to establish calculation standard, namely, testing a few representative models to determine calculation parameters such as ENCUT and *k*-point is sufficient, and then be applied these parameters to other systems. Moreover, re-performing convergence tests for every independent model will increase resource consumption by 3–5 times, which is not justifiable in scientific computing. Thus, the convergence tests were conducted a few representative models.

For LDH systems, the convergence tests were performed on the three representative models of Zn₂Fe (2Zn-Fe), Zn₂Fe-H_v (2Zn-Fe-H) and Zn₂Fe-O_v (2Zn-Fe-OH) (Figure 5). As shown in Figures 5(a)–(c), the system energy stabilizes as the global orbital cutoff radius increases. Considering the calculation accuracy and computing resources, a global orbital cutoff radius of 5.0 Å was adopted for all the LDH systems. Under this cutoff, the effect of KPOINTS on the energy was evaluated using the Zn₂Fe catalyst. As shown in Figure 5(d), the energy stabilizes as the *k*-point grid is gradually refined. Thus, a 3×2×1 *k*-point was applied to all LDH systems in this study.

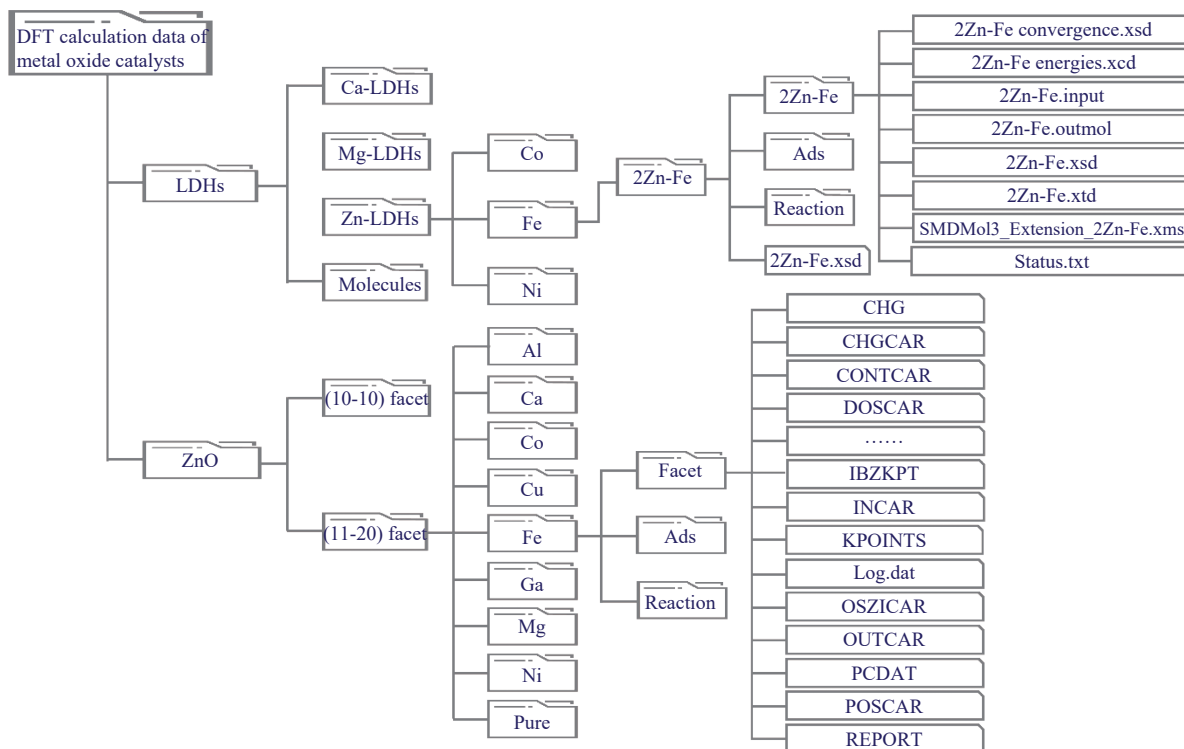


Fig. 2 The file storage directory structure of the Zn₂Fe and FeZ2 in DFT calculation data of metal oxide catalysts

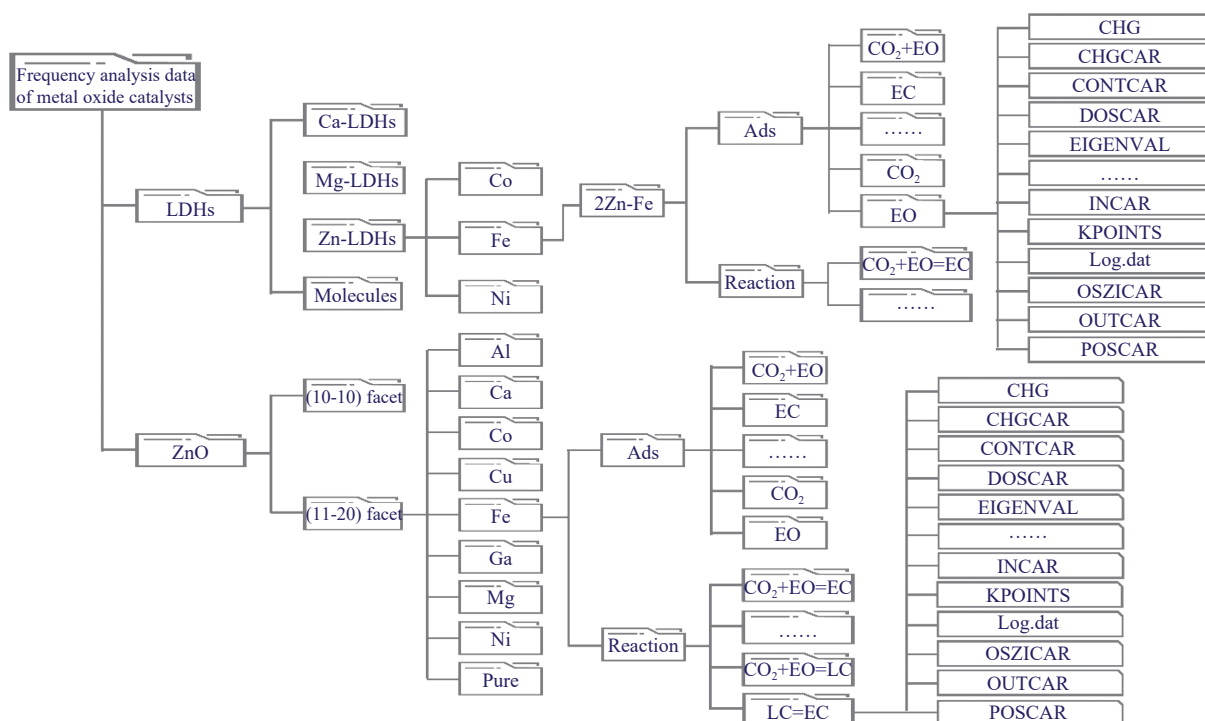


Fig. 3 The file storage directory structure of the Zn₂Fe and FeZ₂ in frequency analysis data of metal oxide catalysts

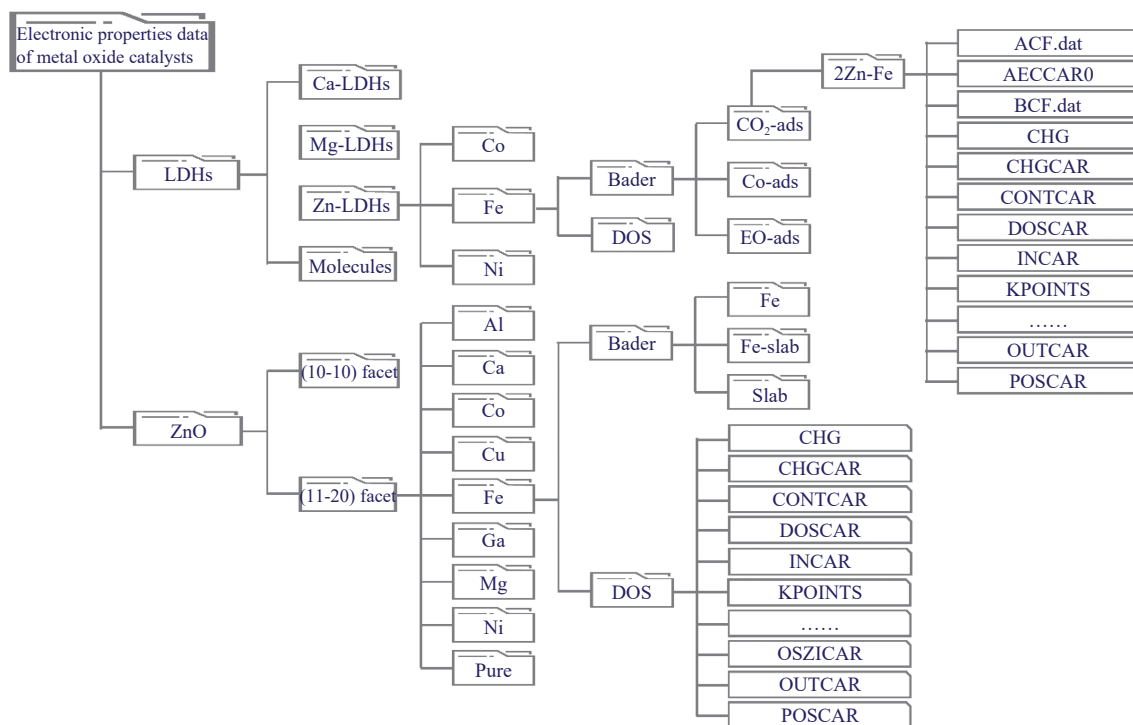


Fig. 4 The file storage directory structure of the Zn₂Fe and FeZ₂ in electronic properties data of metal oxide catalysts

For ZnO systems, the convergence tests were carried out the PZ1(ZnO(10-10)) and PZ2(ZnO(11-20)) (Figure 6). As shown in Figures 6(a), (b), the energy change between adjacent test points stabilizes when ENCUT increases to 350 eV. To balance calculation accuracy and computing resources, an ENCUT of 400 eV was used for all ZnO

systems in this study. Based on this cutoff, we further explored the influence of KPOINTS on the energy. As shown in Figures 6(c), (d), the energy tends stabilizes with increasing the k -point density. Thus, the $3 \times 2 \times 1$ and $2 \times 2 \times 1$ k -point for ZnO(10-10) and ZnO(11-20) are employed in this study, respectively.

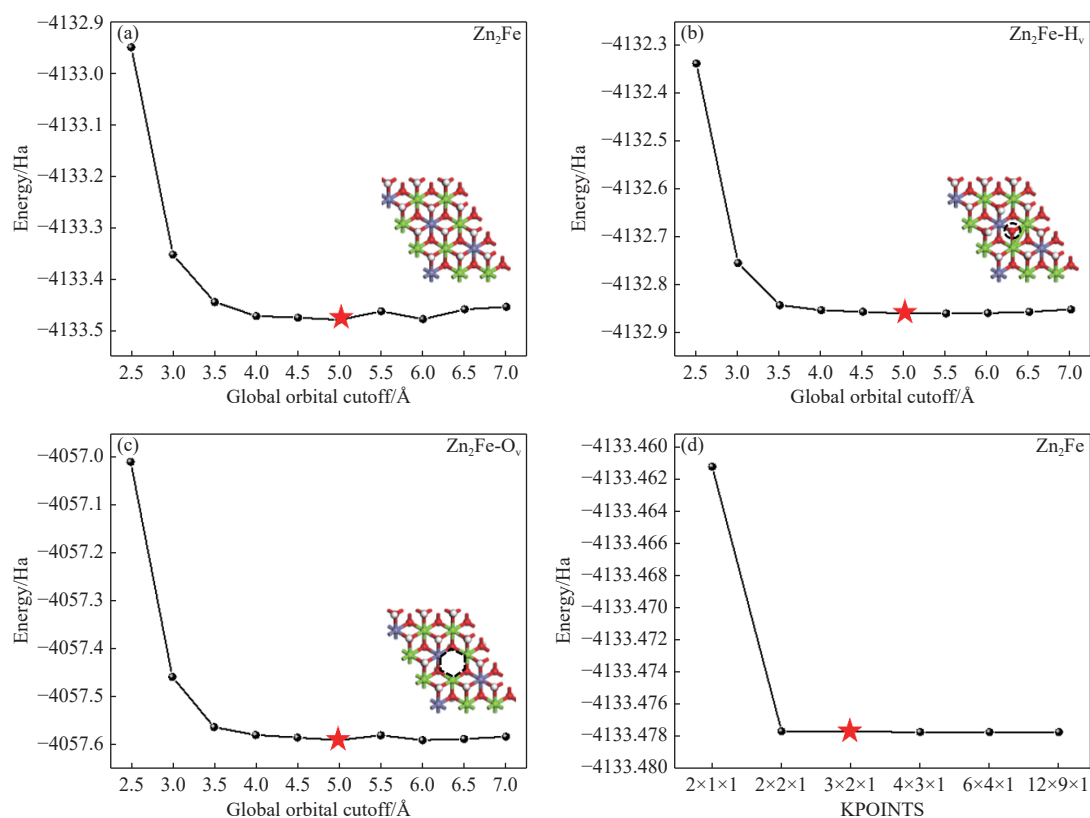


Fig. 5 The relationship between global orbital cutoff radius and energy for (a) Zn₂Fe, (b) Zn₂Fe-H_v and (c) Zn₂Fe-O_v catalysts; (d) The relationship between *k*-point and energy for Zn₂Fe catalyst

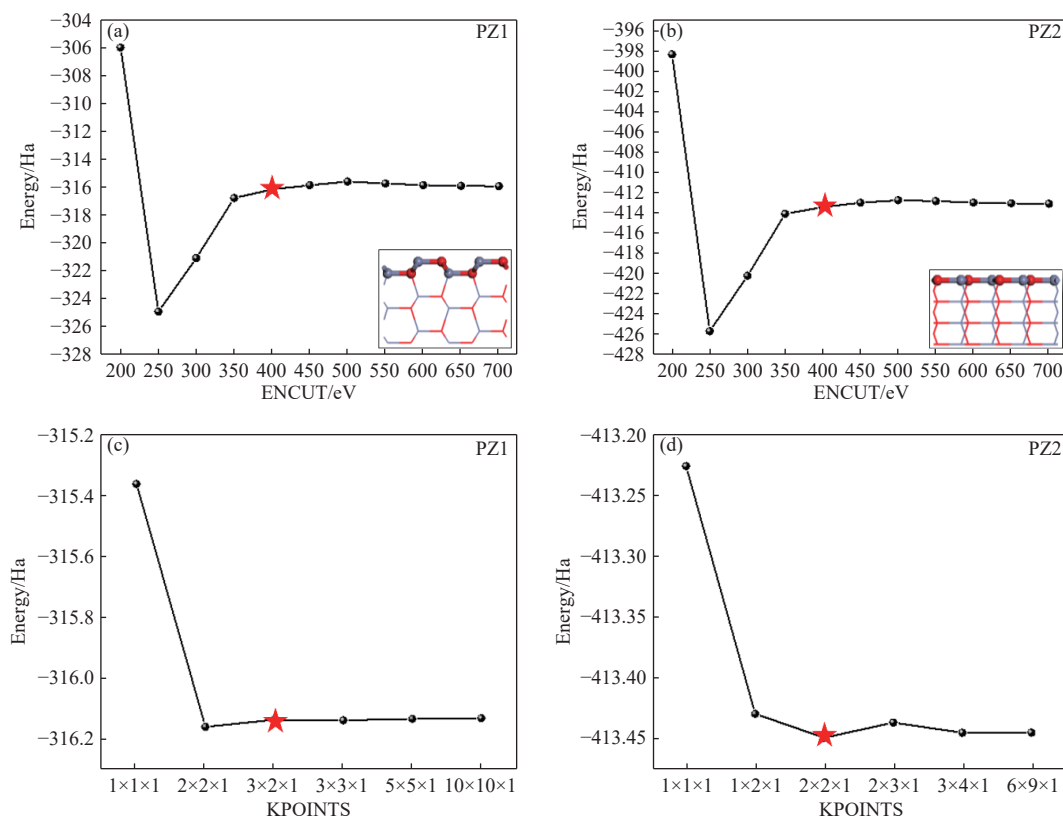


Fig. 6 The relationship between cut-off energy and energy for (a) PZ1 and (b) PZ2 catalysts; (c) The relationship between *k*-point and energy for (c) PZ1 and (d) PZ2 catalysts

It is noted that the separate convergence verification is not required for the Bader charge and density of states calculations, as their accuracy depends on the high-precision self-consistent field (SCF) calculations used to generate charge density files (including CHG and CHGCAR). Additionally, the parameters used in these calculations have already passed the convergence tests during the preprocessing stage.

Based on the convergence tests described above, the numerical stability and reliability of the computed results are ensured. The estimated numerical error for the relative energies is within ± 0.15 eV.

3.2 Convergence criterion

The convergence criterion is a standardized definition of the computational process. In high-throughput computing, multiple systems correspond to the different computing software, such as VASP, Material studio, CP2K. Thus, adopting the strict convergence criteria for energy, force and displacement is necessary during the process of structural optimization and TS search to ensure that each parameter meets the required accuracy thresholds, as different software packages have distinct convergence mechanisms.

For Dmol³ module (LDH systems), its convergence criteria including energy, force, and displacement are defined in the calculation task settings. After completing the calculation, verify whether these values in the last step of the output file (.outmol) have fallen below the threshold set initially. When the prompt “Message: DMol³ job finished successfully” appears, indicating that the calculation has successfully converged. For VASP software package (ZnO systems), confirm whether the electron step (EDIFF) and ion step (EDIFFG) have less than the thresholds set in the input file (INCAR) by checking the output files (OUTCAR and OSZICAR). In the OUTCAR file, if the sentence “reached required accuracy-stopping structural energy minimization” appears, indicating that the calculation has reached the convergence standard. Alternatively, you can check the last line of the OSZICAR file to estimate whether the energy change value (dE) in has already less than the threshold. If so, the electron step has converged. Additionally, the “!” at the end of each line indicates that the ion step calculation has completed.

3.3 Handling failed tasks

In high-throughput computing workflow, computational failures are inevitable. The calculation failure is attributed to two type issues: code errors (such as geometric structure problems, non-convergence of electronic steps, and exhaustion of computing resources or insufficient memory) and abnormal calculation results (such as structural distortion and significant changes in magnetic moment). For geometric structure issues, first determine whether the problem origin from an unreasonable calculation model. If

so, reconstruct a reasonable model and recalculate. If not, the relative value is calculated based on the pristine model energy obtained through self-consistent calculation, thereby eliminating the influence of structural distortion on the calculation results accuracy. The non-convergence of the electronic step is the most common failure. This issue can be solved by adjusting the calculation parameters (such as increasing MIXING, improving the accuracy of ALGO or increasing NELMIN) or gradually enhancing the calculation accuracy through multiple relaxations. When computing resources are exhausted or memory is insufficient, these issues can be addressed by increasing the number of convergence steps or expanding the memory capacity. In case of substantial alterations to the magnetic moment, it is advisable to first perform non-spin-polarized calculations to obtain a stable charge density, and then use this as a basis for spin-polarized calculations. All files presented in this dataset are correct.

3.4 Identification and handling of outliers

The successful execution of a task does not guarantee the generation of rational data. Thus, the original data must be statistically and physically cleaned before storing or analyzing. For the outliers, we first check whether the final energy and force have truly converged to the set threshold. If the convergence criteria are not met and the number of force convergence steps has been reached, we increase the number of convergence steps and recalculate. If the convergence standard is met, we further examine whether structural distortion has occurred. If so, we will calculate the relative value based on the model energy obtained from self-consistent calculation. If not, we recalculate at the original or a similar point.

In summary, the combination of strict convergence test and control, along with the handling of the failed tasks and outliers, ensures that the data presented in this work are both physically meaningful and directly comparable with experimental results, thereby offering reliable theoretical insights into the CO₂ cycloaddition reaction over metal oxide catalysts.

4 Data value

All calculations in this dataset were performed using first-principles methods within the DFT framework. This theoretical approach offers significant advantages in analyzing structural stability, evaluating adsorption ability and catalytic performance, and interpreting electronic properties. It not only provides theoretical support for experimental observations but also offers deeper insights into the underlying reaction mechanisms.

Based on this reliable theoretical foundation, the dataset integrates 102 metal oxide catalysts, including 84 LDH structures with varying divalent (M^{2+}), trivalent (M^{3+}) cations, M^{2+}/M^{3+} ratios and defect types, as well as 18 ZnO

systems with different crystal plane orientations and metal dopants. It provides complete calculation source files, which 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 foundation for extended investigations, enabling advanced tasks such as transition state searches and detailed electronic structure analysis. Data users may also directly utilize the pre-optimized structural data available in the dataset to efficiently initiate simulations of other relevant properties, thereby significantly enhancing research productivity. This dataset systematically integrates 102 metal oxide catalysts, which provides 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 dataset are available for direct download and are compatible with computational formats used by Materials Studio and VASP software. Due to copyright restrictions, pseudopotential files are not included in the package. All first-principles calculation data (with no external experimental data are used for comparison) presented in this paper—including structure optimization, frequency analysis, Bader charges analysis and DOS calculations—are stored in the Science Data Bank (SDB) operated by the Computer Network Information Center, Chinese Academy of Sciences. The dataset is titled “Dataset of CO₂ cycloaddition reactions over metal oxide catalysts” with the following identifiers: Dataset DOI:<https://doi.org/10.57760/sciencedb.30198>; Dataset CSTR:<https://cstr.cn/31253.11.sciencedb.30198>. The dataset is accessible for download upon request *via* the platform and may be used for subsequent research.

Example 1 (Screening of High-Performance Catalysts): By screening systems with low overall energy barriers for CO₂ cycloaddition reaction, several promising high-activity catalysts can be rapidly identified, offering clear targets for experimental synthesis.

Example 2 (Exploration of Structure-Performance Relationships): Researchers can systematically extract data from this dataset, such as Bader charge transfer of EO or CO₂, *p*-band centers of O atoms, and *d*-band centers of metal atoms across all systems, and correlate these descriptors with the overall energy barriers for CO₂ cycloaddition.

Example 3 (Search Methods): Researchers can search the specific cases according to the folder structure. **First**, select the corresponding compressed package (including DFT calculation data of metal oxide catalysts.zip, frequency analysis data of metal oxide catalysts.zip and electronic

properties data of metal oxide catalysts.zip) based on the calculation aims and enter this major category folder. **Second**, within the major category folder, locate the target according to the required catalyst type (including LDHs and ZnO). **Third**, after entering the specific catalyst folder, then enter the corresponding subfolder according to the divalent cation type (e.g., Zn-LDHs, Ca-LDHs and Mg-LDHs) for LDHs or the crystal facet type (e.g., (10-10) and (11-20)) for ZnO. **Finally**, within this folder, navigate to the corresponding subfolder according to the trivalent cation type (e.g., Fe, Co and Ni) for LDHs, or the metal dopant type (e.g., Mg, Ca, Al, Ga, Fe, Co, Ni and Cu) for ZnO. It is noted that the M²⁺/M³⁺ ratio and defect type can then be identified from the folder naming for LDH systems.

6 Conclusions

This study employs first-principles calculations to construct a comprehensive dataset of 102 metal oxide catalysts for the CO₂ cycloaddition reaction. Leveraging these data, we systematically reveal how variations in metal dopant types and ratios, surface defect types, and the crystal plane orientations affect catalytic performance across elementary reaction steps. Electronic structure analysis—including Bader charge and DOS analysis—elucidate the physical mechanisms underlying performance differences at the electronic level, thereby providing a theoretical foundation and screening criteria for the rational design of efficient metal oxide catalysts for CO₂ cycloaddition.

This dataset not only presents trends in reaction energies, but also establishes a clear structure-performance relationship linking catalyst structural features (e.g., metal dopant types and ratios, surface defect types, and the crystal plane orientations) to electronic descriptors (e.g., Bader charge transfer of EO or CO₂, *p*-band centers of O atoms, *d*-band centers of metal atoms), and ultimately to catalytic activity. This deepens the atomic-scale understanding of the CO₂ cycloaddition reaction mechanism. This work demonstrates how high-throughput calculations can generate domain-specific data that is both information-rich and mechanistically explicit.

To further advance data-driven research paradigms, future efforts will focus on developing feature extraction code that is easily integrated, deployed, and cross-platform. This will facilitate the generation of well-structured and readily exchangeable data files, thereby reducing computing resources consumption, enhancing efficiency and portability, and enabling seamless integration of the resulting feature tables with mainstream machine learning frameworks and algorithm libraries. Furthermore, this scientific dataset will be continuously enriched through experimental validation and will remain openly accessible. We firmly believe that data realizes its full value only through circulation and sharing, and we anticipate that this openness will catalyze a new data-driven paradigm in CO₂ cycloaddition research.

Declaration of Competing Interest

The authors declare no conflict of interest.

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金属氧化物催化剂上 CO₂ 与环氧乙烷环加成反应的数据库

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摘要: 金属氧化物催化剂由于其成分可调、易于分离、可重复使用和成本低等优势, 在 CO₂ 环加成反应中具有广泛的应用前景。研究表明, 金属掺杂剂的种类和比例、表面缺陷类型以及晶面取向是影响催化性能的关键因素。尽管该类催化剂在 CO₂ 环加成反应中表现出应用潜力, 但相关基础数据的匮乏严重制约了对其反应机制的深度解析与全面理解。针对这一现状, 本研究建立了一个系统且全面的金属氧化物催化剂(水滑石和氧化锌)数据库, 包括了金属掺杂剂类型和比例、缺陷类型和晶面取向变化等多维信息。基于高通量计算, 获得了一个涵盖基元反应能量、振动频率、巴德电荷和态密度在内的多维数据集, 并构建了贯穿参数设置与计算结果两个层面的严格质量控制体系。该数据库提供了完整的原始计算文件, 为深入探究金属氧化物催化剂在 CO₂ 环加成中的催化性能、结构-性能关系和反应机制提供了可靠且关键的数据支撑。

关键词: 金属氧化物; 水滑石; 氧化锌; CO₂ 环加成; 数据库; 第一性原理计算

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