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Few-shot learning for screening 2D Ga₂CoS_{4-x} supported single-atom catalysts for hydrogen production

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ABSTRACT

Hydrogen generation and related energy applications heavily rely on the hydrogen evolution reaction (HER), which faces challenges of slow kinetics and high overpotential. Efficient electrocatalysts, particularly single-atom catalysts (SACs) on two-dimensional (2D) materials, are essential. This study presents a few-shot machine learning (ML) assisted high-throughput screening of 2D septuple-atomic-layer Ga₂CoS_{4-x} supported SACs to predict HER catalytic activity. Initially, density functional theory (DFT) calculations showed that 2D Ga₂CoS₄ is inactive for HER. However, defective Ga₂CoS_{4-x} ($x = 0-0.25$) monolayers exhibit excellent HER activity due to surface sulfur vacancies (SVs), with predicted overpotentials (0–60 mV) comparable to or lower than commercial Pt/C, which typically exhibits an overpotential of around 50 mV in the acidic electrolyte, when the concentration of surface SV is lower than 8.3%. SVs generate spin-polarized states near the Fermi level, making them effective HER sites. We demonstrate ML-accelerated HER overpotential predictions for all transition metal SACs on 2D Ga₂CoS_{4-x}. Using DFT data from 18 SACs, an ML model with high prediction accuracy and reduced computation time was developed. An intrinsic descriptor linking SAC atomic properties to HER overpotential was identified. This study thus provides a framework for screening SACs on 2D materials, enhancing catalyst design.

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1. Introduction

Significant attention is being directed toward designing energy storage and conversion systems that are both cost-effective and eco-friendly, with high efficiency [1,2]. This interest stems from the potential of water electrolyzers and metal-air batteries to facilitate the clean and sustainable use of molecular hydrogen (H₂). The operation of these systems depends on the oxygen evolution reaction (OER) and the hydrogen evolution reaction (HER), which are two key half-reactions [3–5]. The HER holds particular significance within these electrochemical systems, and conventional electrocatalysts like platinum and noble metal oxides have been widely utilized in this regard [6–8]. However, the applicability of these precious metal group elements is hindered by their high cost and limited availability, making it necessary to explore alternative catalysts [9–11]. Extensive research is currently being carried out to develop more cost-effective catalysts, which involves the development of non-metal catalysts and alloys made from economically

viable elements. Moreover, ongoing studies focus on decreasing the size of precious metal components to increase the active sites within the catalyst, thereby improving catalyst efficiency [12–14]. The main objective of these ongoing studies is to create efficient and affordable catalysts for the HER that can overcome the limitations associated with traditional precious metal-based catalysts. By achieving this, the practical implementation of hydrogen production technologies can be achieved, thereby advancing clean and sustainable energy solutions. These innovations will be pivotal in facilitating the broad adoption of H₂ as a clean energy source, ultimately contributing to the realization of a sustainable and environmentally friendly future.

Two-dimensional (2D) materials have garnered significant interest recently due to their exceptional properties, including a large surface area and unique electronic structures. This class of materials includes various compounds such as 2D transition-metal dichalcogenides [15,16], MXenes [17–20], metal-organic frameworks (MOFs) [21–23], carbon-based nanomaterials, and others [24–27]. Despite extensive research into known 2D materials, their performance in the HER is still not comparable to that of the noble metal Pt. Thus, it is imperative to find new 2D catalysts that can match or exceed the HER activity of Pt, particularly for the

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sustainable production of hydrogen through water electrolysis. Commercial Pt/C, one of the most frequently used Pt-based HER catalysts, exhibits an overpotential of approximately 50 mV in acidic electrolytes, but its overpotential is significantly higher in alkaline electrolytes, highlighting the inherent limitations of Pt in such environments [7,28–30]. Recently, a novel category of 2D materials, denoted as MA_2Z_4 , was computationally designed and experimentally synthesized on Cu, Mo, and W bilayer surfaces [31–35]. Within this framework, M, A, and Z represent specific elements, namely Si/Ga, metal (Mo/W/Co), and N/S, respectively. Ga_2CoS_4 with the monolayer structure, consisting of seven atomic layers arranged as S–Ga–S–Co–S–Ga–S, exhibits remarkable mechanical properties and stability under ambient conditions [36]. However, despite these notable characteristics, the catalytic properties of this newly discovered 2D material remain largely unexplored.

To mitigate the previously mentioned limitations, we present an innovative approach that merges density functional theory (DFT) calculations with machine learning (ML) to speed up the rational design of SACs on a 2D $\text{Ga}_2\text{CoS}_{4-x}$ substrate for HER. Initially, we examined the structural stability and electronic properties of a 2D Ga_2CoS_4 monolayer in the presence and absence of sulfur vacancies. Subsequently, we calculated the decomposition energies for 18 prominent transition metals, covering the 3d group (Co, Cr, Cu, Fe, Mn, Ni, Ti, and V), 4d group (Ag, Mo, Pd, and Ru), and 5d group (Au, Ir, Pt, Re, Ta, and W), specifically looking at the energy required to dissociate a single atom from a metal nanoparticle. Subsequently, DFT calculations were conducted to assess the HER catalytic efficacy of SACs composed of these 18 metal atoms embedded on the 2D $\text{Ga}_2\text{CoS}_{4-x}$ ($x = 0\text{--}0.25$) surface. The resulting data from these calculations formed the basis for training and testing an ML algorithm aimed at predicting the HER catalytic performance of SACs made from the examined transition metals. An intrinsic descriptor was developed to assess the catalytic efficiency of SACs on the 2D $\text{Ga}_2\text{CoS}_{4-x}$ substrate for HER. The integration of ML into this process significantly reduces computational demands, allowing for more rapid and precise predictions of catalytic performance compared to traditional DFT methods. This advanced methodology not only enhances mechanistic understanding but also provides quantitative recommendations for the selection of appropriate transition metals and the optimization of experimental conditions to develop SACs with desired electrocatalytic properties. This strategy, known for its cost-efficiency and high predictive accuracy, can be used in other domains of energy materials design.

2. Computational details

2.1. DFT calculations

The electronic and catalytic properties were analyzed using first-principles calculations, employing the plane-wave basis set and the projector augmented wave (PAW) method within the DFT framework [37]. The exchange-correlation effects were treated using the generalized gradient approximation (GGA) with the Perdew–Burke–Ernzerhof (PBE) functional, as implemented in the Vienna Ab initio simulation package (VASP) [38]. Charge transfer was analyzed using the Bader method [39]. The strong on-site Coulomb interaction of localized *d*-electrons in transition metals (TMs) was addressed using the Hubbard model, commonly referred to as the PBE+*U* method [40]. The *U* values for the *d*-orbital of Co-atom were assigned as 3.30 eV, while for other TMs, the values were adopted from those reported in the literature [41–43]. A plane-wave energy cutoff of 520 eV was set, ensuring that the total energy was minimized until variations were less than 10^{-5} eV between iterations, and forces were converged to within

10^{-2} eV/Å. For *k*-point sampling, a $15 \times 15 \times 1$ Γ -centered grid was applied. Gaussian smearing with a width of 0.20 eV was used for the density of states calculations. To evaluate thermodynamic stability, we analyzed phonon dispersion and vibrational properties using a $6 \times 6 \times 1$ supercell within the density functional perturbation theory framework in conjunction with Phonopy [44–46]. Additionally, we conducted Ab-initio molecular dynamics (AIMD) simulations within a canonical ensemble (NVT, where the number of particles, volume, and temperature is conserved), employing a time step of 2 fs throughout 10 ps at a temperature of 300 K [47,48].

2.2. Few-shot learning algorithm

A machine learning technique is implemented to establish a regression correlation between structural and electronic parameters with the overpotential for hydrogen adsorption, leveraging a dataset from DFT computations. The machine-learning model is constructed to forecast the HER catalytic performance of SACs on 2D $\text{Ga}_2\text{CoS}_{4-x}$ substrate. The model architecture features two main components: an embedding module for data integration and a convolutional neural network (CNN) for predicting the overpotential η^{HER} . The CNN includes an input layer for query-support data, seven hidden layers (five convolutional and two fully connected layers), and an output layer for η^{HER} prediction. The activation function for each hidden layer is the rectified linear unit (ReLU), defined as $f(x) = \max(0, x)$, where *x* is the output of the hidden layer. The dataset is partitioned into training and testing subsets, with the support set chosen based on extreme data values. Each training iteration involves concatenating each query data point (1×5) with the entire support set to generate a new input tensor (4×5). The model is trained using the mean square error (MSE) loss function to minimize prediction errors.

3. Results and discussion

3.1. Structure, stability, and HER catalytic activity of 2D $\text{Ga}_2\text{CoS}_{4-x}$

The three-dimensional (3D) bulk Ga_2CoS_4 material exhibits a standard layered configuration where van-der-Waals (vdW) forces connect the individual layers. Fig. 1(a) illustrates the optimal structure of 3D Ga_2CoS_4 , with the computed lattice constants being $a = b = 3.67$ Å and $c = 12.26$ Å. These parameters align with previous findings [36] ($a = b = 3.65$ Å and $c = 12.06$ Å), thereby validating the theoretical calculations conducted in this study. The initial structure of the 2D single-layer Ga_2CoS_4 was derived from a recently developed 2D materials database [49]. Upon local and global optimization, the optimized lattice parameters for the 2D Ga_2CoS_4 monolayer were determined to be $a = b = 3.66$ Å, as shown in Fig. 1(b). The similarity in lattice constants between 2D and 3D Ga_2CoS_4 suggests weak layer interactions, facilitating easy exfoliation of the 2D single layer, much like the 2D graphene nanosheet. Fig. 1(b) depicts a top and side view of a 2D Ga_2CoS_4 monolayer, which forms a sandwich-like arrangement where a CoS_2 monolayer is enclosed by two GaS monolayers. Each sulfur atom on the outer surface is bonded to three gallium atoms, whereas the sulfur atoms in the inner layer are bonded to three cobalt atoms and one gallium atom. Another crucial characteristic is the electrostatic potential, shown in the right panel of Fig. 1(b), which is flat in the vacuum region and symmetrical around the cobalt layer.

The electronic properties of the 2D Ga_2CoS_4 monolayer were calculated and are shown in Fig. 1(c). The material exhibits a metallic nature, with states near the Fermi-level primarily arising from the 3*d*-orbital of cobalt and 3*p*-orbital of sulfur, and to a lesser extent from the 3*d*-orbital of gallium. According to the projected

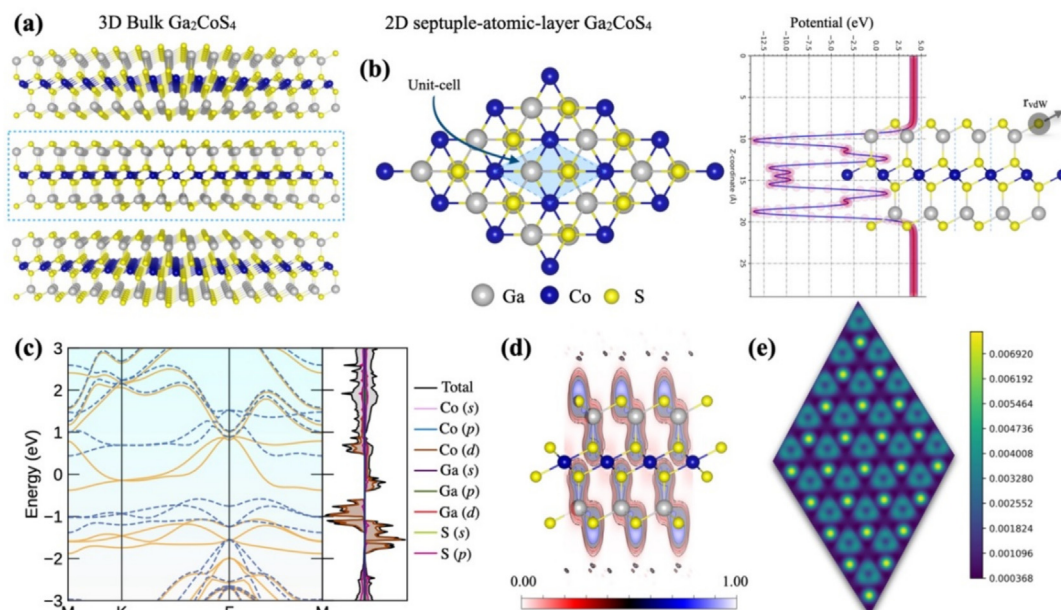


Fig. 1. Detailed analysis of the structure and electronic properties of 3D bulk and 2D monolayer Ga_2CoS_4 . (a) The optimized crystal structure of 3D bulk Ga_2CoS_4 . (b) Top and side views of the exfoliated 2D Ga_2CoS_4 monolayer along with its potential average. (c) Spin-polarized band structure and projected density of states for the 2D monolayer Ga_2CoS_4 . (d) Computed localized charge density profile along the z -direction. (e) Computed charge density of the 2D Ga_2CoS_4 monolayer at the Fermi level, measured to be approximately 12.7 Å above the surface plane, with values ranging from 3.6×10^{-4} to 7.6×10^{-4} electrons $\text{\AA}^{-3}$. The highest values of the Fermi level are located at the atomic sites. The STM image displays the charge density at the Fermi level.

density of states (DOS) for Ga_2CoS_4 , along with Co and S1 atoms (where S1 is bonded to both Co and Ga, and S2 is bonded only to Ga), and when compared with the projected band structure and DOS through PBE+ U method, it is found that the two bands crossing the Fermi level are mainly derived from the spin-up electrons of Co and S1 atoms (Fig. S1). This metallic characteristic makes 2D Ga_2CoS_4 monolayer suitable as a vector for energy-related devices. Fig. 1(d) illustrates the computed localized charge density of Ga_2CoS_4 , where negatively charged S-atoms are encircled by positively charged Ga- and Co-atoms, indicating charge transfer from Ga- and Co-atoms to S-atoms. The Bader charge analysis depicts that each S-atom gains about 2.17e and 1.24e from the adjacent Ga- and Co-atom in 2D Ga_2CoS_4 , respectively. Additionally, the scanning tunneling microscopy (STM) image shown in Fig. 1(e) was generated using first-principles methods to assist experimental research. This STM image clearly shows the atomic structure, with S-atoms appearing brighter than Ga- and Co-atoms. To assess the thermal and dynamic stability of the 2D Ga_2CoS_4 monolayer, phonon dispersion calculation and AIMD simulation were conducted at 300 K for 10 ps, as depicted in Fig. S2(a and b). The finding indicates that the phonon shows positive frequencies, and the structure remains intact after the simulation, confirming the high stability of the 2D Ga_2CoS_4 monolayer. Furthermore, the combination of stable phonon modes, robust structural integrity under thermal conditions, and the favorable atomic arrangement suggests that the synthesis of this monolayer could be achievable using contemporary techniques such as chemical vapor deposition (CVD) or molecular beam epitaxy (MBE). Notably, these methods have been successfully employed in the synthesis of other 2D materials within the same family, such as the layered MoSi_2N_4 , as reported by Hong et al. [50]. The use of Ga, Co, and S as constituent elements, which are commonly utilized in the thin-film fabrication, further supports the feasibility of experimentally realizing the 2D Ga_2CoS_4 monolayer.

Furthermore, to explore the HER activity on the pristine and defective 2D $\text{Ga}_2\text{CoS}_{4-x}$ monolayer, a 3×3 supercell of pristine Ga_2CoS_4 monolayer is initially constructed and three distinct

adsorption sites are examined for H-atom adsorption, namely on a Ga-atom (T_{Ga}), on an S-atom (T_{S}), and in the center of the Ga-S hexagonal ring (H). Full optimization revealed that the S atom site was the most energetically favorable for hydrogen adsorption, forming an S–H bond with a length of 1.052 Å. The free energy variation (ΔG_{*H}) was computed to be about 2.45/2.55/2.71 eV for $T_{\text{S}}/T_{\text{Ga}}/H$, respectively, indicating thermodynamically unfavorable H-adsorption on the pristine 2D Ga_2CoS_4 monolayer and thereby suggesting that the pristine 2D Ga_2CoS_4 is highly inert to H-adsorption and is not a viable catalyst of HER. Subsequently, we examined the catalytic activity for hydrogen adsorption on the 2D Ga_2CoS_4 monolayers with surface sulfur vacancies (SV). Different defective configurations were generated by removing a S-atom from the surface of various supercells: 2×2 , 2×3 , 3×3 , 3×4 , 4×4 , and 5×5 . These configurations resulted in SV concentrations of 12.5%, 8.3%, 5.6%, 4.2%, 3.1%, and 2.0%, respectively (Fig. S3). For illustration, we focus on the relaxed structure of the 3×3 supercell of 2D Ga_2CoS_4 supercell with one surface SV, as shown in Fig. 2(a), since other SV concentrations yield similar structures. Fig. 2(a) demonstrates that removing a surface S-atom prompts the three adjacent Ga-atoms to move slightly toward the SV center. In the 2D materials family, the occurrence of surface atom vacancies is almost inevitable. This occurs mainly because the structural disorder caused by defects increases entropy, which generally correlates with a reduction in the thermodynamic stability of the material. Nonetheless, some defects might be thermodynamically advantageous under certain conditions [51–53]. We have also assessed the challenge of introducing sulfur vacancy by calculating formation energy as detailed in the Supporting Information (SI, Appendix S1). As summarized in Table 1, the formation energies calculated for S-rich conditions range from 6.090 to 6.343 eV with increased C_{SV} . This increase is anticipated because materials with more vacancies tend to have higher energy levels, which are associated with reduced thermodynamic stability. Conversely, under S-poor conditions, the values shift from –2.405 to –1.802 eV, indicating a strong potential for introducing surface SVs in such an environment.

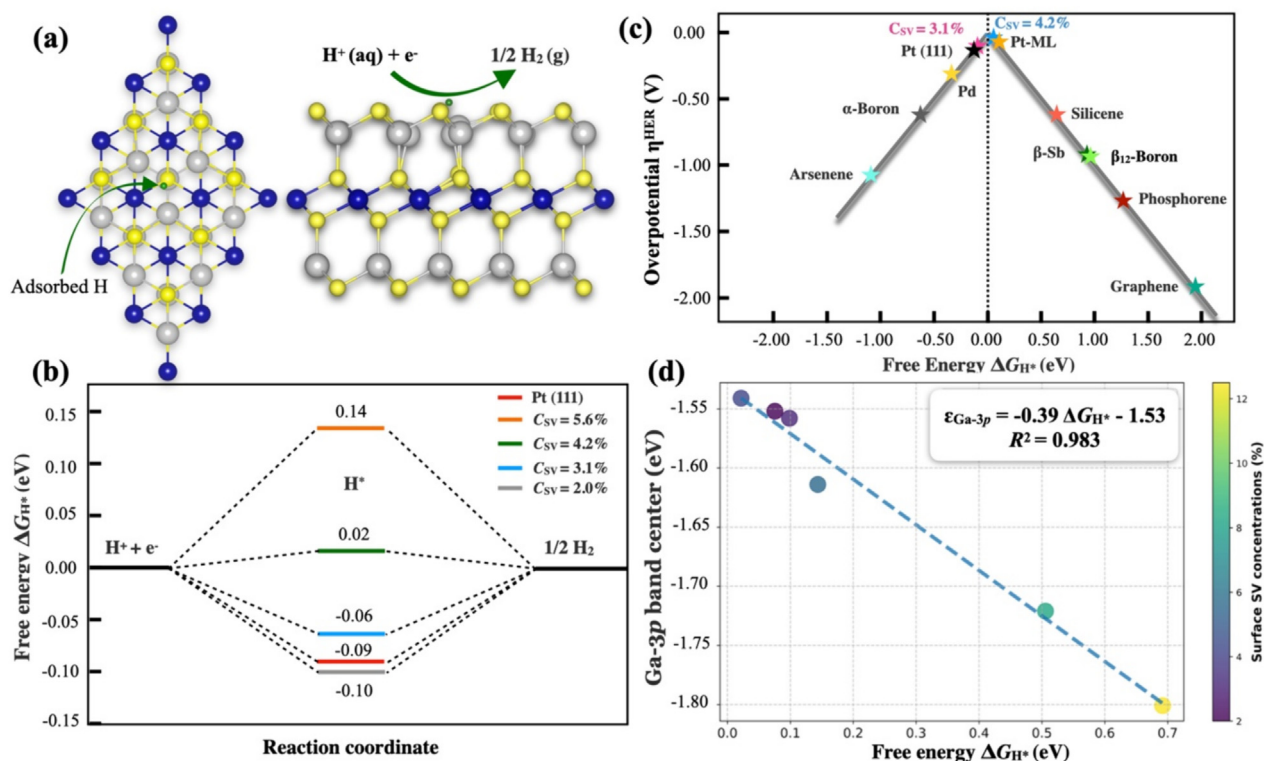


Fig. 2. (a) A top and side view of hydrogen atom adsorbed on the 3×3 supercell of defective 2D Ga₂CoS₄ substrate with a single sulfur vacancy. (b) Free energy diagram for the HER on 2D Ga₂CoS_{4-x} substrate at various C_{SV}. (c) HER activity volcano plot for 2D Ga₂CoS_{4-x} compared to other 2D materials previously reported [54–60], including platinum as a benchmark [7]. (d) Correlation between the Ga-3p band center ($\epsilon_{\text{Ga-3p}}$) and the Gibbs free energy change (ΔG_{H}^*) for hydrogen adsorption on 2D Ga₂CoS_{4-x} substrate with varying C_{SV}.

Table 1

Comparative data of adsorption energies (ΔE_{H}^*), the $\Delta \text{ZPE-TAS}$, Gibbs free energies (ΔG_{H}^*) for H-adsorption on 2D Ga₂CoS_{4-x} substrate with various sulfur vacancy ratio (C_{SV}), and formation energies (E_{form}) of the corresponding defective systems. $\Delta E_{\text{C}} = \Delta \text{ZPE-TAS}$ indicates the correction energy term.

Supercell	C _{SV} (%)	E _{form} (eV)	ΔE _H [*] (eV)	ΔE _C (eV)	ΔG _H [*] (eV)
2 × 2	12.5	6.343	0.461	0.23	0.691
2 × 3	8.3	6.299	0.265	0.24	0.505
3 × 3	5.6	6.172	-0.096	0.24	0.144
3 × 4	4.2	6.152	-0.218	0.24	0.022
4 × 4	3.1	6.129	-0.302	0.24	-0.062
5 × 5	2.0	6.090	-0.343	0.24	-0.103

As previously mentioned, 2D Ga₂CoS₄ demonstrates very high HER activity based on the calculations of the Gibbs free energy as detailed in Appendix S2 in SI, with a ΔG_H^{*} value of 2.45 eV. Our detailed investigation into the catalytic properties of 2D Ga₂CoS_{4-x} with varying concentrations of surface SVs shows a significant enhancement in HER activity, with ΔG_H^{*} values ranging from -0.103 to 0.691 eV as summarised in Table 1 (Fig. 2a and Fig. S4). Notably, for C_{SV} below 5.6%, the ΔG_H^{*} values are comparable to or even lower than that of widely used Pt-catalyst, suggesting near thermoneutral hydrogen adsorption (Fig. 2b). This implies that 2D Ga₂CoS_{4-x} with a low sulfur vacancy ratio can potentially outperform commercial Pt-based catalysts in HER activity. The optimal HER activity is observed at a C_{SV} of approximately 4.2%, where the overpotential is predicted to be around 0.022 eV, suggesting that by precisely controlling the surface sulfur vacancy ratio, 2D Ga₂CoS_{4-x} can be tuned to act as an efficient catalyst for hydrogen production. Furthermore, a volcano-curve assessment outlined in the SI (Appendix S3) and depicted in Fig. 2(c) indicates that 2D Ga₂CoS_{4-x} with C_{SV} of 4.2% and 3.1% demonstrates significantly higher HER activity compared to other 2D materials

and Pt-based catalysts. The underlying mechanisms behind the enhanced HER activity are attributed to the introduction of spin-polarized defect states near the Fermi level, which enhances hydrogen adsorption (Fig. S5). Additionally, our analysis based on the *p/d*-band center theory (Appendixes S4 and S8 in SI) reveals a direct correlation between the SV concentration-dependent HER activity and the energy shift of $\epsilon_{\text{Ga-3p}}$ values as illustrated in Fig. 2(d). These findings underscore the potential of 2D Ga₂CoS_{4-x} as an effective HER catalyst.

3.2. HER catalytic activity of transition metal atoms

In the next step, we employed a defective 2D Ga₂CoS_{4-x} surface to anchor 18 single TM atoms from the 3d, 4d, and 5d groups, creating a series of SACs labeled as TM@Ga₂CoS_{4-x}. The top and side views of fully optimized structures are illustrated in Fig. 3(a) and Figs. S6–S8, which were used to evaluate their long-term catalytic stability. Stability during reactions is a key factor for the durability of TM@Ga₂CoS_{4-x} catalysts, necessitating a strong binding of single metal atoms to the 2D Ga₂CoS_{4-x} surface to avoid aggregation. The binding energies (E_{b}) were calculated as detailed in the SI (Appendix S5). A negative E_{b} value is generally considered favorable as it indicates the formation of stable single-atom sites and prevents metal atom aggregation, as noted in previous studies. Table S1 shows that all the TM@Ga₂CoS_{4-x} systems investigated exhibited negative E_{b} values, indicating that the TM atoms are stably anchored to the 2D Ga₂CoS_{4-x} substrate. Furthermore, the difference between the binding energy and the cohesive energy (E_{c}) of TM atoms was computed to assess the metal-support interaction in TM@Ga₂CoS_{4-x}. Our results, presented in Fig. 3(b), demonstrate that for most TM@Ga₂CoS_{4-x} candidates, the ΔE_{b} values were negative, indicating a propensity for spontaneous dispersion rather

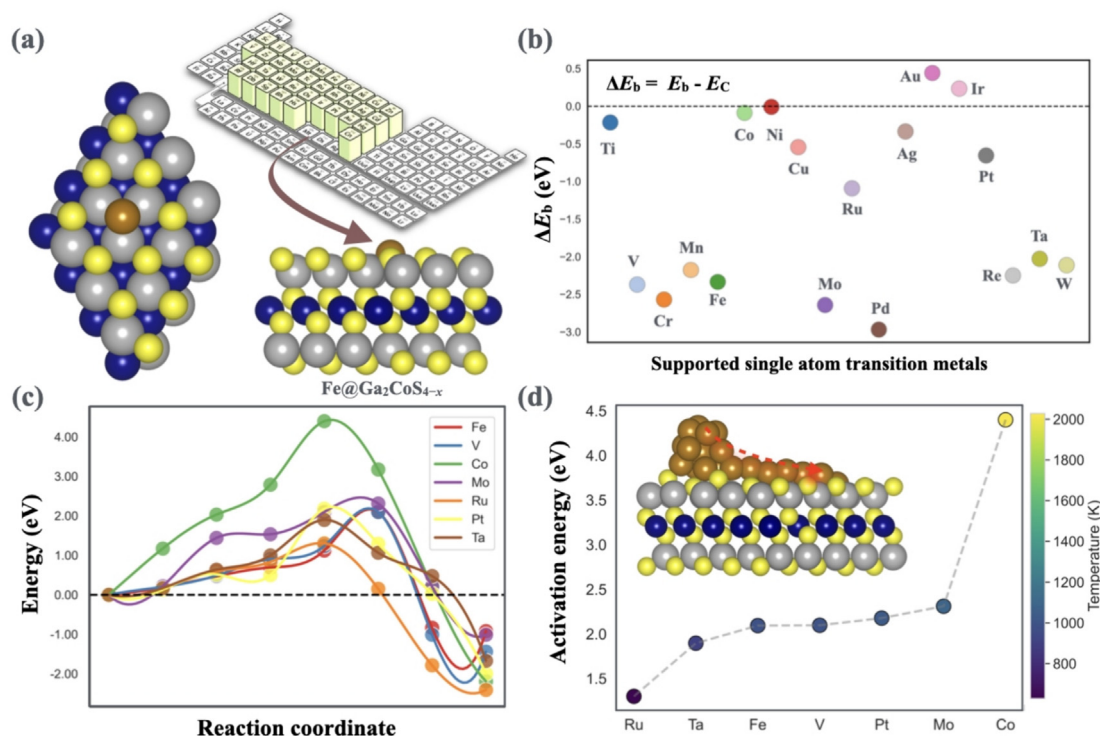


Fig. 3. (a) Top/side views of the 2D Fe@ Ga₂CoS_{4-x} substrate. (b) Computed adsorption energy change and the cohesive energies of TM atoms for TM@ Ga₂CoS_{4-x} substrates. (c) Minimum energy pathway illustrating the detachment process of a single TM atom from a host nanoparticle to binding on 2D Ga₂CoS_{4-x} substrate. (d) Activation energy and the corresponding temperature requirements for the detachment of a single TM-atom from the host nanoparticle, as predicted by transition state theory. The inset displays the fully relaxed pathway for the decomposition of a Fe-atom from a Fe-nanoparticle.

than clustering on the surface. However, for Au, Ir, and Ni, as shown in Fig. 3(b) and Table S1, the ΔE_b values were slightly positive, suggesting a potential for clustering on the 2D Ga₂CoS_{4-x} surface, since agglomeration is more likely when $\Delta E_b > 0$. Additionally, TM@Ga₂CoS_{4-x} stability in an electrochemical environment was assessed using the dissolution potential (U_{diss}), as detailed in Appendix S5 and Table S1. A positive U_{diss} against the standard hydrogen electrode indicates stability against TM-atom dissolution. Only three candidates show a negative value and may be unstable. Most TM@Ga₂CoS_{4-x} systems show effective anchoring by the 2D Ga₂CoS_{4-x} substrate. Favorable E_b and U_{diss} values suggest that 2D Ga₂CoS_{4-x} can prevent TM atom clustering or dissolution, offering a promising substrate for active sites.

Additionally, the specific electronic properties of TM atoms interacting with the substrate are critical in maintaining their dispersion and avoiding clustering on the substrate. In this context, Bader charges analysis and electronic DOS serve as effective methods for quantitatively assessing charge redistribution and Coulomb interactions. Charge transfer values from the TM atom to the 2D Ga₂CoS_{4-x} substrate, as shown in Table S1, range from 0.44e to 1.55e. This leads to all the metal atoms acquiring a positive charge. This indicates a significant electrostatic interaction between the 2D Ga₂CoS_{4-x} substrate and the TM atoms. Figs. S9–S11 illustrate that as the occupation of TM-*d* orbitals increases, there is a corresponding increase in charge transfer from left to right across each group, consistent with the electronegativity trends of the TMs (Table S1). For instance, Ta-atom shows the highest charge transfer of 1.55e with an electronegativity of 1.51, whereas Au-atom has a charge transfer of 0.44e with an electronegativity of 2.54. Positively charged TM atoms play a pivotal role in enhancing hydrogen adsorption, thus benefiting the catalytic process. Additionally, TM@Ga₂CoS_{4-x} exhibits excellent electrical conductivity due to the partially filled TM-*d* orbital states, which facilitates charge

transfer during the HER. This property, demonstrated in Figs. S9–S11, is essential for improving HER performance and lowering the overpotential.

Further investigation was conducted into the fundamental mechanisms governing the dispersion of TMs on a 2D Ga₂CoS_{4-x} surface. As illustrated in Fig. 3(c), the energy barrier of the system was examined corresponding to the scenario when a single TM detaches from a TM nanoparticle and subsequently attaches to an SV site on a 2D Ga₂CoS_{4-x} surface. As the detachment process initiates, the energy steadily rises until reaching a peak upon full detachment of the TM. This peak value signifies the decomposition energy barrier that must be surpassed for the TM to detach from the nanoparticle. A lower decomposition energy barrier indicates an easier detachment of the single TM atom from the metal nanoparticle. For practical applications, we estimated the requisite temperature and the activation energy, depicted in Fig. 3(d), for a single TM atom to leave the host nanoparticle using the transition state theory formalism outlined in the SI (Appendix S6). The analysis reveals that the required temperature for seven chosen single TM atoms to detach from the nanoparticle ranges between 730 and 1850 K. However, for Co, the required temperature exceeds 1900 K, rendering it impractical to synthesize Co single TM atoms on a substrate without deteriorating it.

We then evaluated the catalytic activities for the HER of eighteen transition metals adsorbed on the 2D Ga₂CoS_{4-x} substrate. The HER mechanism is typically represented by a three-state model [61]: (I) the initial state ($H^+ + e^-$), (II) the intermediate state (H^*), and (III) the final state (H_2). This model involves two main stages: (1) hydrogen adsorption ($H^+/e^- + * \rightarrow H^*$) and (2) hydrogen release ($H^+/e^- + H^* \rightarrow H_2 + *$). For each TM@Ga₂CoS_{4-x} system, we placed a hydrogen atom atop each TM atom and performed full optimizations to calculate the Gibbs free energy change for hydrogen adsorption (ΔG_H^*). The results are shown in Fig. 4(a and b) and

summarized in Table S2. The primary indicator of catalytic HER activity is ΔG_{H} . Optimal HER activity corresponds to $|\Delta G_{\text{H}}|$ values that are close to zero, as values that are too high or too low are less favorable for HER. Ideally, ΔG_{H} should be near zero, meaning that the Gibbs free energy of H-adsorption should approximate that of the product or reactant. The optimal catalytic activity is typically achieved if ΔG_{H} is in the range between -0.2 and 0.2 eV or less than -0.09 eV (Pt). In Fig. 4(b), the calculated ΔG_{H} values for the optimal TM@Ga₂CoS_{4-x} systems are shown, identifying six systems (with TM=Au, Pt, Ag, Cu, Pd, and Ni) that have moderate H binding strengths and overpotentials (η^{HER}) close to that of Pt (111) with an overpotential of 0.09 V [61]. To provide a comprehensive comparison, we evaluated the HER catalytic performance of Ag@Ga₂CoS₄ and Pt@Ga₂CoS₄ under conditions where no surface sulfur vacancies were present. Pt and Ag were adsorbed onto a pristine $3 \times 3 \times 1$ supercell of Ga₂CoS₄, as shown in Fig. S12. Our findings indicate ΔG_{H} of approximately 0.38 eV for Ag@Ga₂CoS₄ and 0.24 eV for Pt@Ga₂CoS₄, which are higher than those for systems with surface defects (Fig. S12). The *d*-band center theory, as elaborated in Appendix S4, suggests that the HER activity of TMs is associated with the position of their *d*-band centers. We computed the *d*-band center (ε_d) for each TM atom, illustrated in Fig. 4(c), and established a correlation with the HER activities of TMs@Ga₂CoS_{4-x}. As shown in Fig. 4(d), there is a clear linear relationship between ε_d and η^{HER} for TMs@Ga₂CoS_{4-x}. Specifically, as the *d*-band center becomes more negative, the overpotential η^{HER} decreases, indicating an enhancement in HER activity. This trend suggests that the *d*-band center position is also a critical factor in determining the catalytic efficiency of TMs for the HER.

3.3. Few-shot ML for HER catalytic activity prediction

Finally, we have introduced a ML algorithm aimed at predicting the HER catalytic activity to address the difficulties of efficiently evaluating the HER catalytic properties of various SACs. The

proposed workflow, depicted in Fig. 5, integrates DFT calculations with ML methodologies. This integration allows us to uncover descriptors and rules with high predictive power for the development of new catalytic materials. Importantly, the ML protocol does not require detailed knowledge of specific reaction steps; instead, it relies on general data about the transition metals. This feature makes it particularly useful for exploring new catalyst systems with unestablished reaction mechanisms, facilitating the data-driven identification of key features that provide rapid insights into potential mechanisms. The primary focus of this study is the HER overpotential. The workflow is structured into three different steps, where we build upon these steps by employing a state-of-the-art few-shot learning approach, which has been proven to be highly effective in domains with limited data availability. Few-shot learning is especially valuable in catalysis, where data acquisition can be both costly and time-consuming. Notably, few-shot learning models have been successfully applied in other fields, such as drug discovery, to predict compound activities with high precision despite limited experimental data [50,62,63]. Inspired by these applications, we have tailored our few-shot learning model to the specific challenges of catalytic research, particularly in predicting the HER catalytic activity of SACs on a Ga₂CoS_{4-x} substrate.

Our few-shot learning model is designed to address the inherent complexities associated with SAC systems, such as surface reconstruction, leaching, and defects. By integrating this approach with a novel intrinsic descriptor that links SAC atomic properties to HER overpotential, we have significantly enhanced the ability of the model to predict HER overpotential with high accuracy. The model architecture, which combines a feature extraction module with a fully connected neural network (FCNN), is optimized for both prediction accuracy and computational efficiency (Appendix S9). This ensures that our model can effectively process and analyze complex datasets, leading to more reliable predictions. Additionally, the incorporation of DFT-derived features into our

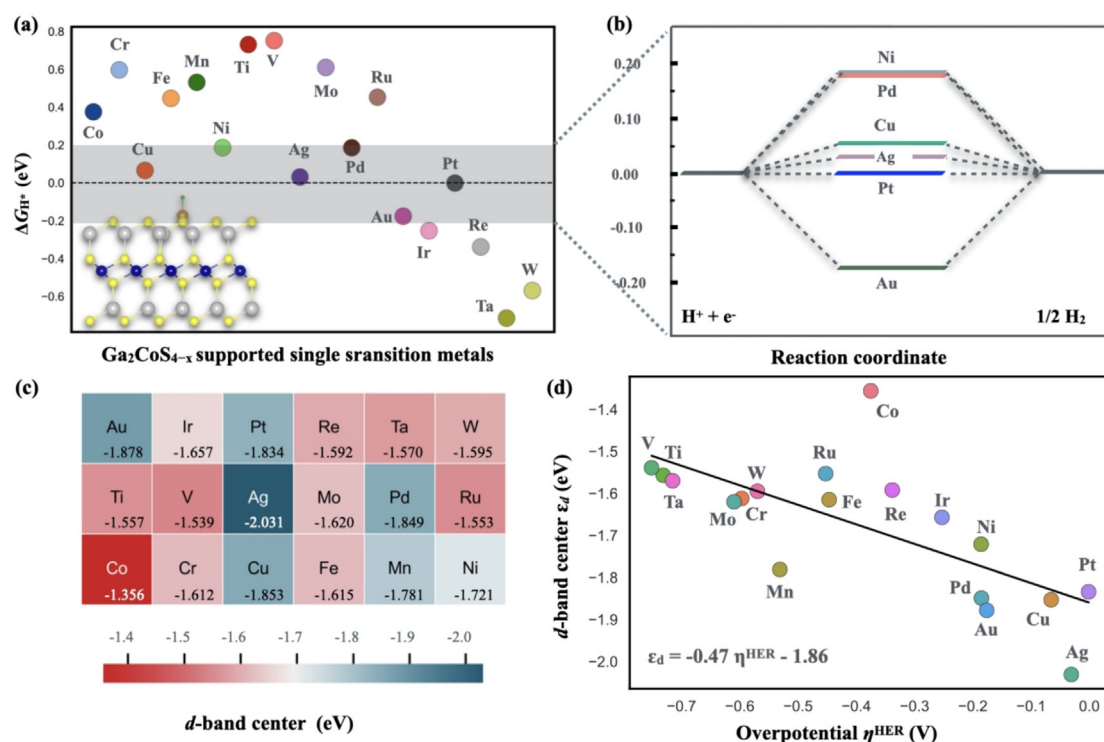


Fig. 4. (a, b) The computed Gibbs free energy change for hydrogen adsorption ΔG_{H} on 2D TMs@Ga₂CoS_{4-x} substrate and the optimal TM@Ga₂CoS_{4-x} catalysts. (c) The *d*-band center positions of TM-atoms on a 2D Ga₂CoS_{4-x} substrate. (d) The linear regression between the *d*-band center position and the HER overpotential η^{HER} .

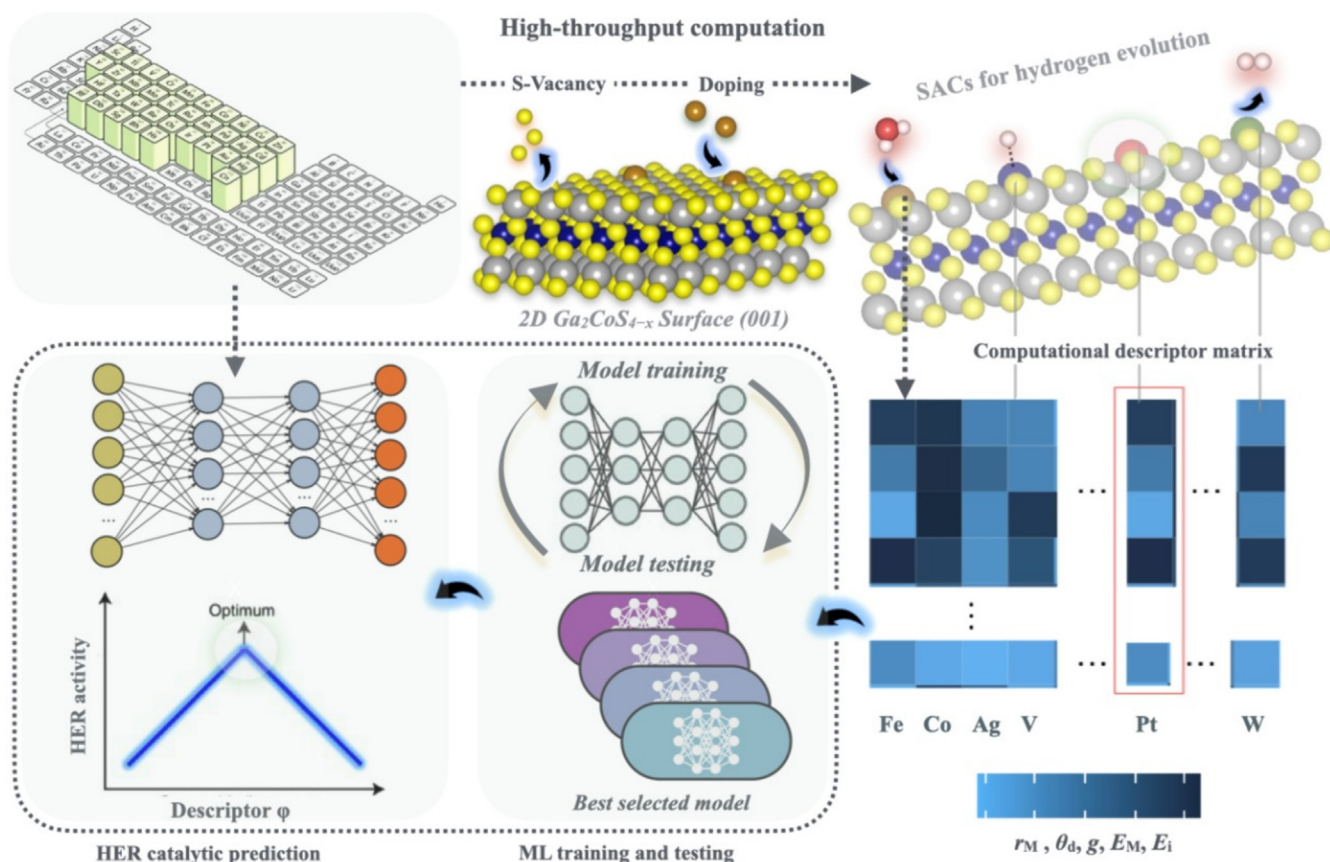


Fig. 5. The comprehensive workflow illustrating the integration of TMs@ $\text{Ga}_2\text{CoS}_{4-x}$ selection, DFT calculations, performance correlation, and the precise prediction of catalytic properties through DFT and machine learning techniques. This approach leverages high-throughput computational methods to identify key descriptors and rules, enhancing the predictive accuracy for developing new and promising HER catalysts.

model enhances its robustness by capturing the subtle nuances of catalytic activity. The workflow is structured into three different steps, as follows.

- **Data compilation:** In the first stage, we collect HER overpotential η^{HER} data from DFT calculations for a group of eighteen TMs on a 2D $\text{Ga}_2\text{CoS}_{4-x}$ substrate. This data will serve as the foundation for our ML model. To thoroughly capture the electronic properties affecting electron transfer and reaction energy during HER, we identify five essential parameters associated with the TMs. These parameters include atomic characteristics (metal atomic radius r_M and d -electron count θ_d), periodic classification (group number g), and reactivity indicators (electronegativity E_M and the first ionization energy E_i). The input data ($x_i = [r_M, \theta_d, g, E_M, E_i]$) are combined with the target data (η^{HER}) for the eighteen TMs, forming a comprehensive dataset for the ML model training.
- **Machine learning-based training and testing:** In this stage, we construct a multi-layered ML model to derive features from input data at various abstraction levels. The previously gathered dataset is randomly split into two groups: a testing subset containing two input-target pairs for Pd and Pt, and a training subset consisting of the remaining sixteen TMs. To address the limited data, we employ a few-shot learning technique, which is suitable for scenarios with sparse data [63–66]. For the case of a single transition metal atom supported on a 2D $\text{Ga}_2\text{CoS}_{4-x}$ substrate, each with distinct coordination environments, few-shot learning is used to capture the geometric spatial relationships between the transition metal atom and Ga atoms. The atomic parameters identified earlier serve as node features,

with connections formed based on bonding states. This topological information is fed into the ML model, which consists of five convolutional layers and two fully connected layers. The convolutional layers extract multi-level features that describe the interactions between the transition metal atom and the Ga atoms. The final outputs of the convolutional layer are then processed by the fully connected layers to predict the overpotential of transition metal atom through regression.

- Using PyTorch [67], the ML model was developed and trained with two separate data subsets. The Adam optimizer [68] was employed during training, maintaining a fixed learning rate of 0.001 across 125 epochs. The mean squared error served as the loss function, and the model performance was measured using relative deviation. After 60 training iterations, the model with the lowest relative deviation on the testing subset was selected as the optimal one. This training and evaluation processes were specifically targeted at assessing the HER catalytic activity of single transition metal atoms on the 2D $\text{Ga}_2\text{CoS}_{4-x}$ substrate. The predictions of HER activity by the optimal ML model showed a maximum deviation of 7.63% compared to those obtained via DFT calculations.
- **Model prediction:** In the final stage, we leverage the most effective ML models derived in the preceding stage to forecast the HER catalytic activity, particularly the η^{HER} of transition metal SACs on the 2D $\text{Ga}_2\text{CoS}_{4-x}$ substrate with the single transition metal atoms being the remaining eleven transition metal species. The comprehensive dataset encompassing input parameters for all the transition metal species is meticulously documented in Table S3. To accelerate the process of finding the best catalyst candidates, we define a novel intrinsic catalytic

HER-activity descriptor. This enables the establishment of a “volcano-curve” relationship between η^{HER} and the descriptor [69], shown in Fig. 6(a).

Based on the Pearson correlation coefficient (PCC) of different features (as delineated in Fig. 6b) and the reaction energy in HER, we discern five parameters, where E_i , g , θ_d , and E_M exhibit positive correlations with the HER catalytic activity, while r_M demonstrates an inverse relationship. Consequently, we derive an initial expression for the intrinsic catalytic activity descriptor (φ) as $\theta_d \times g \times E_i \times (E_M / r_M)$. Considering the crucial role of metal binding on the 2D $\text{Ga}_2\text{CoS}_{4-x}$ substrate, we integrate the electronegativity (E_j) and atomic radius (r_j) of the 2D $\text{Ga}_2\text{CoS}_{4-x}$ substrate into the φ , culminating in the expression.

$$\varphi = \theta_d \times g \times E_i \times (E_M / r_M + E_j / r_j) \quad (1)$$

Fig. 6(a) illustrates the “volcano-curve” relationship, depicting the dependency of η^{HER} on the intrinsic descriptor φ for SACs composed of various transition metals on the 2D $\text{Ga}_2\text{CoS}_{4-x}$ substrate, based on both DFT calculations and ML predictions. Our result indicates that Pt, Ag, and Cu occupy the peak of the volcano, representing the optimal HER catalytic activity. This highlights that Pt and Ag, among non-radioactive transition metals, exhibit superior HER catalytic activity on a $\text{Ga}_2\text{CoS}_{4-x}$ substrate. The volcano relationship between η^{HER} and φ in Fig. 6(a) is crucial for identifying SACs with favorable HER catalytic properties. Additionally, the volcano relationship calculated from the logarithm of exchange current density $\log(i_0)$ versus ΔG_{H}^* for various catalysts further reflects the HER activities of these SACs. The volcano diagram in Fig. 6(c) serves as a straightforward method to visualize, compare, and enhance the activity, thereby aiding in the design of different metal catalysts.

The ML model notably enhances the speed and precision in identifying volcano-curve relationships by efficiently predicting HER activity, as shown in Fig. 6(d). To evaluate computational efficiency, we compared the central processing unit hours (CPU-h) necessary for DFT calculations and ML predictions of HER activity. Utilizing the VASP package on a 128-core supercomputer, DFT calculations require approximately 20.0 CPU-h per ΔG_{H}^* and η^{HER} value, which amounts to 1000 CPU-h on a single-core CPU. In comparison, ML predictions, including model training, testing, and prediction, are accomplished in less than 1 min on a single-core CPU. This demonstrates that ML predictions for the catalytic activity of SACs on 2D $\text{Ga}_2\text{CoS}_{4-x}$ substrate are vastly more time efficient but as accurate as the DFT calculations. Consequently, the significant reduction in computational cost achieved through the ML-driven discovery process effectively mitigates the limitations of DFT calculations in identifying high-performance SACs on 2D $\text{Ga}_2\text{CoS}_{4-x}$ substrate. Our work demonstrates an ML-accelerated approach for the prediction of the overpotential of HER and shows the HER overpotential of SACs on the 2D septuple-atomic-layer $\text{Ga}_2\text{CoS}_{4-x}$. However, the influence of the substrate on the HER activity needs to be further investigated.

4. Conclusions

In conclusion, we have demonstrated a rational design methodology for 2D septuple-atomic-layer $\text{Ga}_2\text{CoS}_{4-x}$ supported SACs that show enhanced catalytic activity for the HER, using a combination of DFT and ML techniques. Our research highlights that the incorporation of transition metal nanoparticles into a stable SAC configuration on a 2D $\text{Ga}_2\text{CoS}_{4-x}$ substrate is controlled by the energy barrier of single metal atom detachment from the nanoparticle. An intrinsic descriptor has been identified, showing a strong

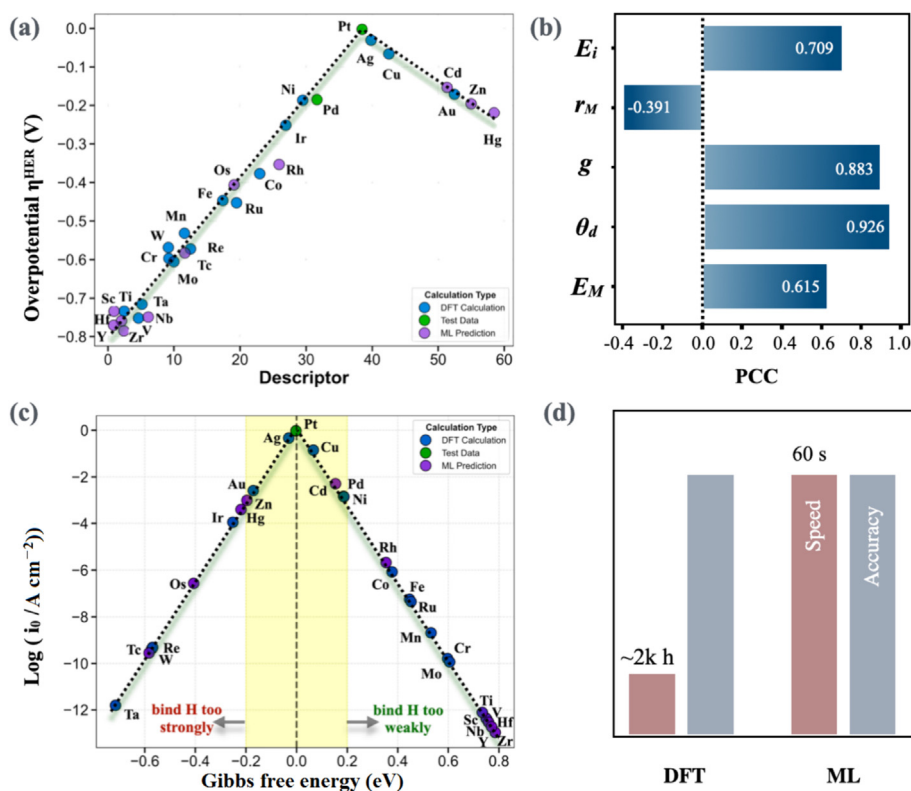


Fig. 6. (a) Catalytic activities for the HER as a function of the intrinsic descriptor φ for TMs on a 2D $\text{Ga}_2\text{CoS}_{4-x}$ substrate. (b) PCC for the five atomic properties of SACs on a 2D $\text{Ga}_2\text{CoS}_{4-x}$ substrate. (c) Exchange current density for hydrogen adsorption on SACs on a 2D $\text{Ga}_2\text{CoS}_{4-x}$ substrate. (d) Comparison of computational costs involved in predicting HER catalytic activity of TMs@ $\text{Ga}_2\text{CoS}_{4-x}$ systems using DFT compared to ML techniques. cm^{-2} .

correlation with the HER catalytic activity of SACs on 2D Ga₂CoS_{4-x} substrate, following a volcano-curve plot relationship, which enables the efficient selection of optimal SACs. The ML model, featuring a few-shot learning algorithm, significantly reduces the computational time required to determine the HER catalytic activity of SACs on 2D Ga₂CoS_{4-x} substrate. This ML-accelerated, simulation-based design approach holds significant promise for expediting the discovery of effective SACs for a wide range of catalytic applications.

CRediT authorship contribution statement

Nabil Khossossi: Writing – original draft, Visualization, Validation, Resources, Project administration, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Poulumi Dey:** Writing – review & editing, Supervision, Software, Project administration, Funding acquisition, Conceptualization.

Data availability

All data supporting the conclusions of this research are provided in the main text and [Supplementary Information](#). Additional datasets are available from the corresponding authors upon reasonable request. Fully optimized structures, input files, and Python scripts used in the DFT calculations are stored at https://github.com/NabKh/SACs_HER_2024.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jechem.2024.09.009>.

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