

# High-Throughput Computational Discovery and Intelligent Design of Two-Dimensional Functional Materials for Various Applications

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**CONSPECTUS:** Novel technologies and new materials are in high demand for future various applications to overcome the fundamental limitations of current techniques. For example, the nanomaterials are in high demand for the miniaturization of electric devices. Spintronics is one of the most viable solutions for green electric devices. The single-atom catalysts can provide a 100% utilization rate. Heterojunctions hold promise for applications in energy conversion and storage. Two-dimensional (2D) materials show promising applications in various applications because they can be tailored to the specific property on which a technology is based and may be compatible with other technologies. Although the number of experimentally discovered 2D materials is growing, the speed is very slow and only a few dozen 2D materials have been synthesized or exfoliated since the discovery of graphene. Recently, a novel computational technique, dubbed high-throughput computational materials design, has become a burgeoning area of materials science, which is a combination of quantum mechanical theory, materials information, and database construction with intelligent data mining. This new and powerful tool can greatly accelerate the discovery, design, and application of 2D materials by creating a database containing a large number of 2D materials with calculated fundamental properties and then intelligently mining (via high-throughput automation or machine learning) the database in the search for 2D materials with the desired properties for particular applications, such as energy conversion/storage, catalysis, water purification, electronics, and optoelectronics.

In this Account, we summarize our recent progress in the emerging area of 2D materials discovery, database construction, 2D functional materials design, and device development by quantum-mechanical modeling, high-throughput calculations, and machine learning inspired by the materials genome concept. We developed an open 2D materials database—2D materials encyclopedia (2DMatPedia; <http://www.2dmatpedia.org/>), which includes a variety of structural, thermodynamic, mechanical, electronic, and magnetic properties of more than 6000 two-dimensional materials. Using high-throughput computational screening and machine-learning techniques, we identified exotic 2D materials and heterojunctions with desired properties for several applications, such as the electrocatalysis of hydrogen and nitrogen evolution reactions, photocatalysis for water splitting, high Curie temperature ferromagnetic materials, ferromagnetic(ferroelectric) tunnel junctions, piezo(ferro)electricity, and excitonic solar cells. Our open 2D materials database with high-throughput calculations and proper advanced models will greatly reduce the experimental effort in trial and error, narrow the scope for both experimental and theoretical explorations, and thus boost the fast and sustainable development in the area of 2D materials. Despite the significant progress and successful deployment of materials informatics, data-driven materials discovery, high-throughput calculations, and machine learning as a major game change in the area of 2D materials science and technology, future challenges remain in several aspects, which are summarized in the Outlook.

## 1. INTRODUCTION

Two-dimensional (2D) materials and van der Waals (vdW) heterostructures,<sup>1</sup> a new degree of freedom in the materials space, have attracted tremendous interest in the applications of (opto)electronics,<sup>2,3</sup> spintronics,<sup>4</sup> valleytronics,<sup>5</sup> twistrionics,<sup>6</sup> and slidetronics,<sup>7</sup> such as (photo)electrocatalysis,<sup>8,9</sup> ferro-(piezo)electricity,<sup>10,11</sup> spin(orbit) tunnel junctions,<sup>12,13</sup> and flexible solar cells.<sup>14,15</sup> This extremely fast development of 2D materials and vdW heterostructures is prompted by their various and tunable properties, such as recently reported unconven-

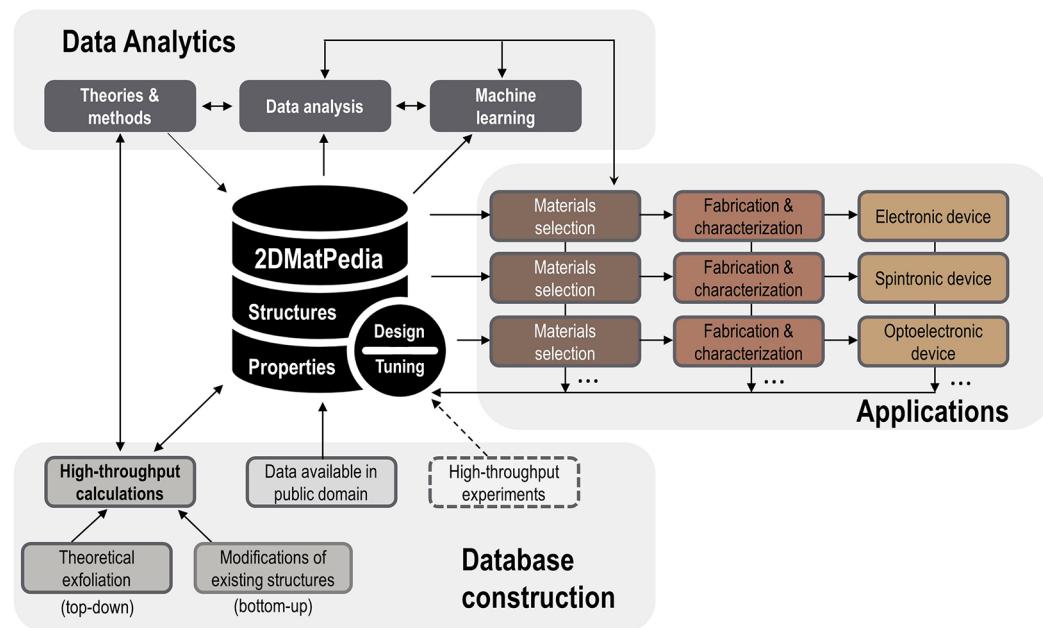
tional superconductivity in magic-angle stacked bilayer graphene and spin(valley) polarization in rhombohedral trilayer

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**Scheme 1.** Infrastructure of 2D Materials Database Construction, Data Analytics, and Technological Applications

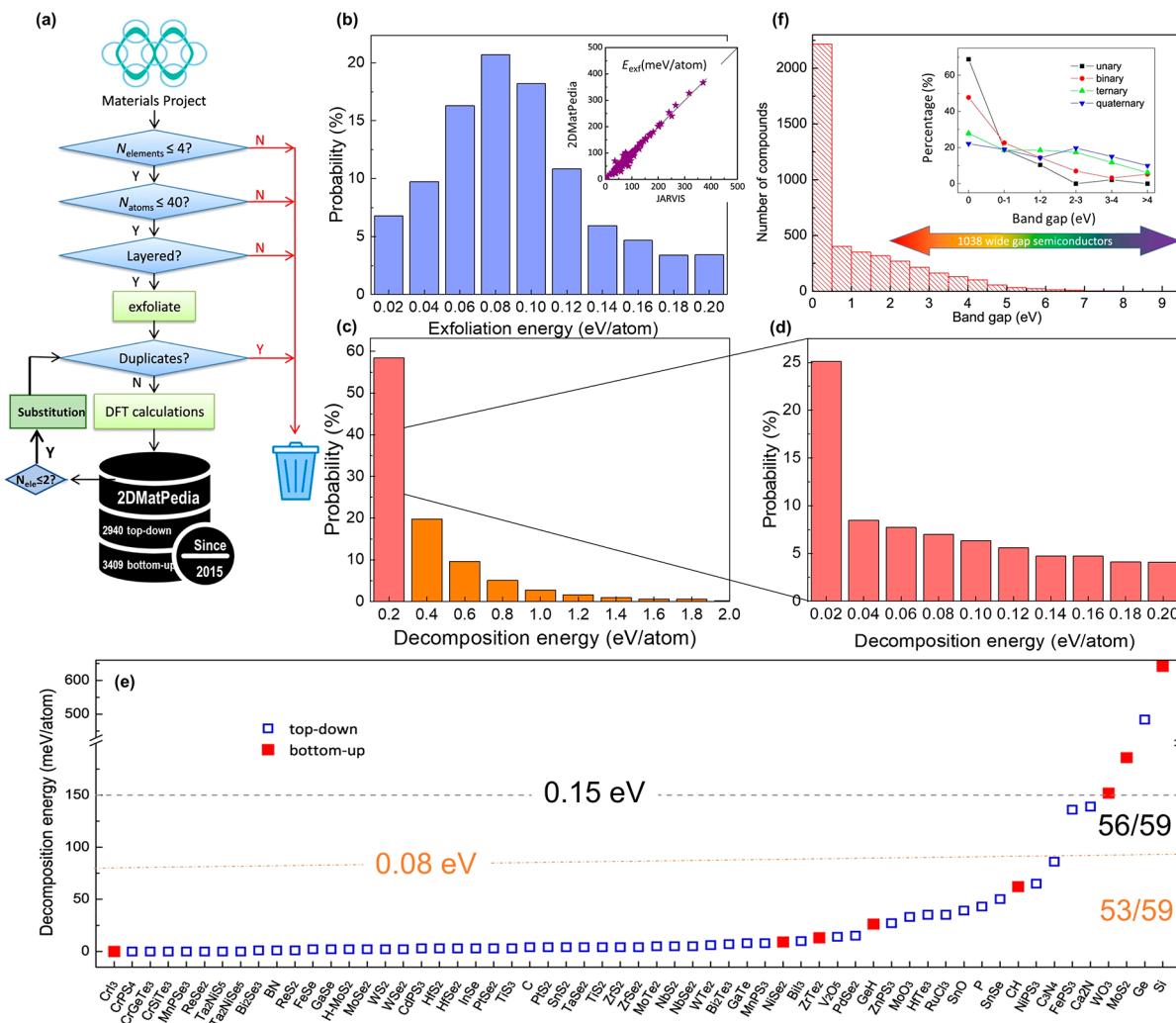
graphene<sup>14</sup> and 17 years since the birth of graphene, respectively.<sup>6,16</sup> The other driving force in the rapid development in the field of 2D materials is the discovery of a wealth of new 2D materials by high-throughput experiments<sup>17</sup> and high-throughput calculations.<sup>18–20</sup> In particular, the latter fuels the 2D materials pool by several orders of magnitude through two channels. The first and most natural approach is called top-down, which is inspired by the experimental exfoliation of graphene from layered graphite. Using high-throughput screening on 5619 experimentally known layered 3D compounds, Mounet et al. identified 1036 candidates that might be easily exfoliated to be monolayer 2D materials.<sup>19</sup> They further developed a free and open 2D materials database named Materials Cloud to store such a large portfolio of 2D materials and the vibrational, electronic, magnetic and topological properties of 258 compounds.<sup>19</sup> Using the same top-down approach and assisted by high-throughput calculations, Choudhary et al., Gjerding et al., and Zhou et al. also constructed open 2D materials databases JARVIS,<sup>21</sup> Computational 2D Materials Database (C2DB),<sup>20</sup> and 2DMatPedia,<sup>18</sup> which include 812, 1500, and 2940 exfoliable 2D materials, respectively. Besides the physical top-down approach, we also propose a chemical bottom-up design that doubles the number of 2D materials to ~6000 (**Scheme 1** and details in **Section 2**).

The combination of the exponential increase in computing power with matured, cost-effective, and empirical parameter-free density functional theory (DFT) codes promotes high-throughput DFT calculations, which have shown success as a powerful tool in the area of new materials discovery, such as the high-throughput-enabled development of the Materials Project Database<sup>22</sup> and the Magnetic Topological Materials Database.<sup>23</sup> This calculation-first approach is especially effective in searching the complex heterogeneous catalysts. It can efficiently guide experimentalists in finding high-performance catalysts without having to synthesize them first. For instance, Zhong et al. identified high-performance Cu–Al-based CO<sub>2</sub> electrocatalysts from 244 different copper-containing intermetallics in the Materials Project Database using high-throughput DFT calculations, which was verified subsequently by experiment.<sup>24</sup>

Hannagan et al. reported the high-throughput-calculation-led experimental discovery of a single-atom-alloy catalyst for propane dehydrogenation.<sup>25</sup> It is worth noting that the high-throughput calculations in the field of materials science are defined as “an automatic and overwhelming flow from ideas to results”, enabling scientists not only to discover thousands of new materials but also to calculate a variety of their properties as part of a single study. It can thus generate a huge number of materials-related data sets which fuel interest in the application of machine learning (ML) to further accelerate the discovery and design of new materials.

With the rapid development of high-throughput computational techniques in materials science, our ability to generate materials’ “big data” has greatly surpassed our capability to analyze it with conventional approaches, underscoring the emergence of machine learning. ML and its deep-learning (DL) subset, encompassing statistical algorithms and modeling tools used for finding patterns in high-dimensional data, have been successfully employed in the materials science discipline in the past decade.<sup>26</sup> For instance, ML and DL models have shown their power in predicting the crystal stability,<sup>27</sup> nanoparticle synthesis,<sup>28</sup> diffraction pattern recognition,<sup>29</sup> battery technologies,<sup>30</sup> and design of molecules, lead-free perovskites, and heterogeneous catalysts.<sup>26,31,32</sup> The key role of ML in materials science is to extract useful information or patterns in the processing–structure–properties–performance relationships (PSPP), which are difficult to establish on the basis of the conventional laws of physics and chemistry. Once the materials PSPP relationship is established by ML techniques, one can design new materials with targeted performance using the inverse design (PPSP).

In our group, we leverage on our core expertise in computational 2D materials as well as high-throughput screening and machine-learning techniques to develop capabilities in 2D materials informatics and a 2D materials platform, including (i) database construction, (ii) data analytics, and (iii) applications (**Scheme 1**). This platform offers a unique, easy-to-implement, and powerful solution for discovering new 2D



**Figure 1.** (a) Workflow of both the top-down and bottom-up design of 2D materials. (b) Statistical analysis of the exfoliation energy of 2D materials in 2DMatPedia. The inset shows a comparison of the exfoliation energy with that in JARVIS. (c and d) Statistical analysis of the decomposition energy of all materials in 2DMatPedia. (e) Calculated decomposition energy for 59 known experimentally grown 2D materials. (f) Statistical analysis of the band gaps for the relatively stable 2D materials in 2DMatPedia. The inset represents the band gap distribution of different compounds. Reproduced with permission from ref 18. Copyright 2019 Springer Nature.

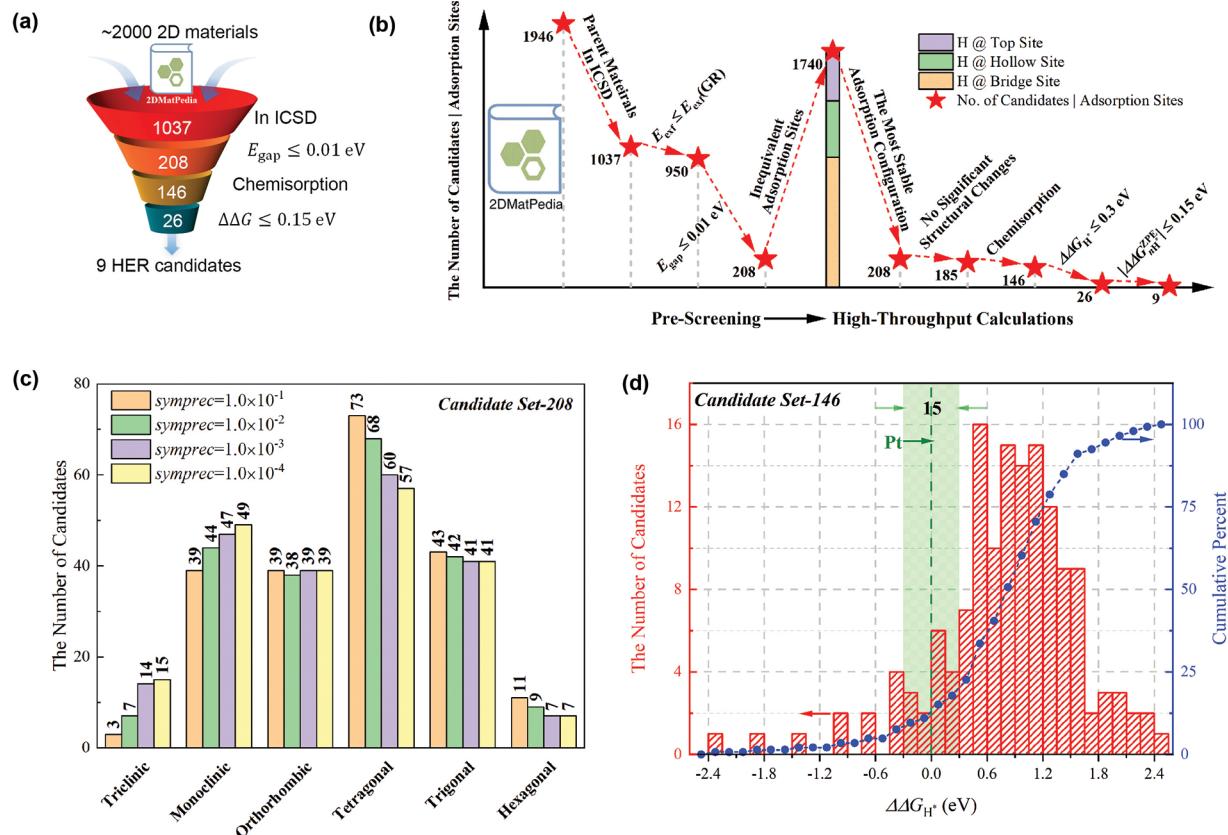
materials, designing the desired functionality, and exploring their technological applications.

## 2. DATABASE CONSTRUCTION

Using the high-throughput screening of a bulk materials database, we have identified a large data set of 2D materials (2DMatPedia).<sup>18</sup> There are a total of 6351 materials in the current database, whereas 2940 were obtained by exfoliating existing layered materials (top-down approach), 3409 were obtained by the chemical substitution of 2D materials (bottom-up approach), and 2 were obtained from the literature via neither a top-down or bottom-up approach. The bottom-up approach starts from the 35 unary and 755 binary compounds obtained from the top-down approach. Only the same-column elements are used for substitution. The elements from H (atomic number 1) to Bi (83) in the periodic table are considered, excluding the lanthanoid series (58–71). The reason we chose only the unary and binary 2D materials for substitution is that the number of compounds exponentially increases with the number of elements by element substitution. For example, on the basis of the existing ternary 2D compounds

(around 3000), more than 180 000 new ternary compounds can be generated. Clearly, this is extremely difficult for DFT-based high-throughput calculations. Instead, we are now applying machine-learning techniques to prescreen a large number of materials and select the predicted stable compounds for further high-throughput calculations. This work is ongoing with respect to expanding 2DMatPedia. The workflow is summarized in Figure 1a. We started from >80 000 inorganic compounds in the Materials Project Database (v2018).<sup>22</sup> In the initial stage, we focused on compounds with simple and layered structures. We then performed high-throughput DFT calculations to calculate the exfoliation energy, optimize these 2D structures, and calculate their properties, which were stored in the 2DMatPedia Database. The unary and binary 2D materials obtained from the top-down approach were then used as initial structures for bottom-up elemental substitution. Structure matching was applied to ensure unique structures for further high-throughput DFT calculations.

In the top-down process, we screened all bulk materials from the database of the Materials Project (v2018) for layered structures with a topology-based algorithm and theoretically

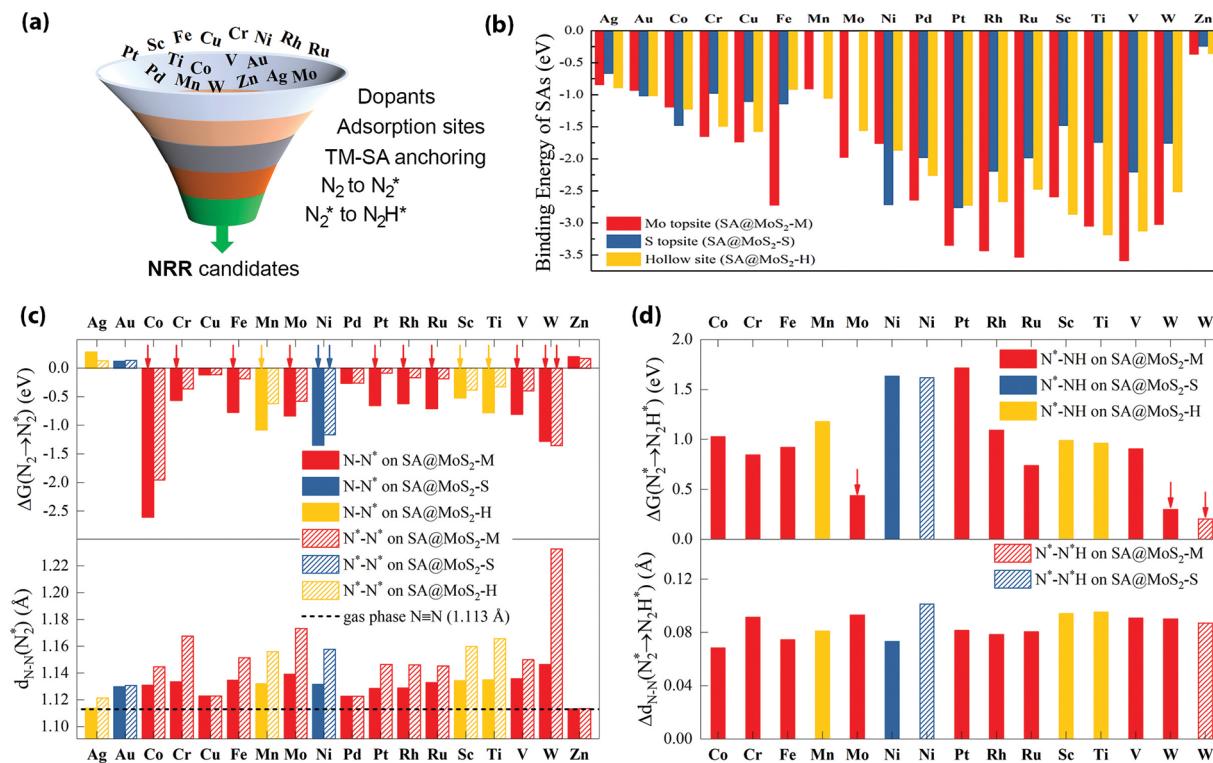


**Figure 2.** (a) Schematic of screening high-performance 2D HER electrocatalysts from the 2DMatPedia database. (b) Detailed criteria of prescreening and high-throughput screening. The total number of considered top-down 2D materials from 2DMatPedia is 1946. Each candidate set is labeled as Candidate Set- $X$ , where  $X$  is the set size. The stacked column shows the statistics on the inequivalent adsorption sites in Candidate Set-208. (c) Statistical results of Candidate Set-208 for a series of symmetry precisions. (d) Distribution of the hydrogen adsorption Gibbs free energy ( $\Delta\Delta G_H^*$ ) with respect to Pt on the catalyst candidates in Candidate Set-146 in the most stable adsorption configuration. The area shaded in light green shows the screened good candidates. Reproduced from ref 8. Copyright 2020 American Chemical Society.

exfoliated them into monolayers. The exfoliation energy, which is the energy required to isolate a layer from its layered parent, of all layered compounds was calculated and is in good agreement with the results in JARVIS<sup>21</sup> (Figure 1b and inset). Our statistical analysis shows that the number of compounds with exfoliation energies in the range of 0.06–0.10 eV/atom is the largest. Interestingly, this is also the range where graphene is located (0.067 eV/atom). Taking the cutoff of the exfoliation energy as 0.08 eV/atom, 54% of the 2D compounds can be easily exfoliated from their layered bulk parent materials. Besides the exfoliation energy, the decomposition energy, which is the energy needed to decompose a given material into a set of most-stable materials (energy above the hull), was calculated to investigate the stability of monolayer 2D materials. Figure 1c shows that the number of compounds decreases dramatically with the decomposition energy, and most of the 2D materials (>50%) have a decomposition energy of less than or equal to 0.2 eV/atom. Taking a close look at Figure 1d, we note that around one-fourth of the compounds have a decomposition energy of less than or equal to 0.02 eV/atom. By checking the decomposition energy of 59 known experimentally synthesized monolayer materials, we find that most of them (53/59) have a low decomposition energy (<0.08 eV) (Figure 1e). Thus, a decomposition energy threshold (or energy above the hull for bulk materials) of 0.08 eV/atom has been used to identify “stable” 2D materials, which is also adopted in other computational screening work.<sup>33</sup> On the basis of this criterion,

we find that 48% of the 2D compounds in 2DMatPedia might be stable. For the screened, stable 2D compounds (2231), our statistical analysis of the space group and elements shows that among them the largest portion of compounds is oxides (around one-fourth), followed by chloride and bromides. The anions are loosely distributed with around 150 compounds containing H as the largest portion, followed by V and Cu. The top seven most popular space groups are  $P\bar{3}m1$ ,  $P\bar{1}$ ,  $P2_1/c$ ,  $C2/m$ ,  $P2_1/m$ ,  $P4/nmm$ , and  $Pmmn$ . Materials in these 7 groups (out of a total of 60 2D space groups) compose 58% of the whole database.

In the bottom-up design process, we generated new 2D materials by the chemical substitution of elements in known 2D materials with others from the same group in the periodic table (from H to Bi, excluding radioactive, lanthanide, and actinide elements). We performed high-throughput calculations to obtain the band structures of all 6000 compounds as shown in Figure 1f and the inset. Furthermore, the structural, electronic, magnetic, piezoelectric, electrocatalytic, and energy properties of these 2D materials were consistently calculated by high-throughput DFT calculations. The details of computational methodology, recorded data, and technical validation can be found in ref 18. The whole database is publicly available at <http://www.2dmatpedia.org/>. This 2D materials database platform provides a starting point for further material screening, data mining, data analysis, and artificial intelligence applications. Some properties and applications of 2D materials discovered on



**Figure 3.** (a) Schematic of screening steps of 18 TM-SAs@MoS<sub>2</sub> for NRR. (b) Calculated binding energy of 18 TMs on monolayer MoS<sub>2</sub> at 3 different adsorption sites. (c) Gibbs free energy change of N<sub>2</sub> adsorption and the N–N bond length of the adsorbed N<sub>2</sub> on various TM-SAs@MoS<sub>2</sub>. The configurations labeled by arrows are selected to proceed to the next NRR step, consisting of the hydrogenation of adsorbed N<sub>2</sub><sup>\*</sup>. (d) Changes in the Gibbs free energy and the N–N bond length upon the hydrogenation of adsorbed N<sub>2</sub><sup>\*</sup>. Mo and W SAs (red arrows) are selected for the further evaluation of their NRR activities. Reproduced with permission from ref 35. Copyright 2020 Elsevier.

the basis of 2DMatPedia will be discussed in the following sections.

### 3. INTELLIGENT DATA MINING OF TWO-DIMENSIONAL FUNCTIONAL MATERIALS

Leveraging the 2DMatPedia database, one can perform intelligent data mining and high-throughput screening to discover functional materials with desired properties for particular applications. To demonstrate this, in this section we first show some examples from our group, including high-throughput and/or machine-learning screening and the design of 2D electrocatalysts, electrides, piezoelectrics, ferroelectric materials, and heterostructured solar cells. Finally, we also summarize recently published papers that utilize the data sets in the 2DMatPedia database.

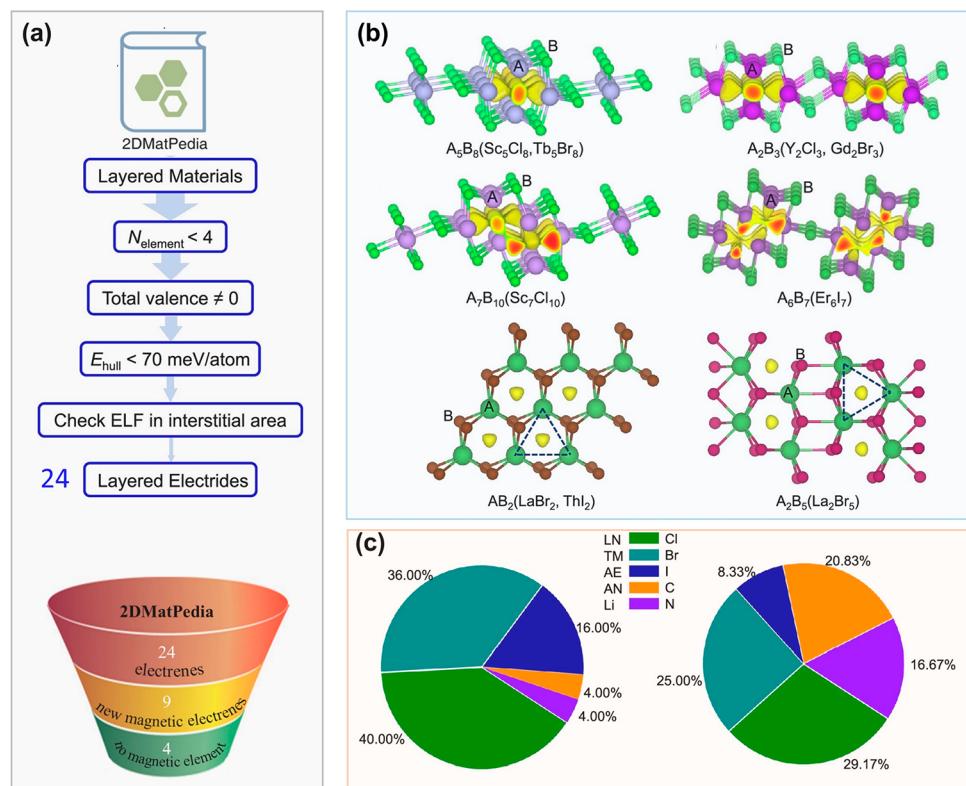
#### 3.1. Electrocatalysts

Electrocatalysis is of great importance to the conversion, storage, and utilization of renewable energies for energy sustainability.<sup>24,26,32</sup> The hydrogen evolution reaction (HER) is the simplest but most promising electrochemical process, which is the cathodic half-reaction of water splitting that produces hydrogen (an energy-dense clean fuel).<sup>8,34</sup> The nitrogen reduction reaction (NRR) is another important electrochemical reaction for providing ammonia fertilizers from abundant N<sub>2</sub> in the atmosphere.<sup>35</sup> The electrocatalysts play a key role in achieving a high conversion efficiency. However, the best electrocatalysts for these two reactions are either noble metal-based or still lacking, especially for NRR under ambient conditions. Among a variety of intensively explored materials, low-dimensional materials (e.g., 0D single-atom catalysts<sup>35,36</sup>

and 2D materials<sup>8</sup>) stand out, where their profound quantum confinement effect significantly modulates the electronic structures and thus the catalytic performance. In addition, the high ratio of exposed surface atoms in such materials greatly improves the atom utilization, which is especially beneficial to the development of noble metal-based electrocatalysts in terms of cost considerations.

**3.1.1. HER of Two-Dimensional Materials.** There is a dilemma in discovering and developing catalysts with 2D materials. Usually, the catalytic performance of experimentally synthesized stable 2D materials is below expectations because of their “stability” (chemical inertness). For example, the active sites of MoS<sub>2</sub> are located only at the fractional edges, whereas the large basal planes are inert for HER.<sup>37</sup> Thus, the methods reported to improve its catalytic performance are either the maximizing of active edge sites or the activation of the inert basal plane, such as by defect engineering.<sup>38</sup> Both require additional complicated treatments. It is thus desirable to search for 2D materials with intrinsically active basal planes, which could naturally provide a much higher density of active sites. Our 2DMatPedia database includes around 3000 top-down 2D materials, providing a wealth of 2D materials for discovering high-performance HER candidates by high-throughput screening<sup>8</sup> or machine-learning<sup>39</sup> algorithms.

Figure 2a,b shows the screening processes and the corresponding criteria. We first applied three criteria to prescreen 2D materials in 2DMatPedia: (i) their parent layered materials that have been experimentally reported in the Inorganic Crystal Structure Database (ICSD), (ii) the upper bound of the exfoliation energy of 268 meV/atom ( $4E_{\text{exf}}^{\text{graphene}}$ ), and (iii) metals with high electrical conductivity. Through the



**Figure 4.** (a) Workflow of the high-throughput screening procedure for layered electrides. The decreasing size of the light-blue arrow indicates the number of candidates after each screening step. Through the process, 24 layered electrides were identified, and 4 among them are ferromagnetic without magnetic elements. (b) Six types of new structure prototypes of nine new electrides which have never been reported before. (c) Statistical information for the 24 layered electrides screened in this work. Cation (left panel) and anion (right panel) distributions by percentage. Reproduced from ref 41. Copyright 2019 American Chemical Society.

above prescreening criteria, we identified 208 compounds, denoted as Candidate Set-208, for further investigation. Most candidates in this set are mainly in the monoclinic, orthorhombic, tetragonal, or trigonal crystal system as shown in Figure 2c. The bridge, hollow, and top adsorption sites of each candidate in Candidate Set-208 are considered, resulting in a total of 1740 adsorption sites. We then performed intelligent high-throughput calculations to automatically iterate over all possible adsorption sites. Afterward, these candidates were further screened according to their stability upon hydrogen adsorption as well as the hydrogen adsorption regime (Figure 2b), through which 208 candidates were further narrowed to 146 (Candidate Set-146). Figure 2d shows the distribution of the Gibbs free energy ( $\Delta\Delta G_{\text{H}^*}$ ) in Candidate Set-146. We finally shortlisted 15 candidates with  $|\Delta\Delta G_{\text{H}^*}| \leq 0.3 \text{ eV}$  and 9 best candidates with a more rigorous criterion of  $|\Delta\Delta G_{\text{H}^*}| \leq 0.15 \text{ eV}$  and the inclusion of the case-by-case zero-point energy contribution. These nine promising candidates are  $\text{IrTe}_2$ ,  $\text{Ce}_4\text{C}_2\text{Br}_5$ ,  $\text{C}_8$ ,  $\text{Pr}_4\text{C}_2\text{Cl}_5$ ,  $\text{Ba}_2\text{Cu}_2$ ,  $\text{NbS}_2$ ,  $\text{NbSe}_2$ ,  $\text{Ti}_2\text{Se}_2$ , and  $\text{TaSe}_2$ , which we use for further experimental investigations. It is worth noting that this 2D HER screening work is updated by the continuous development of the 2DMatPedia database.

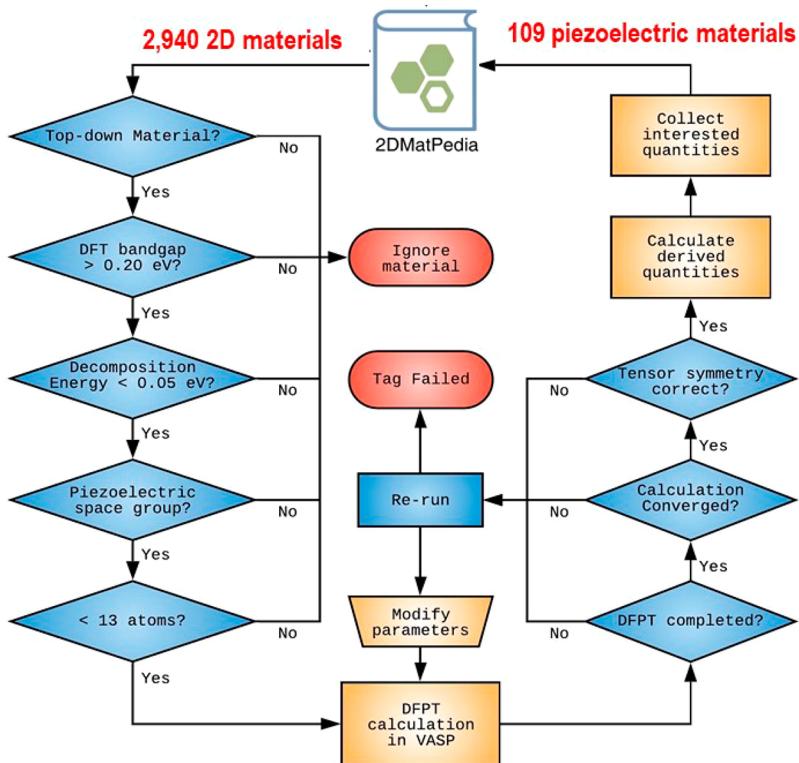
### 3.1.2. NRR on a Two-Dimensional Material Support.

Single-atom catalysts (SACs) have recently been intensively explored with regard to the electrochemical nitrogen reduction reaction because of the 100% utilization, high activity, and high selectivity in which N-doped graphene is widely used to support SACs.<sup>36,40</sup> Through high-throughput DFT screening, we found that monolayer  $\text{MoS}_2$  is a potential alternative substrate to anchoring transition-metal (TM) SACs for NRR.<sup>35</sup> It is found

that the single Mo atom anchored on top of the Mo site of  $\text{MoS}_2$  has the best NRR performance (the lowest limiting potential and best selectivity against HER) among Ag, Au, Co, Cr, Cu, Fe, Mn, Mo, Ni, Pd, Pt, Rh, Ru, Sc, Ti, V, W, and Zn. The estimated limiting potential of  $\text{Mo@MoS}_2\text{-M}$  is around  $-0.44 \text{ V}$  via the distal mechanism. Our results further confirm the stability of  $\text{Mo@MoS}_2\text{-M}$  as well as its good selectivity to NRR against the hydrogen evolution reaction.

Figure 3a shows the workflow of our high-throughput screening process for NRR. Eighteen transition metals are selected for SACs on three different sites (Mo top, S top and hollow) of monolayer  $\text{MoS}_2$ . On the basis of the high-throughput binding-energy calculations (Figure 3b), we identified the most stable anchoring site on  $\text{MoS}_2$  for each transition-metal single atom. Because  $\text{N}_2 \rightarrow \text{N}_2^*$  is the first reaction step and  $\text{N}_2^* \rightarrow \text{N}_2\text{H}^*$  is one of the potential limiting steps in NRR, we first calculated the Gibbs free energy change of these two steps as the screening criteria (Figure 3c,d). On the basis of this result, we shortlisted two promising candidates,  $\text{Mo@MoS}_2\text{-M}$  and  $\text{W@MoS}_2\text{-M}$  (Figure 3d). Through further systematic investigations of the stability, activity, and selectivity against HER,  $\text{Mo@MoS}_2\text{-M}$  is finally identified as the best NRR catalytic system.

In summary, a high-throughput screening approach is a powerful tool for discovering 2D materials having good electrocatalytic performance or enabling synergistic catalysis with the nanocatalysts as the support. Beside monolayer 2D materials, a rational design, such as the heterostructure and Janus structure, can effectively improve the catalytic performance.<sup>9</sup>



**Figure 5.** Workflow of the high-throughput screening to identify piezoelectric 2D materials. Several criteria were chosen to ensure the stability and existence of dipoles. Several (109) candidates are identified from 2940 2D materials. Reproduced with permission from ref 51. Copyright 2022 Springer Nature.

### 3.2. Layered Electrides and Electrenes

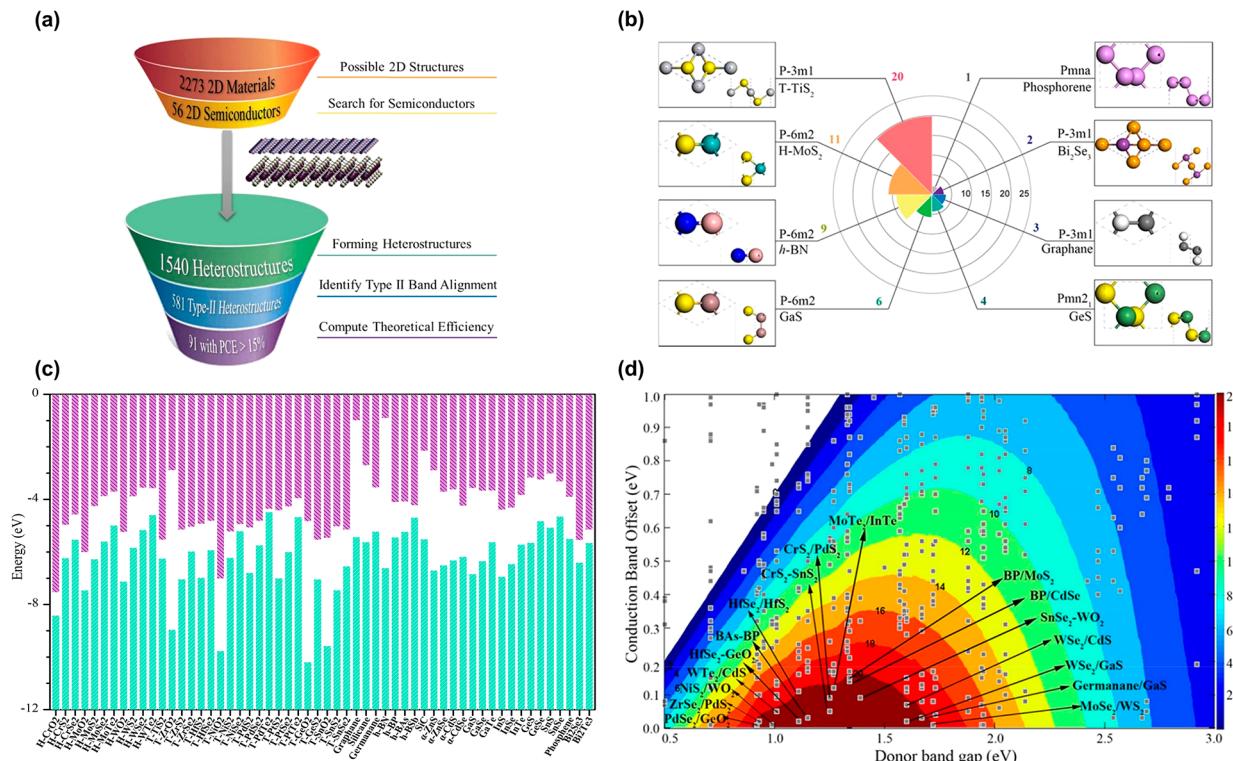
Electrides or electrenes in a monolayer are intrinsically electron-rich materials, in which excess electrons (called anionic electrons) are confined in geometrically interstitial sites and loosely bound, acting as anions.<sup>41,42</sup> It has been demonstrated that electrides exhibit many unique properties and have potential applications, such as superconductivity, topological matters, and catalysis. Despite their exotic properties, electrides, especially layered electrides, are scarce, with only a few compounds grown experimentally. By high-throughput searching for layered materials with anionic electrons in 2DMatPedia, we identified 24 layered electrides (Figure 4a).<sup>41</sup> Nine of them have unique compositions that have never before been reported (Figure 4b). Statistical information for the 24 layered electrides shows that the ions are mainly early transition metals, alkaline earth metals, lanthanides, and actinides (Figure 4c). Interestingly, we found nine magnetic electrenes. To our surprise, four of them ( $\text{LaBr}_2$ ,  $\text{La}_2\text{Br}_5$ ,  $\text{Sc}_7\text{Cl}_{10}$ , and  $\text{Ba}_2\text{LiN}$ ) contain no magnetic elements.<sup>42</sup> The underlying physics of long-range ferromagnetism in electrenes is unrevealed by low-energy effective Hamiltonian models.<sup>42</sup> These screened materials significantly increase the number of layered electrides and 2D ferromagnetic materials and expand the exploration scope and depth in the area of electrenes and ferromagnetic materials.<sup>43,44</sup>

### 3.3. Two-Dimensional Ferromagnets

Magnetism has been explored in 2D materials for more than a decade. (See ref 12 and references therein.) In 2017, the first observations of intrinsic ferromagnetism in pristine 2D crystals were reported for  $\text{Cr}_2\text{Ge}_2\text{Te}_6$  and  $\text{CrI}_3$ .<sup>44</sup> Soon after, a variety of 2D (anti)ferromagnetic materials, such as  $\text{Fe}_3\text{GeTe}_2$ ,  $\text{VSe}_2$ ,  $\text{MnSe}_2$ , and  $\text{MnBi}_2\text{Te}_4$ , were reported experimentally. (See the

review in ref 42 and references therein.) Most reported 2D magnets work only at low temperatures. However, this does not fundamentally exclude the possibility of high-temperature 2D magnets. Efforts toward this goal using high-throughput DFT calculations and machine learning have shown promise.

We have carried out high-throughput spin-polarized calculations on all 2D materials in 2DMatPedia,<sup>18</sup> and we have identified 230 compounds with magnetic moments greater than  $0.1\mu_B$  per unit. We further set at least one magnetic element (Gd, Fe, Cr, Mn, Co, Ni, V, and Rb) and at least one heavy element ( $Z > 49$  ensuring a large SOC) per formula unit. Notice that a large SOC is necessary to provide uniaxial magnetic anisotropy, which can resist thermal fluctuations by opening up a magnon excitation gap or breaking the gapless long-wavelength excitations (spin wave), thus lifting the so-called Mermin–Wagner–Hohenberg restriction (under the isotropic magnetic Heisenberg model) and generating long-range magnetic order and high-Curie-temperature ferromagnets. The solution to the Mermin–Wagner restriction by Onsager of the 2D Ising model shows that the large magnetic anisotropic energy in 2D materials can provide uniaxial anisotropy, resulting in a magnetic phase transition at  $T_c > 0$ . On the basis of the above screening criteria, we then selected 83 candidates for further magnetic coupling calculations within ferromagnetic and several antiferromagnetic configurations. We finally identified 40 ferromagnetic candidates. Their Curie temperature is obtained by an analytical solution based on the 2D Ising model, and nine of them may be ferromagnetic at room temperature. Utilizing the magnetic information above as the data sets, we developed machine-learning models to classify the 2D materials into the respective categories of nonmagnetic and magnetic materials with an accuracy of 93.4% via the gradient boost algorithm. The latter is



**Figure 6.** (a) Workflow of the design of heterostructures and high-throughput screening criteria for high-efficiency type-II solar cells. (b) Polar histogram of the eight categories with their space groups, representative materials, and the number of structures involved. (c) Band alignment of all of the 2D materials with respect to the vacuum level. (d) Simulated power conversion efficiency of all type-II heterostructures studied in this work. The background with different color regions refers to different levels of PCE (color bar on the right). The heterostructures with PCE greater than 20% are labeled in this figure as A/B, where A represents the donor and B represents the acceptor. Reproduced from ref 14. Copyright 2018 American Chemical Society.

further classified into antiferromagnetic and ferromagnetic materials with an accuracy of 94.4% using the random forest classifier. A web-based AI application for predicting 2D magnetic materials has been developed and can be found at <https://twodferromagnetism-model.herokuapp.com>.

Such 2D ferromagnetic materials information, which agrees well with recent experimental reports, is expected to foster practical applications of 2D magnetism, such as 2D spintronic or spin-orbitronic devices.<sup>13</sup> Spin-valley locking and spin-valley polarization have been reported in 2D materials (e.g., MoS<sub>2</sub>) on magnetic substrates.<sup>45–48</sup> Magnetic tunnel junctions with 2D magnets as tunneling barriers exhibit giant tunneling magnetoresistance.<sup>12</sup> New concepts of spin field-effect transistors based on 2D magnets have been reported as well. Furthermore, the exotic spin textures, quantum phases, and quasiparticles in 2D magnetic materials and heterointerfaces can lead to new methods of computation and communication. We envision that those successive breakthroughs in 2D magnets could usher in a new era of information technologies with exciting applications in computing, sensing, and memory.<sup>3,4,49</sup>

### 3.4. Two-Dimensional Piezoelectrics

Piezoelectric materials enable the interconversion between mechanical and electrical energy. This is made possible by the change in polarization of the material when it is stretched or compressed. As such, piezoelectric materials are integral components of intelligent, multifunctional devices and drive a multi-billion-dollar industry through their applications as sensors, actuators, and energy harvesters. The recent thrust toward flexible nanoscale devices creates a need for 2D

piezoelectric materials, such as wearable sensors and smart material applications that require a large voltage signal in response to a small amount of physical deformation.

So far, the discovery of 2D piezoelectric materials has mostly been by trial and error or materials intuition, for example, by searching for either in-plane or out-of-plane 2D ferroelectric materials,<sup>10,50</sup> because ferroelectrics must be piezoelectric. However, there is no direct relationship between the piezoelectric coefficient and the ferroelectric coefficient, and a material with a large ferroelectric polarization may have a very small piezoelectric coefficient. Experimentally, it is also challenging to quantitatively compare the piezoelectric coefficients of 2D materials. To address this problem, we performed a systematic high-throughput search through the 2DMatPedia database to discover 2D piezoelectric materials and identify candidates with large intrinsic piezoelectric coefficients.<sup>51</sup> Figure 5 shows the workflow with screening criteria. We identified 109 2D piezoelectric materials from 2940 top-down 2D materials in 2DMatPedia. Interestingly, 46 piezoelectric materials show out-of-plane piezoelectricity. Our findings provide a platform for the development of flexible nanoscale piezoelectric devices, such as high-precision actuators, wearable electronics, and energy harvesters.

### 4. RATIONAL DESIGN OF van der Waals HETEROSTRUCTURES

The combination of two or more different monolayers into one vertical stack via van der Waals forces, i.e., 2D heterostructures, not only tremendously increases the number of 2D materials but

also brings about a variety of new properties, due to the synergistic or coupling effect, that do not exist in each constituting material.<sup>1</sup> Such heterostructures have already led to numerous exciting properties and a range of applications, such as field-effect tunneling transistors, plasmonic devices, and light-emitting diodes.<sup>1</sup>

To identify van der Waals vertical heterostructures with high power conversion efficiencies (PCEs) for the target application of flexible solar cells, we conducted a high-throughput screening (Figure 6a) of the band alignments of 1540 vertical heterostructures formed by 56 2D semiconducting materials (Figure 6b).<sup>14</sup> On the basis of the band alignment of two different semiconducting compounds (Figure 6c), we identified more than 90 heterostructures with estimated PCEs that were greater than 15%, of which 17 heterostructures (labeled in Figure 6d) are predicted to have PCEs greater than the best value (20%) reported or proposed in the literature. Taking this example, we demonstrate that van der Waals heterostructures not only increase the members of the 2D materials family but also realize interesting phenomena for a variety of applications. More 2D van der Waals heterostructures and their properties will be provided in 2DMatPedia, such as Z-scheme heterojunction photocatalysts for overall water splitting.

## 5. OTHER PUBLICATIONS RELATED TO THE 2DMatPedia PLATFORM

Finally, we briefly summarize some recent papers utilizing the data sets in 2DMatPedia. Using the similar bottom-up approach in 2DMatPedia, Sorkun et al. used machine learning to develop a “virtual” 2D materials database with 316 505 compounds.<sup>52</sup> The accuracy of the predicted band gaps with ML models is tested and validated by 2DMatPedia data sets.<sup>52</sup> From Materials Cloud, Karmodak et al. screened out only 15 promising HER catalysts from 258 2D materials with  $E_{\text{exf}} < 35 \text{ meV}/\text{\AA}^2$ .<sup>53</sup> For deeper data mining, Wu et al. used a deep-learning method (at near-DFT accuracy) to screen out 38 high-performance HER catalysts from 6531 2D materials.<sup>39</sup>

The electronic structures are the main part of 2D materials databases. By combining 2DMatPedia with C2DB, Liang et al. developed a rational machine-learning model for the band gap and exciton binding energy of 2D materials, which can reveal the physical pictures behind the predicted quantity by machine learning.<sup>54</sup> Liu et al., via screening the 2DMatPedia database, identified 15 monolayer atomic crystals hosting the unique topological flat-band feature near the Fermi level.<sup>55</sup> Schleider et al. identified 15 new topological 2D materials by screening 2D material databases with ML algorithms.<sup>56</sup> Kabiraj et al. developed an ML model to predict the ferromagnetism of 2D materials with the test set of new and complex compositions in 2DMatPedia.<sup>57</sup> Lu et al. developed active learning by integrating ML with high-throughput calculations, which first learns ferromagnetic materials from C2DB and then predicts 542 ferromagnetic and 917 antiferromagnetic materials from Materials Cloud and 2DMatPedia databases.<sup>58</sup> Leveraging the van der Waals interaction between 2D heterostructures for novel solid lubricant and superlubricant applications, Fronzi et al. used a machine-learning approach to create a data set of the interlayer energy and the elastic constant of 18 million van der Waals heterostructures based on 6000+ monolayer 2D materials in 2DMatPedia.<sup>59</sup> Data analytics and applications based on 2DMatPedia can be expected to increase rapidly.

## 6. CONCLUSIONS AND OUTLOOK

We aim to develop a capability in 2D materials informatics, revolutionize new 2D materials discovery, and shorten the time from discovery to applications, enabled by the development of the world’s largest comprehensive 2D materials database (2DMatPedia), high-throughput screening and ML models, web toolkits, and device applications. Our next goals are to (i) further increase the collection of candidates in 2DMatPedia through a bottom-up approach, alloying, forming heterostructures, and collecting 2D materials data from the public literature, (ii) adapt advanced AI algorithms for the analysis of a variety of material properties to find descriptors to accelerate the design of new 2D materials, and (iii) utilize the platform to screen desired 2D materials for specific applications in close collaboration with experimentalists.

Despite the significant progress and successful deployment of the materials genome, data-driven materials discovery, high-throughput calculations, and machine learning as a major game changer in the area of 2D materials science, future challenges remain:

- (1) Different research groups may create their own databases containing the calculated properties of existing and hypothetical 2D materials, such as JARVIS, C2DB, Materials Cloud, and 2DMatPedia. Their collaboration and integration will be of great use to researchers and users in the field of 2D materials science. Thanks to OPTIMADE, a user-friendly materials-design platform, several materials databases (including 2DMatPedia; <https://www.optimade.org/providers-dashboard/>) have recently been integrated.
- (2) New complex 2D materials may not be discovered by either the top-down or bottom-up approach, such as recently synthesized  $\text{MnBi}_2\text{Te}_4$ . AI-enabled literature mining to collect 2D materials and their properties from the public domain is an important step in building a comprehensive 2D materials database.<sup>60</sup>
- (3) High-throughput calculations have a so-called speed–accuracy trade-off. Most large computed materials databases are constructed using conventional DFT functionals for computational efficiency, which have well-known issues in strongly correlated materials, van der Waals heterostructures, and band gap materials. This needs to be addressed by methodological advances in the future.
- (4) Limited by the DFT core, the high-throughput calculations still cannot handle a very large number of samples, such as the catalysis with complex reaction paths and active reaction sites. One solution is to apply active learning, a machine-learning-assisted DFT calculation.<sup>24</sup> The other solution is to develop models to access beyond-DFT scales at near-DFT accuracy, such as materials/physics-based deep-learning algorithms.<sup>27</sup>
- (5) AI tools are being eyed with suspicion by some scientists because of the “black box” models which are as opaque to our understanding as the data patterns themselves. Interpretable AI models for materials science are strongly desired in the future. The materials science, physics, and chemistry communities may learn from the ML community to transfer and implement advanced algorithms into their domains with discipline interpretability. The recent effort led by Jeffery C. Grossman at MIT is a good example in this direction. They implemented graph

convolutional neural networks, a widely used algorithm for computer vision, to successfully explain the prediction of material properties.<sup>61</sup>

- (6) Finally, the computational community has a close collaboration with the experimental community. Any new discovered or designed candidate 2D materials for high-performance applications must be verified and validated by experiments.<sup>24</sup> Inversely, experimental feedback can be used to optimize the theoretical models to provide more accurate calculations and predictions.

We believe that future efforts in the computational 2D materials community are certainly not limited to the six points listed above. The family of 2D materials is continuously growing in terms of both variety and quantity, and almost every new 2D material has unique properties for one or more applications. The rapid development of this field will finally accelerate the progress of novel 2D materials from discovery to deployment in new technologies.

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