Screening and understanding Li adsorption on 2-dimensional metallic materials by learning physics and physics-simplified learning

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1. Appearance frequency of elements and crystal systems in the subset

In this work, because the GCN potentials and random forest models are built based on atomic properties and geometric information, the models should have transferability across different compositions and structures. However, if the chosen subset is highly biased, compositionally or geometrically, then the data from the subset might not be representative for human or machine learning models to the learn the underlying pattern in the whole materials space of interest. To check if the sampled subset is biased, we show the appearance frequency of elements and crystal system of the 180 materials subset and the 3,499 metallic 2D materials in Figure S1, from which one can see that, compared with all the 2D metallic materials, the element and crystal system distribution in the 180 materials subset are not biased to one end or concentrated on one location, therefore we think that the subset is not biased.
Figure S1. **a** and **b** Appearance frequency of elements and crystal system in the 180 materials subset and 3,499 2D metallic materials, respectively.

2. **Distribution of distances between adsorbed Li and the nearest atom in the 2D materials**

The distribution of adsorption configurations used for training the GCN potentials is of interest as those configurations are generated from active sampling. The distribution of distance between adsorbed Li and the nearest atom in the 2D materials is plotted in Figure S2, from which one can see that sites close to the 2D sheets are sampled more frequently while sites with moderate and large distance are sampled with similar frequency. This behavior is in accordance with the physical intuition that energy variation with respect to interatomic distance is larger when atoms are closer to each other, which shows that the active sampling in this work tends to sample more data points from the region harder to learn while ensures that the whole space of interest (space of distance here) is included in the training set.
Figure S2. Distribution of distances between adsorbed Li and the nearest atom in the 2D materials.

3. Comparison between linear regression, support vector regression and random forest

Figure. S3. a, b and c. Mean Average Error (MAE, in eV) of leave-one-out cross-validation versus regularization term (linear regression and support vector regression) or number of estimators (random forest) for linear regression, support vector regression and random forest, respectively.

4. Discussion about learning $E_{\text{ads}}$, $E_{\text{cp}}$ and $\Phi$ by random forests.
As discussed in the main text, learning $E_{\text{ads}}$ is essentially learning $E_{\text{cp}}$ and $\Phi$ at the same time, making learning $E_{\text{ads}}$ more challenging than learning $E_{\text{cp}}$ or $\Phi$ alone. In addition to the MAEs as shown in the paper, here we plot the feature importance from learning $E_{\text{ads}}$, $E_{\text{cp}}$ and $\Phi$ in Figure S3, from which we can see that the top features from learning $E_{\text{ads}}$ are almost equally composed of top features from learning $E_{\text{cp}}$ and $\Phi$, showing that when learning $E_{\text{ads}}$, the random forest model is trying to simultaneously learn $E_{\text{cp}}$ and $\Phi$.

Figure S4. a, b and c Feature importance from random forest models for learning $E_{\text{ads}}$, $E_{\text{cp}}$ and $\Phi$, respectively. Only features with high importance are plotted.

5. **Standard deviation of covalent radius versus adsorption height.**

Figure S5. Adsorption height versus standard deviation of covalent radius of components in 2D materials.
6. **SHAP analysis of the impacts of features on model output \((E_{cp})\)**

A SHAP analysis\(^1\) is performed and the impacts of features on model output \((E_{cp})\) are plotted in Figure S5. One can see that the most impactful features from the SHAP analysis are mostly overlapped with the most correlated features from \(R^2\) scores shown in Figure 3a and most important features from feature importance shown in Figure S3b. One of advantages of SHAP analysis is that it summarizes the most impactful features and how those features impact the model output concisely. For example, one can see that larger standard deviation of covalent radius of elements, the model tends to predict larger \(E_{cp}\), and larger number of atom per area, the model tends to predict larger \(E_{cp}\), which provide similar information as in Figure 3 in a more concise way. Here add the SHAP analysis as a supplement to the \(R^2\) correlation in Figure 3 and feature importance in Figure S3.
**Figure S6.** Distribution of the impacts (SHAP values) of compositional and structural features on the model output ($E_{cp}$), respectively. The color represents the feature value (red high, blue low), and here only the top 12 features with the highest sum of absolute SHAP values are shown.

### 7. Enhancing Li adsorption ability of graphene

**Figure S7.** Structures of B-doped graphene and F-functionalized graphene, respectively.

### 8. Li-Li interaction energy and substrate relaxation energy

In this work, the Li-Li interaction energy term ($E_{electro.}^{Li-Li}$) is not considered as there is only one Li introduced into each supercell of 2D material to simplify the problem. Even though the Li-Li interaction is not considered, the obtained $E_{ads}$ is still valuable as generally materials with more negative $E_{ads}$ at dilute concentration have higher average voltage and $E_{ads}$ at dilute concentration is extensively used as a metric to measure the Li adsorption ability of materials$^{2-20}$. The energy term related to substrate relaxation ($E_{relaxation}^{subs}$) is also not considered in this work because calculating this
term requires geometry relaxation of substrate, which is expansive for high-throughput screening. In fact, due to the openness of 2D materials\textsuperscript{21}, in general the degree of structural change associated with Li adsorption is small for 2D materials such as (doped-) graphene\textsuperscript{12, 22}, g-C\textsubscript{3}B\textsubscript{23}, MXene\textsuperscript{2} and MoS\textsubscript{2}\textsuperscript{24}, and $E_{\text{relaxation}}^{\text{subs}}$ is proved to contribute much less than other energy contributions for 2D MoS\textsubscript{2} and VS\textsubscript{2} in both 2H and 1T phases\textsuperscript{25}, suggesting that structural relaxation for Li adsorption on 2D materials is not as important as that of Li insertion into bulk materials where strong structural deformation is observed\textsuperscript{26, 27} because Li is confined in all the three dimensions in space. Here, ignoring $E_{\text{electro}}^{\text{Li-Li}}$ generally makes $E_{\text{ads}}$ more negative, and ignoring $E_{\text{relaxation}}^{\text{subs}}$ makes $E_{\text{ads}}$ more positive because substrates are not relaxed in the DFT calculations. These two contributions can be included in future high-throughput screening works with more time-efficient first-principle calculation methods or more data-efficient machine learning potentials.

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