

An Initial Design-enhanced Deep Learning-based Optimization Framework to Parameterize Multicomponent ReaxFF Force Fields

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ReaxFF is an empirical interatomic potential capable of simulating reactions in complex chemical processes and thus determine the dynamical evolution of the molecular systems. A drawback of this method is the necessity of a significant preprocessing to adapt it to a chemical system of

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interest. One of the preprocessing steps is the optimization of force field parameters that are used to tune interatomic interactions to mimic ones obtained by quantum chemistry-based methods. The optimization of these parameters is a very complex high dimensional problem. Here, we propose an INitial-DEsign Enhanced Deep learning-based OPTimization (INDEEDopt) framework to be used in ReaxFF parameterization. The procedure starts with a Latin Hypercube Design (LHD) algorithm that is used to explore the parameter landscape extensively. The LHD passes the information about explored regions to a deep learning model for training. The deep learning model finds the minimum discrepancy regions and eliminates unfeasible regions, which originate from the unphysical atomistic interactions, and constructs a more comprehensive understanding of a physically meaningful parameter space. The procedure was successfully used to parametrize a nickel-chromium binary force field and a tungsten-sulfide-carbon-hydrogen quaternary force field and produced improved accuracies in shorter periods time compared to conventional optimization method.

INTRODUCTION

Atomistic-scale insights have been critical to understanding the dynamical evolution of chemically reactive systems and therefore have created a demand for the development of computational chemistry techniques. Quantum mechanics (QM)-based atomistic simulation methods have been commonly used to study the molecular dynamics (MD) because these methods provide accurate energies, charges, and reaction pathways. However, simulation times and sizes are highly constrained by computational cost, and these limitations are some of the motivations behind the development of empirical potentials. The empirical potentials provide fast access to forces; and, in turn, the dynamical evolution of much larger chemical systems for longer simulation times in

various conditions (e.g., temperature, pressure). However, due to fixed connectivity between atoms, traditional empirical potentials have limited capability of modeling the evolution of systems during reactive events. In order to bridge the gap between QM and empirical potential-based methods, several reactive force field-based atomistic simulation techniques have been developed¹⁻⁴. ReaxFF is one of the widely used reactive interatomic potential in this category due to its reliability and transferability between chemical systems. Initially, ReaxFF interatomic potential has been formulized to model hydrocarbons, and then extended through silica, nitramine-based materials to several aqueous and combustion systems⁵. Currently, the ReaxFF method covers over 50 elements; and is applicable to a wide range of chemical systems that are of interest to materials science community (i.e., 2D materials⁶⁻¹³, electrochemistry^{14, 15}, thin films^{16, 17}, nanotubes^{18, 19}, catalysts^{20, 21}).

ReaxFF uses bond-order and charge-dependent functionals to define intra- and interatomic interactions. The simplest form of the total energy equation used in ReaxFF can be given as below, and a detailed and most up-to-date form can be found in Ref²²:

$$E_{system} = E_{bond} + E_{angle} + E_{tors} + E_{Coulomb} + E_{vdW} + E_{over} \quad \text{Equation 1}$$

where E_{bond} , E_{angle} , E_{tors} are energy terms related to bond formation, three-body valence angle strain and four-body torsional angle strain, respectively. The energy term, E_{over} , is used to prevent the over coordination of atoms. The energy terms, $E_{Coulomb}$ and E_{vdW} , are associated with electrostatic and dispersive contributions, respectively. Each separate energy term in Equation 1 contributes to the total energy of the system, which in turn, predicts the dynamical evolution. Each term is calculated by an equation involving several parameters to be optimized to adapt the calculation to

a form specific to a chemical system of interest. A typical ReaxFF force field consists of approximately 100 of these parameters per element type depending on the element. Because the intra- and interatomic interactions are tuned by these parameters, they are optimized to reproduce reference values with reasonable accuracy before moving to production simulations. These reference values form a force field training (FF-training) set, which is composed of, but not limited to, molecular properties (e.g. bond lengths, bond angles, charges and energies etc.), and/or chemical reactions from as simple as bond breaking/formation to more complex such as vacancy dynamics, ion diffusion of reference systems. The reference values, which hereafter will be referred to as “reference properties”, are obtained using QM-based methods if experimental values are not present.

The quality of the production simulations is strictly affected by the quality of the developed force field; therefore, the force field parametrization is one of the most critical parts of the MD studies. ReaxFF uses fixed functions for all chemical systems; hence, application to different chemical systems and/or different applications of similar chemical systems require adaptation of ReaxFF through optimization of the force field parameters. The optimization process requires a comprehensive exploration of the force field parameter landscape. However, optimization of such a large number of parameters, especially for multicomponent chemical systems, is challenging to achieve. A typical force field for a multi-component system contains several hundreds of parameters, a significant part of which can be transferred from previously optimized force fields; however, depending on the complexity of the molecular system, a few tens of parameters should be optimized to reproduce reference properties in a given FF-training set. A parameter space that is composed of tens of parameters can be considered as high-dimensional and can have a complex geometry with many local minima.

Each local minimum in ReaxFF parameter space yields acceptable molecular property values and therefore can be reliably used by researchers for the application of the developed force field in the production stage. However, being stuck in a local minimum may be troublesome from the parametrization perspective. For example, it is a very common practice to update FF-training sets to extend the applications, and this update requires reparameterization of the force field because the previously found local minimum corresponding to an ideal parameter set may no longer be appropriate for newly added reference properties. Another issue that may arise during force field development is due to diversity in the FF-training set. A typical ReaxFF training set is composed of molecular and condensed phase parts, which are also separated into groups such as the equation of states, the heat of formations, etc. It is very likely for a force field parameter set to get stuck in a local minimum that precisely reproduce reference properties corresponding to a specific part of the force field while poorly fitting to the remaining.

The ability to have the information of several local minima in parameter space and so making transitions between these minima is significantly beneficial to detect low discrepancy regions that are capable of precisely fitting to the whole FF-training set, including all sections. The conventional optimization method that is widely used by the force field developers is a sequential one parameter parabolic extrapolation method^{23,24}, which is not capable of switching between local minima to detect lowest error regions in parameter space. In contrast, this method is susceptible to being stuck in a local minimum due to its sequentiality, which also prevents parallelization of the optimization algorithm. Due to these limitations in the conventional method, considerable effort has been directed toward finding a solution to this optimization problem using heuristic methods²⁵⁻³⁰. Iype et al. have used a Monte Carlo (MC) algorithm combined with simulated annealing (SA) procedure to find global optimum values for ReaxFF parameters of magnesium-salt hydrates²⁵.

The application of SA method varies depending on the system; therefore, it cannot offer a common solution to the global force field optimization problem. Several studies have used single and multi-objective genetic algorithm (GA) methods to optimize ReaxFF force field parameters²⁶⁻³⁰. The GA methods require a decent population size to obtain good results and are inefficient for scanning the parameter space comprehensively.

Here we present an INitial-DEsign Enhanced Deep learning-based OPTimization (INDEEDopt) framework that will not only accelerate the force field development for simple chemical systems (e.g., ternary, quaternary or quinary component), but also, enable the development for multi-component ones (e.g., senary or larger), which have become crucial with the advances in materials discovery (e.g., high entropy alloys, inorganic-organic interfaces, etc.). The framework uses machine learning (ML) due to its capability of modeling highly correlated high dimensional data and adapts an initial design algorithm to explore the high-dimensional parameter space comprehensively. Thus, INDEEDopt is capable of finding several local minima in parameter space and produces optimized force fields to be used in the production stage of simulations. Most of the optimization methods developed until today require an assignation of initial values to parameters of interest; however, there is no certain rule to assign these values. INDEEDopt does not require the assignation; in fact, it can be used as a method to generate the initial values that can be further optimized by combining with other optimization methods. The procedure that INDEEDopt framework uses is fully parallelizable and can be used with any ReaxFF-MD software without modifications to the code. INDEEDopt as a concept can be adapted to the parametrization of classical and other reactive force fields in the future if desired.

METHODS

INDEEDopt Force Field Optimization Framework

INDEEDopt framework involves three stages, and a schematic representation of the framework can be seen in Figure 1. In the first stage, the high dimensional parameter space is sampled by means of an initial design algorithm called Orthogonal-maximin latin hypercube design (OMLHD). The OMLHD algorithm can generate parameter combinations within ranges specific to each parameter that are multidimensionally uniformly distributed by reducing the pairwise correlation and maximizing the distance between parameters ³¹. The parameter combinations are given to the ReaxFF-MD code for energy minimization calculations, which returns target property values correspond to each reference property in FF- training set. The parameter combinations and corresponding property values produce a training set for the second stage of the framework, which is the ML model training stage, and hereafter as this training set will be referred to as ML-training set.

The strong correlations of ReaxFF force field parameters create very complex parameter-property relationships because these correlations do not only come from analytical expressions but are also correlated due to the many-body character of atomic interactions specific to the system. In order to handle these complex correlative relationships, we adapted deep learning (DL) as the ML method in the framework, because DL is a promising approach in extracting high-level features from less featured input data ³². The DL involves a type of artificial neural network with multiple hierarchical hidden layers. Using these multi-layered network structures, DL models are capable of nonlinearly mapping the relationships between a given set of inputs and corresponding outputs. This capability makes DL an appropriate method to obtain features from a large data set. In

addition, DL models can be easily parallelized on CPU or GPU architectures, which is one of the goals of this study.

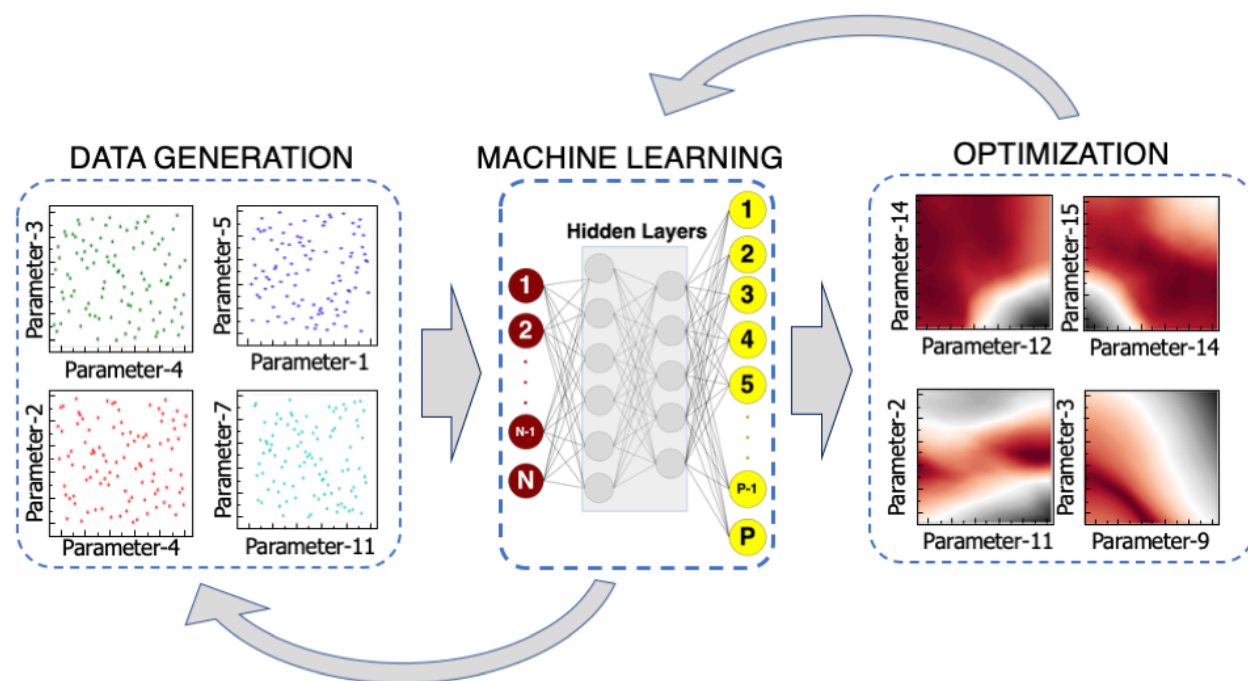


Figure 1. Schematic representation of the DL-based optimization procedure. (Left) Representation of the sampling using the initial design algorithm; (Middle) Deep neural network structure, red circles are input nodes, and yellow circles are output nodes, grey circles are hidden layer nodes; (Right) Representation of the parameter-discrepancy correlation in parameter space.

In the DL model, the ReaxFF parameters (N) are given as an input to a deep neural network (DNN), which is trained to return target molecular property values (P) (Figure 1 (middle)). The DNN used in this study is a fully connected feed-forward type, which transfers data from the input to the output layer through hidden layers. Each layer is composed of several nodes that are connected to each of the other nodes in the previous and next layers. Nonlinear mapping is implemented into neural networks using activation functions. For all layers except the last layer, rectified linear unit (ReLU), which has been proven to accelerate the training process³³, is employed as an activation function, and we use a linear activation function for the last layer. The shape of a neural network

used in DL model affects prediction accuracy³⁴; therefore, to understand the complexity of the neural network used in INDEEDopt, we analyzed the effect of the network shape to the final prediction. The accuracy of prediction was measured according to the mean absolute error (MAE) between the model prediction and test data. According to our analysis, MAE decreases until the number of layers is increased over ten. After ten, the model tends to overfit to the data, thus results in a reduction in accuracy (Figure 2). We used two hidden layers in our model because the accuracy difference between ten and two layers is not significant enough to negatively affect the optimization procedure. The effect of the number of nodes per hidden layer was analyzed by changing the node amount in each layer by multiples of the number of reference property values in the FF-training set. As can be seen from Figure 2, the increase in the number of nodes results in a decrease in MAE, and the network with 900 nodes per layer fits best to the test data. However, the improvement in accuracy is not significant, while the increase in node number is inefficient in terms of computational time. In the light of these analyses, two hidden layers containing a constant number of nodes, which was selected as the size of the FF-training set, is considered as the most appropriate neural network configuration to be used in INDEEDopt framework. For a more complex FF-training set that is composed of thousands of reference property values, there may be a need to assign more hidden layers and more nodes per layer; however, our configuration produced sufficient accuracy for both of our parametrization test cases and should be sufficient for most of the parametrizations conducted in literature. The INDEEDopt code is capable of detecting GPU architecture in the computer system and automatically run on it, so it is advisable to use a larger number of nodes and layers if GPU is accessible to force field developer.

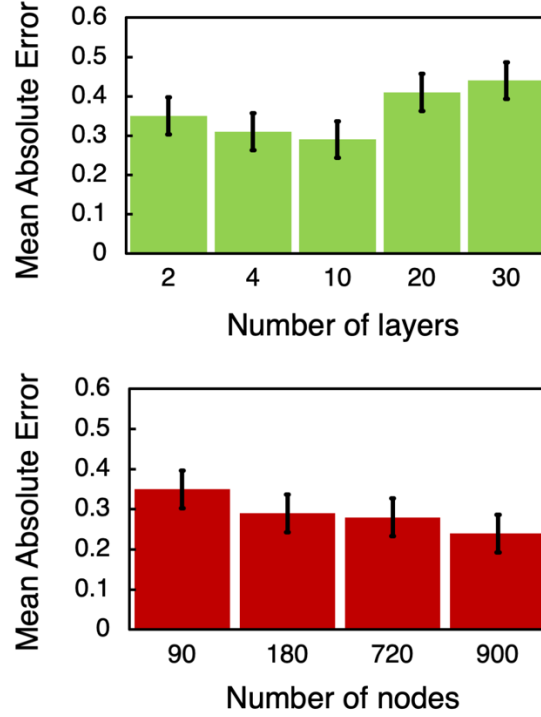


Figure 2. The effect of neural network shape on the accuracy of the DL model. All model training was run until the mean absolute error (MAE) is converged. The change in MAE with respect to the number of hidden layers (up) and the effect of number of nodes per layer to MEA of the trained model (down). The error bars were calculated training each model on the same data with ten repetitions.

The DL training loss function is optimized using Adaptive Moment Estimation (Adam) method, which is computationally efficient and suitable for high-dimensionality due to its adaptive learning rate³⁵. Once the DL model is trained, the accuracy of the model is tested using a test set, which is obtained by randomly selecting a part of the ML-training set as a test set with a split ratio of 0.2. The DL model has an early stopping algorithm, which ensures that the overfitting is avoided by stopping the training at the point where the prediction accuracy is maximum. The maximum fail parameter is set to 100 epochs, while the total number of epochs is selected as 10000. The quality of the DL model predictions is calculated using MAE. The first two stages of INDEEDopt are

iterative and repeat until the DL model converges to an acceptable accuracy. The third stage of the framework, which is the optimization stage, starts once the accuracy is converged. The computational flow diagram of INDEEDopt framework is depicted in Figure 3. We should note that the convergence of DL model is critical, and optimization does not continue until the convergence is achieved. There may be several reasons for a failure/delay in convergence. One of them is the limited sampling of the parameter space, which is very likely a result of a too wide parameter range selection. Another reason may be high uncertainty in parameter-property correlation, which is likely due to the limited representation of troublesome parameters in the FF-training set.

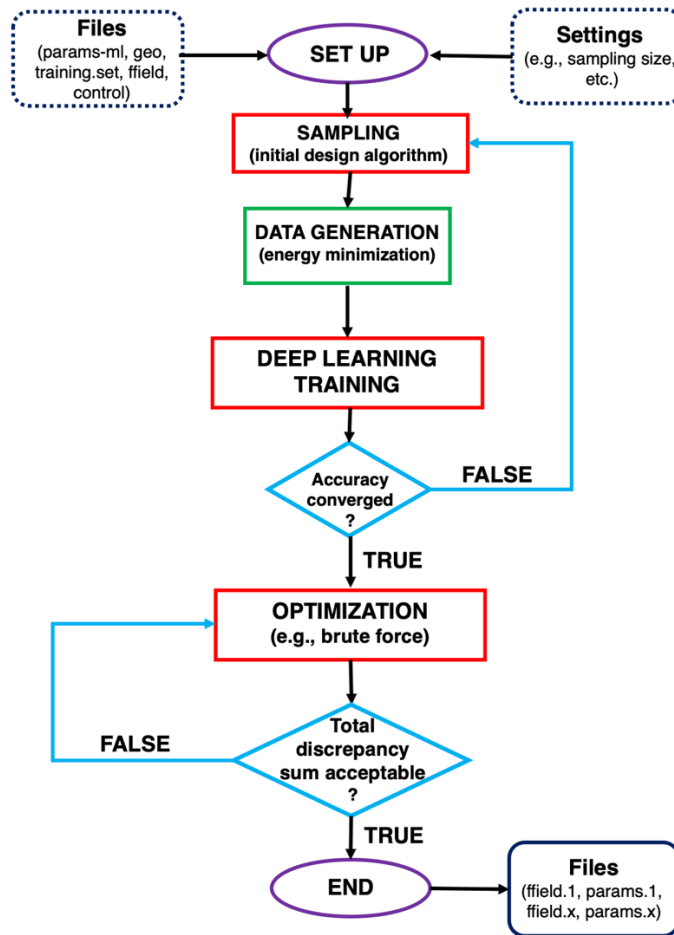


Figure 3. Flow diagram of INDEEDopt framework. Red boxes represent the three main stages of the framework: sampling with initial design algorithms, deep learning model training, and optimization using brute-force method. Blue diamonds represent the conditional transition points in the algorithm. Dark blue dashed boxes represent input files and settings given by the FF developer. The input files that are used in INDEEDopt are same as the ones used in conventional method. Dark blue box represents output files, which are the optimized force field parameters.

In the third stage, a brute force optimization is performed by minimizing the Equation 2.

$$y = \sum_{i=1}^P \left[\frac{x_{i,ML} - x_{i,REF}}{w_i} \right]^2 \quad \text{Equation 2}$$

where $x_{i,ML}$ and $x_{i,REF}$ are the target property obtained by trained DL model, and reference property values in FF-training set, respectively, and w_i is the weight factor, which can be used to adjust the importance of property values during parametrization. The brute force optimization assigns randomly generated values to all parameters at once and calculates the discrepancy (y) between the target and reference property values. This process repeats until stopped by the force field developer when the discrepancy value is satisfactory.

Production of target property values using ReaxFF-MD code takes a significant amount of time during optimization, especially when the reference data set is extensive. This time issue limits the optimization quality as most of the developed algorithms tend to limit their parameter space sampling. To address this issue, INDEEDopt framework calculates the target properties using the trained DL model predictions instead of calling ReaxFF-MD code in the optimization stage. A well-trained DL model is capable of predicting target properties with reasonable accuracy in 3

milliseconds. Therefore, INDEEDopt is capable of exploring a wider parameter space more thoroughly. In addition, the ML-training set, which is generated to train DL model, has information about whole parameter space (in other words, it has the information for several local minima), and therefore can be used to train several parameters sets in parallel by targeting the best local minima in a smaller parameter range similar to Ref ²⁸.

RESULTS AND DISCUSSION

The INDEEDopt framework was tested for two different force field development cases, namely, Ni-Cr binary system for alloy applications and W-S-C-H quaternary system for 2D materials applications. The Ni-Cr system is relatively simple, due to the absence of angular and dihedral terms, which in turn requires optimization of a small number of parameters. The W-S-C-H system is more complex due to several reasons: (1) it has angular and dihedral interactions involved in the training set; (2) the number of parameters to be optimized and the size of the FF-training set is larger than the Ni-Cr one; (3) the force field training set includes both condensed and molecular phases together; therefore, W-S-C-H training was selected as a secondary test case to evaluate the performance of INDEEDopt framework. The same force fields were also parametrized using conventional method, which is the most commonly used method in the literature. All the settings related to energy minimization, molecular geometries in the FF-training set, and weights factors for discrepancy calculations were kept the same for the optimization performances of INDEEDopt and conventional method to be compared.

Ni-Cr force field optimization

The ReaxFF training set of the Ni-Cr alloy system is composed of 90 different reference property values including equations of state and heat of formation energies for different Ni-Cr compositions in fcc and bcc phases. A total of 16 parameters were optimized, which involve bond order terms, bond energy terms, over coordination terms, charge polarization terms and Coulombic interaction terms. A detailed full list of parameters is given in Table S1.

Using OMLHD algorithm, a total of 79636 samples were created to be used as the ML-training set. Each parameter has a corresponding anticipated value range, mostly coming from the chemical nature of the element; however, some parameter ranges are determined by experience. The ranges used for each parameter (in other words, upper and lower limits of parameter search) are given in Table S1. The determination of the ranges is important because wider ranges require more sampling, which is computationally inefficient, may also delay the optimization depending on the complexity of the problem. While exploring the parameter space, some parameter combinations produce meaningless inter or intra-atomic interactions, and if the ranges are large, then a significant amount of the sampling is conducted unnecessarily. For example, the Ni-Cr training set uses a very wide range for parameter-1, which is the bond energy term between Ni and Cr. Our analysis of the correlation between parameter-1 and remaining parameters (Figure S3) shows that a significant part (~80%) of the parameter range produces a large discrepancy meaning that the interatomic interactions are not usable for production simulations. We use the term “infeasible” to refer to these large discrepancy regions and remaining will be referred to as “feasible”. In order to avoid sampling the infeasible parts of the parameter space, INDEEDopt only focuses on regions that produce feasible results after a first complete scan of the parameter space.

A comparison of total discrepancy obtained by INDEEDopt and conventional method for different sections of the training set is given in Figure 4. A detailed comparison of each FF-training set element can be seen in Figure S1. Most of the target properties in the training set are improved using INDEEDopt except some equation of states values Ni_3Cr and Ni_2Cr in FCC and BCC phases, respectively. A significant improvement is observed in the heat of formation energetics, and the lowest total error sum obtained using the conventional method was calculated as 5614.2569, which was reduced to 2804.0238 using INDEEDopt. During the optimization using the conventional method, the heat of formation and equations of state parts of the force field were inversely correlated; therefore, each part was optimized by compromising the accuracy of the other, and both parts were not able to be optimized at the same time. The reason why INDEEDopt succeeded is due to finding another local minimum in parameter space that can satisfy both parts of the force field. The significant differences between some of the final parameter values obtained by INDEEDopt and conventional method also show that the optimum parameter sets are detected in different regions of the parameter space (Figure 5). The final parameters values obtained by both methods are found in Table S1. Some of the parameters were optimized to similar values by both optimization methods, which may be due to a narrow parameter search range assigned by the force field developer. INDEEDopt outperformed the conventional method in terms of timings by producing an optimized parameter set in around two days compared to two weeks by an experienced force field developer. However, we should note that the conventional method was run on a single core while INDEEDopt was run on a hundred cores.

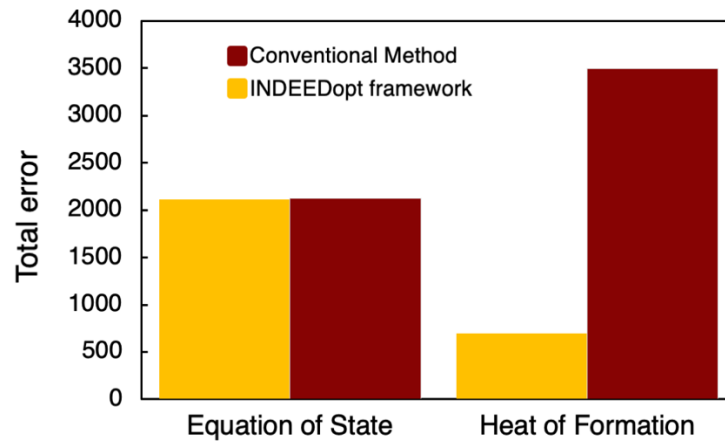


Figure 4. The total error values corresponding to different parts of the nickel-chromium force field training set calculated by INDEEDopt and conventional methods.

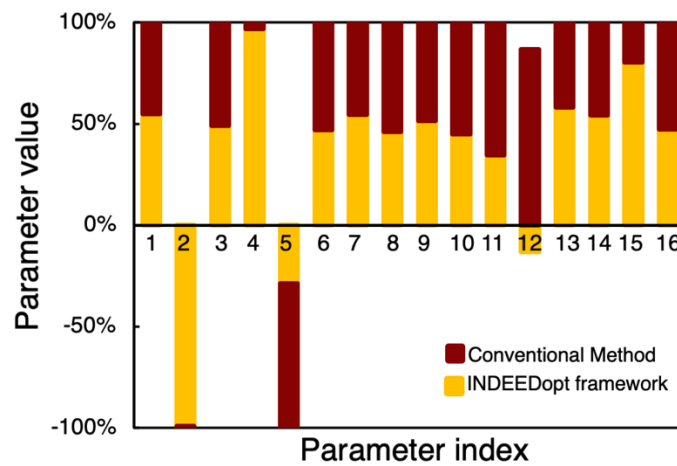


Figure 5. The stacked bar graph representation of the difference between the parameter values in the nickel-chromium training set optimized using INDEEDopt and conventional method. The red represents the conventional, and yellow represents INDEEDopt version of the same parameter. The difference between the percentage values corresponds to the difference between predicted values by each method. The value 50% corresponds to a situation when both methods predict the same values. If the predicted parameter values are negative, then the percentage value is given as negative.

W-S-C-H force field optimization

The parametrization of the W-S-C-H quaternary system is more complex than the binary alloy case because the FF-training set involves both molecular and condensed phase parts. The presence of a condensed phase brings extra challenges to the optimization problem. One of the challenges is that the molecular and condensed phase may be best optimized separately in different local minima in the parameter space, meaning that a well-optimized molecular part does not necessarily produce low discrepancy for condensed phase part as well. In addition, the condensed phase part requires the usage of large molecular configurations to represent a crystalline phase. For example, the W-S-C-H training set has several geometries that are composed of hundreds of atoms, which makes these properties more sensitive to alteration in parameter values, and therefore brings considerable uncertainty to the parameter-property correlations. An additional complexity to the optimization problem comes from the size of the FF-training set. There are 289 reference properties in FF-training set, which is significantly higher than most used in literature to test newly developed optimization procedures. The FF-training set includes heat of formation values for both molecular and condensed phase, defect and ad-atom energetics, some application-specific reaction energetics, strain energetics, and bond and angle energies. The total number of parameters to be optimized is 68, which include bond, angle, and off-diagonal terms. A detailed list of optimized parameters is given in Table S2.

INDEEDopt produced a total of 43700 samples until the DL model is trained to mimic ReaxFF-MD with reasonable accuracy. The total discrepancy sum for different parts in the FF-training set is given in Figure 6 and compared with the ones calculated using the conventional method. A more detailed comparison is also given in Figure S2. The difference between the final optimized parameter values obtained by both methods can be seen in Figure 7. As can be seen from the bar

graphs, INDEEDopt produced a lower total error than the conventional method for angle scan, bond scan, and defect/ad atom parts of the force field. The lowest total error sum obtained using the conventional method was calculated as 5314.4381, which was reduced to as low as 5250.3082 using INDEEDopt. The amount of time spent on optimization was approximately a month with the conventional method and was five days with INDEEDopt. We should note that the optimization criterion in this study was to do better than the conventional method; therefore, the optimization was stopped after the discrepancy value was below the one produced by the conventional method. However, the parameter set was very well developed by experienced FF developers using the conventional method; therefore, it was a challenging test. INDEEDopt spent approximately one day, around thirty million calculations, at the optimization stage to predict a lower discrepancy point than the conventionally obtained one. Therefore, it may not be guaranteed to predict a better point by running the optimization longer because: (1) INDEEDopt predicted a local minimum that is very different from the conventionally obtained one, which can be seen in Figure 7, and there may not be a better point accessible by our optimization method in this specific low discrepancy region; (2) DL model may not be able to make predictions as sensitive as ReaxFF-MD, which prevents revealing a better point. It may be possible to further improve the parametrization by assigning INDEEDopt predicted parameters as a starting point to the conventional method by assigning a narrow parameter search range.

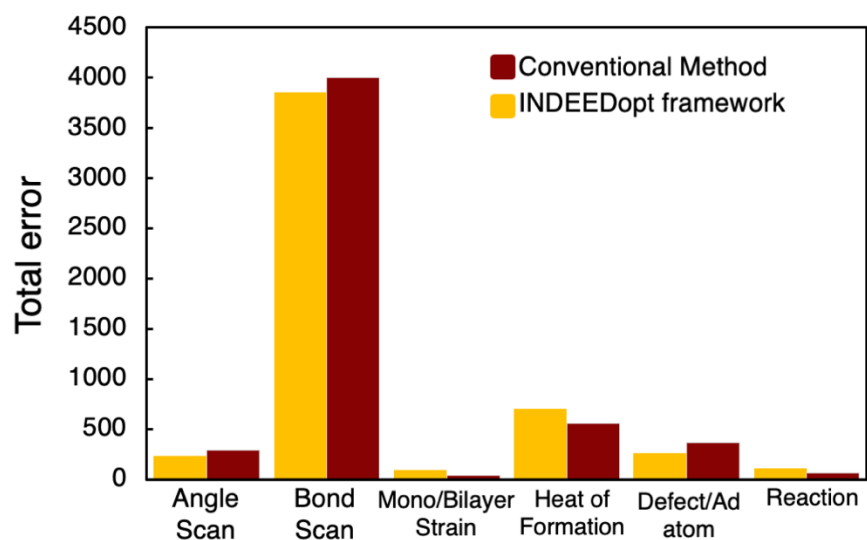


Figure 6. The total error values corresponding to different parts of the tungsten-sulfide-carbon-hydrogen FF-training set calculated by INDEEDopt and conventional methods.

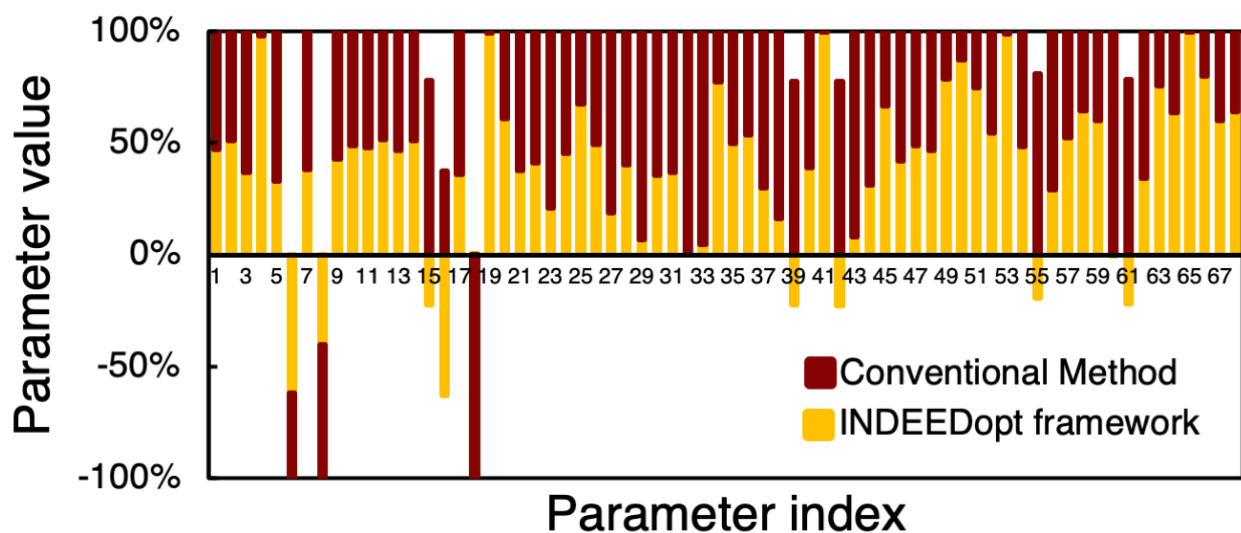


Figure 7. The stacked bar graph representation of the difference between the parameter values in the tungsten-sulfide-carbon-hydrogen training set optimized using INDEEDopt and conventional method. The red represents the conventional, and yellow represents INDEEDopt version of the same parameter. The difference between the percentage values corresponds to the difference between predicted values by each method. The value 50% corresponds to a situation when both

methods predict the same values. If the predicted parameter values are negative, then the percentage value is given as negative.

CONCLUSION

An initial design enhanced deep learning-based optimization framework has been developed to parametrize ReaxFF force fields. The INDEEDopt framework was tested on two different training sets, and the performance was examined comparing with the conventional method. The tests were conducted on Ni-Cr and W-S-C-H training sets, of which lateral is the most challenging due to a large number of properties and diversity in the training set. The framework could produce a lower total discrepancy between ReaxFF values and reference property values compared to the conventional method. The time required for optimization calculations is also lower using INDEEDopt due to its ability to run on multiprocessors. Another advantage of INDEEDopt framework is its ability to be used as an initial parameter generator that can be combined with other optimization methods, which can be valuable in avoiding local minima sticking problem in some other optimization methods and also to fine-tune force fields. Additionally, INDEEDopt can be used by any software that can run ReaxFF-MD energy minimization simulations, which makes it open to a wider user population.

This version of INDEEDopt is fully capable of optimizing ReaxFF force fields with sufficient quality in short periods of time when appropriately used. However, there are some challenges that can be addressed to improve the framework further in future versions. The sampling scheme (first stage of the framework) used in INDEEDopt is computationally the most demanding part due to the high dimensionality of the problem, which can be addressed by developing problem-specific

initial design methods. With the implementation of design of experiments-based optimization procedure for neural network hyperparameter tuning, desired accuracies from the DL model can be achieved in shorter periods of time. The brute-force optimization used in this version of the code is not efficient and the most appropriate method to be used in high dimensional problems; therefore, a problem specific optimization method can be developed to further increase the quality and speed of the framework. The future versions of INDEEDopt will also include subroutines that conduct statistical analysis to give feedback about the FF-training set, which will be very valuable to further improve FF quality.

Supporting Information. The following files are included with this manuscript for free of charge.

Supporting information regarding the details of analysis to supplement the work (PDF).

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