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Method and advantages of genetic algorithms in parameterization of interatomic potentials: Metal oxides

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ABSTRACT

The method and the advantages of an evolutionary computing based approach using a steady state genetic algorithm (GA) for the parameterization of interatomic potentials for metal oxides within the shell model framework are developed and described. We show that the GA based methodology for the parameterization of interatomic force field functions is capable of (a) simultaneous optimization of the multiple phases or properties of a material in a single run, (b) facilitates the incremental re-optimization of the whole system as more data is made available for either additional phases or material properties not included in previous runs, and (c) successful global optimization in the presence of multiple local minima in the parameter space. As an example, we apply the method towards simultaneous optimization of four distinct crystalline phases of Barium Titanate ($BaTiO_3$ or BTO) using an ab initio density functional theory (DFT) based reference dataset. We find that the optimized force field function is capable of the prediction of the two phases not used in the optimization procedure, and that many derived physical properties such as the equilibrium lattice constants, unit cell volume, elastic properties, coefficient of thermal expansion, and average electronic polarization are in good agreement with the experimental results available from the literature.

1. Introduction

The temperature and pressure dependent atomistic molecular dynamics (MD) simulations of material systems lie at the core of many advances in the discovery and optimization of novel materials in a wide variety of applications areas. The accuracy of the material properties resulting from MD simulations, however, is always directly correlated to the quality of the interatomic interaction potentials or force field functions underlying the dynamics. The potential function is comprised of a functional form and an accompanying set of numerical coefficients or parameters fitted to optimize the physical properties of the simulated materials. The functional form is generally an ansatz chosen so as to reproduce known symmetries, crystal structure, and basic physical or chemical properties of the material. An initial dataset comprising of the crystal structures including the lattices constants, bond lengths and angles, and systematic changes in the energies of the system as these values are changed (in other words elastic constants) are obtained either through higher accuracy ab initio quantum mechanical simulations or through experimental measurements. Given such a reference dataset and a chosen ansatz for the functional form, the potential parameters are generally obtained by finding a set that gives the best fit to a defined set of properties or material features [1]. Optimizing the parameters for a given set of the properties is often a combination of local curve fitting to the selected material's characteristic trends followed by closed-loop MD or static structural simulations in which known macro-scale material quantities are targeted by iterative manual adjustment of the chosen parameter sets [2].

The above process by definition is sequential in nature and needs to be wholly repeated whenever a reference dataset is augmented with new materials' structural or properties data. As a result, in the earlier days, for well known semiconductors like silicon more than 30 interatomic potentials or force field functions were developed [3] but mainly the Stillinger–Weber or Tersoff potentials for silicon or their derivatives have been used extensively over the last two decades [4–6]. Similarly, for reactive hydrocarbons the original Brenner potential [7] was developed in the early 90s and has been used extensively over the last two decades [8], but only minor changes or improvements have been added since then [9].

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The conventional local gradient based curve fitting methods, such as steepest descent and conjugate gradient, used in the traditional optimization loop: (i) are inherently local in nature and strongly dependent on the initial configurations for the fitting, (ii) have a tendency to get trapped in or find mainly the nearest local minima in a many dimensional fitting parameter space, (iii) cannot include incremental additions to the reference dataset or the physical properties without sacrificing completely the previous optimization and starting all over again, and (iv) are limited overall by how much data or how many physical properties can be accommodated for a given set of parameters in the optimization procedure. In certain cases when these methods are used, the limitations of a resulting parameterization are a product of the functional form of the potential itself, but many times these limitations result from the sequential nature and local dependency of the optimization loop used by local gradient based techniques. For example, in many cases, the commonly used functional forms have been reported with one set of parameterization for the bulk properties and another set of the parameterization for the cluster or surface properties of the same material.

Concurrently, in recent years there has also been a focus on the generation and sharing of more and more materials structure and properties data through focused programs such as the Materials Genome Initiative (MGI) [10] and the recent OpenKIM project [11], the latter of which seeks to implement a standard infrastructure for introducing novel potential functions which can make the implementation and testing of new force fields more expedient. These type of focused initiatives, along with the broader capacity of high-performance computing resources available to the materials academic and industrial community, are dramatically increasing the range and depth of material characterization data. Such data includes not only the quantum mechanical ab initio density functional theory based simulations of a wide variety of materials structures and properties, but also the synthesis and atomic scale characterization of the structure and properties of the same materials in experiments. It is natural, therefore, that alternate optimization or fitting procedures be investigated for not only the direct search and discovery of new materials and properties from these large databases, but also for the development of the down-the-stream simulation techniques such as MD or Monte Carlo (MC) methods which are explicitly geared towards exploiting the large amount of available data in the literature and online databases. An interesting example of said alternate optimization schemes is that of the neural network (NN) potential, which is actively being explored for the modeling of a variety of materials [12–14]. In the NN setting, the functional form of the potential function is replaced with a network of interconnected nodes categorized as input, hidden and output nodes [15]. The inputs of the NN are coordinates, or generalized coordinates [15], of the system and the output is the system's total energy. The connection between any two nodes of the network is associated with a specific weight, which is the parameter that is adjusted when fitting the NN to a reference potential energy landscape [16]. This innovative approach to modeling interatomic interactions as well as the technique discussed here, are derived from machine learning principles [17,18], which although applied to other fields of research for quite some time, have only recently begun to make significant contributions to MD techniques.

In this work, we investigate an evolutionary computing (EC) based genetic algorithm (GA) method for the optimization of interatomic potentials for molecular dynamics of materials systems. The optimization occurs for a cost function that measures the fitness of the force field parameter set. The optimization or fitness within the GA context is a measure of the agreement between the target values of a reference database, irrespective of the number of structures and properties in the database, and the values produced from a given initial or fitted parameter set. Roughly speaking, the challenges faced by optimization approaches is that, for even a limited range of parameter variations, the fitness function may live on a very rugged landscape often with many closely-spaced local minima.

The general GA method is derived from biological genetic theory and Darwinian evolutionary principles [19–21]. In the GA method, a randomly generated set of parameters, which constitute the so-called population pool, are adjusted through recombination and mutation operators [22] to improve the parameter sets according to a metric, also known as the fitness. A reference dataset of the atomic configurations and their energies obtained from quantum simulations are stored and the GA subsequently refers to the reference dataset continuously during the fitting procedure. A randomly generated population of parameter sets is initially created, and the GA then begins the stochastic process of recombining and mutating the population pool to iteratively generate better and better evolved parameter sets over subsequent generations which are in agreement with the entire reference dataset.

GA-based evolutionary techniques have been used for a wide range of applications, including the optimization of gas transmission lines in petroleum piping [23], satellite scaffolding design [20], pattern recognition and image analysis [24], microcircuit design [25], and drug design [26] for pharmaceutical applications. In more recent work, Oh et al. [27,28] predicted and optimized the configuration of nanoalloys consisting of Pt-Ag, Pt-Au and Pt-Cu, by using a GA engine to drive an embedded atom model (EAM) energy evaluation to determine the lowest total energy configurations of each bimetallic combination. Within the context of the development and optimization of force field functions, Wang and Kollman [21] were one of the first to use a GA method to create a parameterization for an interatomic potential consisting of bonded and non-bonded force field components for non-reactive bio-molecular systems. The implementation presented here is derived directly from Globus et al. [29,30], where GA was employed to reproduce the Stillinger–Weber potential parameters for silicon [4] as an initial test case.

In this work we develop and extend the method to the optimization of parameters for interatomic potentials for metal oxides, which are known to exist in multiple phases at different temperatures. As an example, we apply the GA method to the parameterization of Barium Titanate ($BaTiO_3$ or BTO), for which a reference dataset is first created using a DFT method, and then we explicitly show that it is possible to (i) optimize the multiple phases of BTO in a simultaneous single run, (ii) include incrementally more and more data in the reference dataset during the optimization loop, and (iii) include both the near-equilibrium and far-from-equilibrium configurations in the optimization. All four known phases of BTO, rhombohedral, orthorhombic, tetragonal and cubic, are simultaneously fitted within a single GA parameterization run, and the results are found to be in good agreement with the entire DFT reference dataset, as well as with the available experimental measurements of the basic mechanical and thermal properties of BTO reported in the literature.

In Section 2, we describe the details of the GA method as used for the parameterization of interatomic force field functions with an example application for the optimization of the shell model potential [31] for BTO described in Section 3. In Section 4, the main results of the GA fitting to multiple phases of BTO in a single simultaneous run are reported, and the validation of the evolved parameterization in comparison with experimentally reported values of the basic structural, mechanical, and thermal properties of BTO are described. Finally in Section 5, we summarize the main advantages and suitability of the GA method in fitting parameters of force field functions as larger and larger reference datasets for different materials are developed and made available online.