10º SIMPÓSIO DE ESTRUTURA ELETRÔNICA & DINÂMICA MOLECULAR
SEEDMOL 4 a 8 novembro | 2024 Pirenópolis, Goiás

Study of Mechanical Properties of DHQ-Graphene with Machine Learning Interatomic Potential (MLIP)

Hugo Xavier Rodrigues[1,2], Hudson Rodrigues Armando[1,2], Marcelo Lopes Pereira Júnior[3] and Luiz Antonio Ribeiro Júnior[1,2]

[1]Institute of Physics, University of Brasília, 70910-900, Brasília, Brazil.
[2]Computational Materials Laboratory, LCCMat, Institute of Physics, University of Brasília, 70910-900, Brasília, Brazil.
[3]University of Brasília, Faculty of Technology, Department of Electrical Enginnering, Brasília, Brazil.

rx.oguh@gmail.com, hrodrigues1729@hotmail.com, marcelo.lopes@unb.br, ribeirojr@unb.br

Keywords: Molecular Dynamics, Mechanical Properties, Machine Learning Potentials.

Abstract

In this study, we investigate the mechanical properties of DHQ-graphene, a theoretical corrosion-resistant carbon allotrope, using a combination of density functional theory (DFT) and classical molecular dynamics (MD) methods. We trained a machine learning interatomic potential (MTP) through ab initio molecular dynamics (AIMD) calculations to accurately model these properties. Our findings reveal that the mechanical characteristics of DHQ-graphene, including Young's modulus and Poisson's ratio, differ slightly from those reported in previous studies by Wang et al., likely due to the more stringent self-consistency convergence criteria applied in our DFT calculations. The stress-strain analysis indicates that DHQ-graphene lies on the threshold between ductility and brittleness, a property that could impact its practical applications. Additionally, our comparative study shows that the AIREBO and AIREBO-M parametric potentials approximate the ab initio and MTP results more closely than the ReaxFF and Tersoff potentials. These findings underscore the utility of MTP in accurately describing the mechanical behavior of novel 2D materials, providing a valuable tool for future research in this field.

Introduction

It has been 20 years since the synthesis of the first 2D carbon material called graphene, which initiated the search for new 2D carbon allotropes due to their electronic, optical, thermal, mechanical, and chemical properties [1]. Graphene, for instance, is a stable organic semimetal material that is optically transparent, with high thermal conductivity, a Young's modulus of approximately E = 1.0 ± 0.1 TPa, and a tensile strength of 130 GPa before breaking [2].

To study the mechanical properties of these materials, various methods, both theoretical and experimental, are employed depending on the scale of the structure. For 2D nanomaterials like graphene, small dimensions can reduce accuracy and negatively affect the reliability of experimental measurements of mechanical properties [3]. Therefore, accurate theoretical methods are necessary to investigate these properties.

DFT methods are known for their high accuracy, reproducibility, and flexibility, but they require significant computational

resources and cannot study systems with more than 1000 atoms, often neglecting temperature effects in ab initio calculations of mechanical properties [3. 4]. On the other hand, classical methods use interatomic potentials to describe interactions and can be categorized into two groups: parametric and non-parametric potentials [5]. The first group, which includes empirical potentials, can study the mechanical properties of materials at elevated temperatures with millions of atoms but often lacks accuracy and can overestimate tensile strength values, predicting nonphysical stress-strain curves [3,5]. Non-parametric potentials, however, can achieve greater accuracy and be improved through quantum mechanics calculations.

One type of non-parametric potential is the machine learning interatomic potentials introduced by Behler and Parrinello, which are trained using DFT-based datasets [6]. In 2016, Shapeev proposed a new approach for these non-parametric interatomic potentials, called Moment Tensor Potentials (MTP), which use machine learning methods to improve accuracy. [5]

As demonstrated by Mortazavi et al., these potentials can be used to study mechanical properties with classical molecular dynamics, achieving accuracy comparable to DFT calculations [3].

In this work, to study the mechanical properties of a theoretical corrosion-resistant carbon allotrope called DHQ-graphene, which cannot be described using empirical potentials but is similar to graphene, we trained an MTP using the MLIP package and DFT calculations with VASP. We then compared some of our results with those achieved by Wang et al [7].

Methodology

For this study, ab initio molecular dynamics (AIMD) using the Vienna Ab initio Simulation Package (VASP) were used to create a dataset for training the MLIP potentials.

In this work, the generalized gradient approximation (GGA), the Perdew-Burke-Ernzerhof (PBE) exchange-correlation functional [8], and the projector augmented-wave (PAW) method [9] were employed to analyze the mechanical properties of DHQ-Graphene, including elastic constants evaluation and classical stress-strain study. A cutoff energy of 500 eV was used, with a self-consistency convergence criterion of 10^{-7}eV, adopting a 3x4x1 Monkhorst-Pack K-point mesh in AIMDs and a 6x7x1 Monkhorst-Pack K-point mesh for the elastic constant calculations.

The training dataset was prepared through AIMD calculations performed on an 80-atom structure where DHQ-Graphene was stressed and stress-free, with the first being conducted at a fixed temperature and the second with varying temperatures.

With the dataset prepared, we utilized the LAMMPS package to examine the mechanical properties using the trained MTP, with a time step of 0.1 fs, comparing the results with those achieved by Wang et al., the achieved with parametric potentials and with the elastic constants calculations carried out in VASP with seven different applied strains (-0.015, -0.010, -0.005, 0.000, 0.005, 0.010, 0.015).

Results

In this study, we investigated the mechanical properties of DHQ-graphene using DFT calculations and classical dynamics, employing a machine learning interatomic potential trained through AIMD calculations.

The study of DHQ's mechanical properties reveals a small difference between the Young's moduli and Poisson's ratio obtained by Wang et al. and the data calculated using the elastic constants evaluated with VASP, which are as follows:

C11 = 230.2 GPa·nm, C12 = 60.88GPa·nm, C22 = 299.63 GPa·nm, and C44 = 82.43 GPa·nm. In Wang et al.'s work, they found a Young's modulus of 269 GPa·nm with a Poisson's ratio of 0.28 along the a-axis. In contrast, with VASP calculations, we found values of 217.89 GPa·nm and 0.203.

These values, obtained through VASP calculations, were found using a better self-consistency convergence criterion than that used in Wang et al.'s article, and are closer to those achieved through classical dynamics using the MTP with LAMMPS.

Furthermore, the stress-strain curve analysis and elastic constants results show that this material is on the threshold between a ductile and brittle material.

The comparative study of the mechanical properties of DHQ with MTP and the parametric potentials revealed that the AIREBO and AIREBO-M parametric potentials were those that reached values closer to ab initio calculations, and hence also close to those obtained with MTP, differing from the ReaxFF and Tersoff potentials.

Conclusions

This study highlights the effectiveness of using machine learning interatomic potentials (MTP) trained through AIMD calculations to investigate the mechanical properties of DHQ-graphene, a theoretical corrosion-resistant carbon allotrope. The comparison of our results with those obtained by Wang et al. demonstrates the importance of employing accurate self-consistency convergence criteria in DFT calculations, as evidenced by the closer alignment of our Young's modulus and Poisson's ratio values to those derived from ab initio methods.

Moreover, our findings indicate that DHQ-graphene exhibits mechanical properties that place it on the threshold between ductile and brittle materials, a characteristic that could have significant implications for its potential applications. The comparative analysis of different parametric potentials revealed that the AIREBO and AIREBO-M potentials closely replicate the mechanical properties predicted by both DFT and MTP calculations, whereas the ReaxFF and Tersoff potentials show greater deviations.

These results underscore the potential of MTP in providing a more accurate description of the mechanical behavior of novel 2D materials, particularly when empirical potentials fall short. Future studies could further explore the application of MTP to other carbon allotropes and investigate the temperature-dependent mechanical properties to fully realize the potential of DHQ-graphene in practical applications.

References

[1] K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, Y. Zhang, S. V. Dubonos, I. V. Grigorieva, and A. A. Firsov. Electric field effect in atomically thin carbon films. Science, 306(5696):666–669, 2004.

[2] D.G. Papageorgiou, I.A. Kinloch, R.J. Young, Mechanical properties of graphene and graphene-based nanocomposite. Progress in Materials Science, 90:75–127, 2017.

[3] B. Mortazavi, X. Zhuang, T. Rabczuk, A. V. Shapeev. Atomistic modeling of the mechanical properties: the rise of machine learning interatomic potentials. Mater. Horiz., 10:1956–1968, 2023.

[4] Pan, J. Scaling up system size in materials simulation. Nature Computational Science, 1(2):95–95, Feb 2021.

[5] A. V. Shapeev. Moment tensor potentials: A class of systematically improvable interatomic potentials. Multiscale Modeling & Simulation, 14(3):1153–1173, 2016.

[6] Behler, J. & Parrinello, M. Generalized neural-network representation of high-

dimensional potential-energy surfaces. Phys. Rev. Lett., 98:146401, Apr 2007.

[7] Wang X, Chen L, Yuan Z, Rong J, Feng J, Muzammil I, Yu X, Zhang Y and Zhan Z. Dhq-graphene: a novel two-dimensional defective graphene for corrosion-resistant coating. J. Mater. Chem. A, 7:8967–8974, 2019.

[8] Salpeter, E. E.; Bethe, H. A.; A relativistic equation for bound-state problems. Physical Review, 1951, 84, 1232–1242.

[9] Kresse, G.; Daniel, J. From ultrasoft pseudopotentials to the projector augmented-wave method. Phys. Rev. B 1999, 59, 1758.