# MECHANICAL PROPERTIES OF BORON NITRIDE SHEETS USING ML POTENTIAL

**M.Tech.** Thesis

By MAHESH



## DISCIPLINE OF MECHANICAL ENGINEERING INDIAN INSTITUTE OF TECHNOLOGY

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# MECHANICAL PROPERTIES OF BORON NITRIDE SHEETS USING ML POTENTIAL

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Submitted in partial fulfillment of the requirements for the award of the degree

of

Master of Technology

by

MAHESH



## DISCIPLINE OF MECHANICAL ENGINEERING INDIAN INSTITUTE OF TECHNOLOGY

## INDORE

MAY 2024



## INDIAN INSTITUTE OF TECHNOLOGY INDORE

## **CANDIDATE'S DECLARATION**

I hereby certify that the work which is being presented in the thesis entitled **MECHANICAL PROPERTIES OF BORON NITRIDE SHEETS USING ML POTENTIAL** in the partial fulfillment of the requirements for the award of the degree of **MASTER OF TECHNOLOGY** and submitted in the **DISCIPLINE OF MECHANICAL ENGINEERING, Indian Institute of Technology Indore**, is an authentic record of my own work carried out during the time period from July 2022 to May 2024 under the supervision of **Dr. Shailesh I Kundalwal**, Associate Professor, Department of Mechanical Engineering, Indian Institute of Technology Indore.

The matter presented in this thesis has not been submitted by me for the award of any other degree of this or any other institute.  $V(herh_{03}) = 06$ 

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This is to certify that the above statement made by the candidate is correct to the best of my/our knowledge.

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## **Dedicated** To

My Beloved Family

## Abstract

This study explores the introduction of nanostructures, and the areas where they are playing an important role. We investigated the mechanical properties of boron nitride nanosheets while addressing challenges in analysis methods for predicting the accurate results of structural properties. approaches were discussed, emphasizing how they operate, their benefits, limitations, and the necessity for a more effective and efficient solution. Introducing the novel MLIP method with the use of machine learning technique, we thoroughly discussed the stepby-step creation process of MLIP using precise but limited data and the parameters that should be taken care for the construction of an effective potential. Furthermore, we addressed techniques for evaluating its performance and robustness in different environments. With the implementation of the newly formed MLIP in classical molecular dynamics simulation, we outlined how simulation software operates and functions and investigated the mechanical attributes of boron nitride nanosheets, exploring the fluctuations of sheet strength across a range of dimensions, temperatures, and added layers. We obtained a Young's modulus of 993 GPa at 1K, whereas an average breaking stress and breaking strain are around 106 GPa and 0.16, respectively. Our outcome exhibits significant enhancements compared to prior studies, highlighting MLIP's effectiveness in advancing studies with greater accuracy and minimal effort. This study offers comprehensive analysis and theoretical exploration, delivering valuable insights into MLIP and the mechanical properties of boron nitride nanosheets, paving the way for future applications in materials science and engineering.

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## ACRONYMS

AENET	Atomic energy network
AIMD	Ab initio molecular dynamics
ANN	Artificial neural network
BN	Boron Nitride
CFG	Configuration
CMD	Classical molecular dynamics
CVD	Chemical vapor deposition
D	Dimension
DDS	Drug delivery systems
DFT	Density functional theory
EMP	Empirical interatomic potentials
FCC	Face centered cube
GAP	Gaussian approximation potential
hBN	Hexagonal boron nitride
LED	Light Emitting Diode
MAE	mean absolute error

## LIST OF SYMBOLS

- eV Electron Volt
- keV Kilo-electron Volt
- MeV Mega-Electron Volt
- GeV Giga-Electron Volt
- g/cm<sup>3</sup> Gram per cubic meter
- dpa/s Displacements per Second
- MPa Mega-Pascal
- GPa Giga-Pascal
- K Kelvin
- E Energy

## **Chapter 1 : Introduction and Literature Review**

#### **1.1 Introduction to nanostructures and their applications**

Nanomaterials are typically described as substances consisting of components wherein at least one dimension measures below 100 nm. This defining characteristic gives rise to novel physical and chemical properties inherent to the nanoscale. Compared to bulk materials, nanoscale observation reveals variations in the electrical, optical, chemical, thermal, and magnetic characteristics of the same material as shown in Figure 1. The two main reasons why nanomaterials exhibit unique properties compared to bulk materials are:

1. High surface to volume ratio 2. Quantum confinement effects

The considerable elevation in the ratio of surface atoms to interior atoms in nanomaterials enables them to engage more effectively with the environment than bulk materials. Despite this, it's worth noting that as size decreases, the surface-to-volume ratio increases, leading to a proportional effect on properties including transition temperature and solubility. Quantum size effects are intricately linked to the concept of "dimensionality" and confinement, which entails constraining the movement of electrons that would otherwise move randomly to discrete energy levels. As particles are reduced to the nanoscale, the confining dimensions cause energy levels to become discrete, consequently increasing the bandgap of the material. The increase in the bandgap between the energy levels leads to a spectrum of discreet energies reflecting various fluorescent colors [1].



Figure 1: Examining atom arrangement in graphite lead at nanoscale

Reducing particle size to the nanoscale alters properties like melting point, fluorescence, electrical conductivity, magnetic permeability, and chemical reactivity, which change with particle size. Yet, these properties are not exclusively size-dependent.

Nanomaterials became an important materials in various fields due to their unique properties such as grain size on the order of 1–100 nm. Their contribution to environmental sustainability and medicine are incredible. Here are some of the major fields where nanomaterials play important roles.

- 1 *Energy conversion and storage applications* Nanomaterials not only provides an ideal platform to disclose the structure–property relationships, but also leads to dramatically enhanced performances in supercapacitors, rechargeable batteries, and catalysis. They provide numerous fascinating properties, such as abundant active surfaces and open ion diffusion channels, which enable fast transport and storage of lithium ions and beyond [2].
- 2 *Medicine* In controlled drug delivery systems (DDS) offer a sophisticated approach where the drug is precisely transported to its intended site of action, thereby minimizing its impact on vital

tissues, and reducing undesirable side effects. Moreover, DDS serves to shield the drug from rapid degradation or clearance, while concurrently amplifying its concentration within target tissues. Consequently, this method facilitates the achievement of therapeutic outcomes with lower doses of the drug[3].

- 3 Surface coating - Coatings stand as the primary choice for shielding metals against corrosion. Despite this, the susceptibility of polymer coatings to the penetration of corrosive solutions at the metal/coating interface remains a notable concern. However, the integration of nanoparticles into coatings has emerged as a promising solution to enhance their chemical, mechanical, and optical properties. Nanocoating has proven to be a highly effective method for mitigating the impact of corrosive environments, owing to its array of advantages. These include enhanced surface hardness, superior adhesive qualities, long-term durability, as well high-temperature corrosion resistance. Furthermore, as nanocoating serves to improve tribological properties, among other benefits [4].
- 4 *Electronics* By enhancing optoelectronic performance, broadening integration possibilities, and reducing costs, nanomaterials have facilitated significant progress in the electronic field. A delicate equilibrium between conductivity and stretchability is achieved through high aspect ratio, coupled with electronic and optical properties. This balance is essential in designing e-skin materials, including strain sensors, solar cells, stretchable LEDs, biological monitoring devices, and transparent conductors, among other applications [5].
- 5 *Lubricants* The combination of robust intra-layer atomic bonding and low interlayer shear strength makes nanomaterials highly promising for lubricating. The two layers in multilayer nanosheets

were bonded by Van der Walls which is weak in nature compared to covalent bond. This weak bonding result in relatively low shear strength, facilitating easy sliding between adjacent layers when subjected to shear force.

6 *Nanofiltration* - Nanofiltration, utilizing membrane pores sized between 0.5 to 2.0 nm and operating pressures situated between those of reverse osmosis and ultrafiltration, harnesses the strengths of both methods: high-solute rejection and low-energy consumption. As nanofiltration technologies rapidly evolve, the market for their applications is experiencing significant growth.

### **1.2 Classification of nanomaterials**

Based on the dimensionality nanomaterials are classified into 4 categories which are shown in Figure 2 [6].



Figure 2: Classification of nanomaterials on the basis of dimensional characteristics

- 1. *Zero-dimensional nanomaterials (0-D)*: these materials have all three dimensions inside the nanoscale range. Nanoparticles, fullerenes, and quantum dots are a few examples.
- 2. One-dimensional nanomaterials (1-D): materials falling under this category only have one dimension that is not part of the

nanoscale. Nanotubes, nanofibers, nanorods, nanowires, and nanohorns are a few examples.

- Two-dimensional (2-D) nanomaterials: this class of nanomaterials has two dimensions that extend beyond the nanoscale. Nanofilms, nanosheets, and nanolayers are a few examples. The first 2D substance to be found was graphene.
- Three-dimensional (3-D) or bulk nanomaterials: materials in this class are not limited to any one dimension of the nanoscale. Bulk powders, nanoparticle dispersions, arrays of nanowires and nanotubes, etc. are all included in this class.

### **1.3 Boron nitride nanosheets and structural properties**

Boron nitride has emerged as a popular material after carbon and its allotropes. From nano electronics to the space industry, it has shown its potential in numerous areas. Structurally similar to carbon, BN forms a hexagonal lattice with alternating boron and nitrogen atoms, and can be found in forms such as nanosheets, nanoribbons, fullerenes, and nanotubes. As shown in Figure 3 [7].



Figure 3: Categorizing hBN based on the arrangement of atoms.

Hexagonal boron nitride (hBN) nanosheets are most popular at current period. Due to its similar structural arrangement to graphene, h-BN is also called white graphene. In contrast to the purely covalent C–C bonds found in graphene, the bonds present in hBN sheets exhibit unique properties, prompting numerous researchers to delve deeper into its immense potential for diverse applications [2]. The bond length of B-N in sp2 hybridized hBN is 147 pm, with a 334 pm separation between consecutive hexagonal planes held together by van der Walls interactions. An h-BN sheet consists of dissimilar adjacent sides, with one following a zigzag pattern and the other following an armchair pattern, as shown in the Figure 4 below.



Figure 4: hBN nanosheet highlighting armchair and zigzag orientation.

## 1.4 Methods to synthesis hexagonal boron nitride

Various approaches have been developed to produce mono- and multilayer hexagonal boron nitride (hBN) with a focus on achieving high yield and quality, along with significant lateral size. Notably, some popular techniques which have been extensively investigated are.

- 1) Mechanical exfoliation
- 2) Liquid exfoliation
- 3) Chemical vapor deposition (CVD)

Each synthesis method offers distinct advantages and disadvantages. Mechanical exfoliation presents a feasible approach for producing crystalline hBN structures, provided it is executed properly. However, the resulting hBN layers are often randomly distributed with limited flake size and low yield. In contrast, liquid exfoliation proves to be an economical method for generating a significant amount of hBN nanosheets Nevertheless, the inconsistent layer quantity, small flake sizes, and potential risk of surface contamination from the chemicals involved may present challenges for large-scale practical production.

In contrast, chemical vapor deposition (CVD) and vapor phase epitaxy processes offer advantage of managing the layer count while facilitating the extensive fabrication of atomically thin hBN nanosheets as shown in Figure 5. Despite this, the demanding synthesis conditions, including high reaction temperatures and exposure to different gas atmospheres, often give rise to defects within the crystal structure. As a consequence, significant variations occur in the material properties of 2D-hBN, ultimately affecting device performance [8].



Figure 5: Diagrammatic representation of the CVD setup utilized for h-BN synthesis

## **1.5 Structural properties of hBN**

hBN has extraordinary bulk properties, including high mechanical rigidity, high thermal and chemical stability, a low dielectric constant

(electrical insulation), and a very low coefficient of friction offered abundant opportunities to investigate various diverse applications. hBN demonstrates considerable promise across various industries, scientific disciplines, and technological fields, particularly in nano-electronics, optoelectronics, field emission, and lubrication under extreme temperature conditions. [9].

BN nanomaterials possess distinct advantages over their carbon counterparts, primarily due to their electrical insulating properties and superior stability at high temperatures and in diverse chemical environments. Despite being electrically insulating, hBN can be effectively tuned to exhibit varying properties and functionalities through several strategies, including doping, substitution, functionalization, and hybridization.

Moreover, the sleek surface of h-BN positions it as an exceptional contender for tribological applications. With a substantial band gap reaching 5.9 eV and seamless lattice parameter alignment with graphene, h-BN solidifies its status as an optimal choice for serving as an epitaxial substrate and gate layer in graphene-based devices [10].

Property	Value		
Appearance	White powder, photostable, odorless		
Appearance	(hexagonal, cosmetic grade)		
Bond length	1.466 Å		
Molar mass	24.82 g mol⁻¹		
Density	~2.1 g/cm <sup>3</sup>		
Structure	Crystal; hexagonal		
Melting point	2.973°C; sublimes		
Surface area	0.82–30 m²/g (varies by grade)		
Refractive index (nD)	1.74		

Table 1: Structural properties of hexagonal boron nitride

Stability	Chemical inert and stable		
Hardness	1–2 on the Mohs scale		
Specific heat capacity	19.7 l/(K·mol)		
(C)			
Std enthalpy of	-254.4 kl/mol		
formation ( $\Delta_{\rm f} {\rm H^o}_{298}$ )			
Gibbs free energy ( $\Delta_f G^\circ$ )	–22 kJ/mol		
Coefficient of friction	<0.3		

## 1.6 Literature review and research gap

Several studies have been conducted on BNS due to their exceptional bulk properties, including a high Young's modulus of 800–850 GPa [11], a thermal conductivity of 300–2000 W/m–K [12], an insulating property with a 5–6 eV band gap [13] and a low coefficient of friction [14]. These features offer multiple opportunities for exploring various applications such as manufacturing sensors and actuators [15, 16], increasing thermal conductivity [17], improving their thermal energy storage performance , efficient thermal management for transformers [18], increasing durability of fuel cells [19]. However, accuracy is essential to guarantee performance, safety, and dependability in crucial fields like space industries like radiation shielding [20], and medical applications, including medication administration. Deviations, no matter how little, might have fatal effects. Therefore, it is crucial to accurately assess the structural qualities.

Many authors have investigated the mechanical characteristics of hBN using various methods. Like, author Han et al. performed the uniaxial tension using, molecular dynamics (MD) simulation and obtained the Young's modulus of 881 GPa [21]. Whereas Zhao and Xue obtained Young's modulus of 716.3 GPa [22]. Li et al. performed the MD simulation and observed the Young's modulus of the pristine hBN sheet as 666 GPa and 642 GPa in zig zag and armchair direction respectively [23]. Similarly, numerous research has been carried out utilizing first

principle method like author Ohba et al. performed first-principles calculation based on density functional theory and obtained Young's modulus of 951.5 GPa [24].

As we examine the above outcomes, we can see that there is so much dispersion in the results obtained by each method for the same material. The disparity in property predictions makes it more challenging to use BN structures across various industries. Thus, it is essential to undertake a thorough examination of structural properties before their utilization.

## Chapter 2 : Current techniques to study the nanostructures

## 2.1 Different techniques currently available

Currently, there are numerous techniques for studying materials at various scales, ranging from the continuum scale to the electronic scale. Each method has its own advantages and limitations, making the selection of the appropriate scale a challenging task that requires balancing computational cost and accuracy, as depicted in the Figure 6. At the continuum scale, properties are assumed to be continuous, which simplifies computations but often results in less accurate outcomes. Conversely, studying materials at the electronic scale yields highly accurate results, but this precision comes at the cost of significant time and financial investment.



Figure 6: Comparing time and length scales across simulation methods: balancing accuracy and computational efficiency

For investigating nanostructures and determining their properties, two most popular techniques that were preferred by researchers are ab initio molecular dynamics (AIMD) simulations based on density functional theory (DFT) [25] and the classical molecular dynamics (CMD) simulations which use predefined empirical interatomic potentials (EIP) for the simulations. DFT based AIMD simulation known for its accurate force and energy prediction, whereas CMD prioritizes flexibility and efficiency [26].

### 2.2 Comparison of AIMD and DFT methods

AIMD simulations based on DFT perform quantum mechanics calculations, which offer an elaborate understanding of materials electronic structures, facilitating the assessment of energy interactions among atoms within the defined configuration. As a result, DFT based simulations yield precise outcomes, positioning them as a popular approach for studying nanomaterial properties such as optical, optoelectronics, catalytic, magnetic attributes, electronic, chemical, and mechanical properties with superior accuracy [27–29]. Despite its sufficient accuracy, the DFT method encounters various challenges. These include a maximum number of atoms, typically limited to a few hundred, simulation duration within a few femtoseconds, high computational costs, and extended computational times of several hours [30]. All these limitations collectively render the DFT an inappropriate method for studying massive structures and accurately evaluating their structural attributes [31].

### 2.3 Empirical potential used for CMD simulations

Conversely, CMD serves as a cost effective method to study the structural properties at nanoscales. It is coupled with various predefined EIPs like Tersoff [32], ReaxFF [33], AIREBO [34], REBO [35], and others, which are mathematical expressions designed to predict the energy and forces acting on atoms within any specified atomic arrangement [36]. These EIPs make CMD simulations highly effective, computationally fast, widely embraced, and an indispensable component for extensive atomistic simulations.

Among the above empirical potential Tersof and tersoff like potentials became most successful for determining the boron and nitrogen interactions in nanostructures. In the Tersoff interatomic potential, uses the following expression to compute the energy E, between atoms or i and j.

$$E = \sum_{i} Ei = \frac{1}{2} \sum_{i \neq j} V_{ij}$$

Where

$$V_{ij} = f_C(r_{ij})[f_R(r_{ij}) + b_{ij}f_A(r_{ij})]$$

Here  $f_R$  (r) and  $f_A$  (r) are the repulsive and attractive pair potentials, respectively. The cutoff function  $f_C$  (r) is defined to restrict the potential's range, thereby conserving computational resources in MDS. Typically, the cutoff distance R is selected to encompass only the first-neighbor shell. In addition,  $b_{ij}$  is the bond order function that determines the strength of the attractive term.

However, the EIP based CMD simulations does not account for electric, magnetic, or optical properties, resulting in significantly reduced accuracy [36, 37]. Additionally, all empirical potentials have few adjustable parameters, which make them parametric in nature, limiting their accuracy and enhancement. Furthermore, EIP's accuracy is still dependent on the reliability of the foundational QM model to which they are calibrated, resulting in inaccurate outcomes when applied beyond their original training, showing low degree of robustness [37].

#### 2.4 Motivation for MLIP construction

The constraints associated with the above mentioned methods inspired researchers to bridge the significant gap between accurate but resource intensive DFT based AIMD simulations and effective yet inconsistent EIP based CMD simulations. And it's understood that classical atomic-scale material modeling computations like CMD rely on having the right potential. To address this challenge, Behler and Parrinello [38] introduced machine learned interatomic potential (MLIP) for the first time in 2007, a novel technique that not only offers more precise predictions but also operates with increased efficiency and reliability.

Similar to EIPs, MLIPs are designed to anticipate the potential energy surface relative to atomic positions, which subsequently simplifies force and stress calculations in CMD simulations [36]. Nevertheless, they are formulated using machine-learning approaches on DFT outcomes, resulting in a notable enhancement in simulation accuracy. MLIPs are not constrained by material specificity and employ a non-parametric functional form, facilitating iterative enhancements in accuracy at the cost of computational efficiency. MLIPs function as a bridge, linking the two described widely used methods, blending the accuracy of DFT with the flexibility of EIP-based CMD simulations. MLIPs comes with various benefits, including significantly faster computation, outperforming DFT simulations by several orders of magnitude, and achieving accuracy close to that of DFT, all while maintaining a lower computational cost. Additionally, their flexible nature allows for increased accuracy through training with new configurations where the cost scales as O(N)in the number of atoms in the system – as compared to O(N3)scaling or higher for conventional DFT methods. Today, MLIPs have become essential for nanostructure studies, outpacing DFT in utilization.

The intensified interest in this field can be attributed to the swift accumulation of data from AIMD simulations, encompassing systems containing up to several hundred atoms. This wealth of QM data has brought to light the limitations in accuracy of numerous empirical potentials. The emergence of MLIP have interrupted the decades-long trend of exponential increase of the computational costs associated with enhancing the accuracy of interatomic potentials. A comparison study of how the MLIP with empirical potentials, year published and variation of cost of computation per atom is shown in the Figure 7



Figure 7: Evaluation of the cost-effectiveness per atom timestep across diverse empirical potentials highlighted in (red) and MLIPs in (blue) with respect to the year of publication.

The figure shows that while the computational cost increases with the implementation of new machine learning potentials, the superior accuracy they provide makes this trade-off worthwhile.

### 2.5 Different forms of MLIP and their comparison

In the past decade, there has been a rapid exploration of various strategies, all grouped under the umbrella term MLIPs. They can be further classified into several classes based on the descriptor and training method utilized for the training the potential. Certain classes of MLIP, including moment tensor potential (MTP) [37], spectral neighbor analysis potential (SNAP) [39], gaussian approximation potential (GAP) [40], and artificial neural network (ANN) potentials developed by the atomic energy network (AENET) [41] are among the most widely used. Each classification has its own set of opportunities and challenges that which are discussed below.

- 1 Neural Network Potential (NNP) The groundbreaking MLIAPs gained momentum with Behler and Parrinello's pioneering work on the silicon model. This model employs twobody and three-body rotationally invariant descriptors, commonly referred to as symmetry functions, which provide inputs to a neural network, which is both differentiable and non-linear in nature Through this methodology, the model accurately forecasts the energy associated with every atom
- 2 Gaussian Approximation potentials (GAP) Gaussian process regression is harnessed to predict energy and force pertaining to individual atoms at the local level by utilizing a sparsified training dataset. How closely prediction points align with training points is evaluated employing the similarity metric within the descriptor space. Originally, GAP descriptors were formulated using the SO(4) bispectrum components, but contemporary GAP potentials preferentially employ the spectrum of power originating from basis functions in SO(3) in conjunction with radial basis. These potentials are now integrated into QUIP/libatoms.
- 3 **Spectral Neighbor Analysis Potentials** (**SNAP**) It utilize SO(4) bispectrum components to represent local atomic density, providing input to a linear model for the anticipation of local atomic energy. This approach has been expanded to include quadratic and neural network energy models, as well as descriptors labeled with chemical information. All necessary code to execute SNAP interatomic potentials is integrated into LAMMPS ML-SNAP software package, which contains a command called pair snap, potential covering various materials, and algorithm for generating indispensable training data
- 4 **Moment Tensor Potential (MTP)** -It is similar to SNAP where scalars which are rotationally invariant are used to energy specific to atoms at a localized level. Consequently, individuals can choose based on material and the area of application and proceed with the development of potential. Recently, a comparison study on the

development of different potentials was conducted by Vijay et. al. [36] using different regression methods including atomic energy network (AENET), gaussian approximation potential (GAP), spectral neighbor analysis potential (SNAP) and its quadratic extension (qSNAP), and moment tensor potential (MTP). Upon analysis, it was shown that AENET exhibited the highest potential for accurately forecasting total energies, and MTP demonstrated superior performance in predicting atomic forces.

## 2.6 Applications and Popularity of Machine Learning Interatomic Potentials (MLIPs)

Several studies have been conducted using MLIP including determination of thermal conductivity of MoS2 alloys [42], study of crack propagation in crystalline silicon [43], Li-based disordered rocksalts determining [44] thermal conductivity in graphene/borophene heterostructures [45], study of graphene phonon dispersion, in-plane thermal expansion [46], validation of mechanical properties of carbon through GAP model [47], determining elastic properties of diamond-type silicon with GAP model [40], prediction of the grain boundary energy in FCC elemental metals [48], extensive study of the mechanical failure and thermal transport properties exhibited by different BC2N monolayers [49], determining thermal conductivity of 2D and 3D structures [50] and many more. Correspondingly numerous studies are in progress to maximize their efficiency by applying different machine learning algorithms and calibrating different parameters [51, 52].

The current work demonstrates the construction of a unique interatomic potential using the ML algorithm. Initially, we explore the formulation of the potential utilizing data that was generated via AIMD method, subsequently, validate the potential through various methods. Afterward, we employ the crafted new potential, determine the mechanical properties of BN sheets subjected to uniaxial strain via CMD simulation across various environmental conditions, and compare the obtained results with previous studies performed via alternative methods, meanwhile evaluating the robustness of the potential.

## **Chapter 3 : Methodology of MLIP construction**

#### **3.1 Data generation for training**

The first step in MLIP construction is to generate the training dataset. But prior to it, we need to be specific regarding the material and application area for the intended use of the MLIP, like hBN sheets in our case. One of the most common methods for generating this dataset is to perform AIMD simulations. To develop a powerful potential with high transferability, we carried out AIMD simulation by utilizing vienna ab initio simulation package (VASP) [53] software package with a unit cell of hBN nanosheet consisting of 64 atoms as shown in Figure 8.



Figure 8. Flowchart representing the overview of MLIP generation and its application in determining structural properties.

We employed Perdew-Burke-Ernzerhof (PBE) functionalization of the generalized gradient approximation (GGA) [54], with projector augmented wave (PAW) [55] potentials. The simulations were performed, without preserving any symmetry, for all the structures till total energies and total forces converged within 0.01 meV and 30 meV/Å, respectively. We performed a self-consistent field calculation for the full structure relaxation. AIMD simulations were performed with

 $\Gamma$ -centered k-point meshes with the plane wave kinetic energy cutoff set to 520 eV.

For better performance, it is crucial to highlight diversity while data generation, accurately capturing configurations most relevant to the targeted applications. While considering the constraints of AIMD, we performed several simulations with a runtime of 2 picoseconds, at various temperatures ranging between 0-2500 K and applied uniaxial strain in both planar directions to generate a high-quality dataset for training and validation. Each simulation produces a multipleconfiguration (CFG) file as output, primarily consisting of the force, stress, and coordinates of each atom, as well as the total energy of the system at each timestep as shown in Fig. 2. For optimal training potential, a standard reference database usually comprises anywhere from  $10^3$  to  $10^4$  configurations [36]. In our case, we obtained 68,235 configurations, encompassing simulations spanning different temperatures and strains along multiple directions.

## **3.2 Sorting and subsampling of the training data**

Each result is examined through a screening process to identify flaws that could hinder efficient training, and any faults observed during the simulation are excluded from the original dataset. Before training MLIP, the dataset needs to be subsampled into a more manageable dataset for computational efficiency by selecting the configurations at specific intervals. We took the subsample at every 25th interval from the original dataset, which allowed us to minimize 96% from 68,235 to 2,728 configurations as shown in the Figure 9



Figure 9: Subsampling the large dataset into training and testing sets

The training of interatomic potentials frequently encounters the challenge of overfitting, where the model replicates reference data excessively, thereby diminishing predictive accuracy. To address this issue, a small validation dataset is typically created from the original dataset to evaluate the model's performance independently [36]. Therefore, the final data set consisting of 2728 configurations is then further split into two sets: training data and validation data, with the distribution of one consisting of 70% and the other 30% of the original data, respectively as shown in Figure 10. Training data is primarily utilized to train the model, while validation data is utilized to evaluate the extent to which our model accurately predicts the outcome when exposed to unseen data.



Figure 10: Flowchart depicting the steps involved in training MLIP, highlighting key stages such as data generation, sub-sampling, and validation procedures

The main objective of MLIP is to enhance precision and minimize the computational time and expenses associated with intricate CMD simulations. Factors like quality of the data set, descriptors, regression model, hyperparameters and number of iterations used for training all play vital roles in determining the accuracy and robustness of newly developed MLIP, but descriptors and regression model are the most crucial determinants. Descriptors define the configuration's features; it helps in transforming the local environments of the atoms obtained from the simulations into a mathematical representation. For effective MLIP training, a descriptor must meet specific criteria, including consistent under coordinate system shifts, rotations, and atomic index permutations, while also exhibiting continuity [56]. On the other hand, regression models employ machine learning algorithms for the given descriptor to identify coefficients for the function that can map the linking energy of atoms. Among the various regression methods discussed earlier, we employed MTP linear regression due to its notable balance between accuracy and computational efficiency, surpassing alternative regression classes [57]. The potential developed through MTP regressor evaluates the energy of an atomic arrangement as the combined effect of individual atom contributions within the sphere of a finite cutoff distance.

#### **3.3 Fundamental behind the fitting the energy curve**

In evaluation process, the MTP regressor helps in determining the coefficients  $\theta$ , while addressing the optimization problem given by:

$$\begin{split} \sum_{k=1}^{K} \left[ W_{e}(E^{MTP}(cfg_{k};\theta) - E^{AIMD}(cfg_{k}))^{2} + \\ W_{f}\sum_{i=1}^{N_{k}} \left| f_{i}^{MTP}(cfg_{k};\theta) - f_{i}^{AIMD}(cfg_{k}) \right|^{2} + \\ W_{s}|\sigma^{MTP}(cfg_{k};\theta) - \sigma^{AIMD}(cfg_{k})|^{2} \right] \to \min_{\theta}, \end{split}$$

$$(1)$$

where  $E^{MTP}$ ,  $f_i^{MTP}$  and  $\sigma^{MTP}$  are the energy, forces and stress tensor respectively, obtained from the MTP method, on the other hand  $E^{AIMD}$ ,  $f_i^{AIMD}$  and  $\sigma^{AIMD}$  are the energy , forces, and stress tensor respectively obtained from the AIMD method. Similarly,  $(cfg_k)$  signifies the configuration pertaining to the k<sup>th</sup> instance, and  $\theta$  represents the coefficients of a linear model. The variable  $N_k$  represents atoms in  $k_{th}$ configuration, with  $W_e$ ,  $W_f$ , and  $W_s$  serving as non-negative weights that quantify the relevance of energies, forces, and stresses. These weights must be calibrated based on the material's chemical structure to achieve an effective potential. The linear model with improved coefficients facilitates the creation of a finely tuned potential energy surface, effectively connecting the reference energies provided in the training dataset [36]. Despite the scenario, achieving optimal hyperparameters requires repeating the training process multiple times. In our instance, we performed over 1500 cycles to achieve an effective coefficient. After obtaining the enhanced coefficients of the newly developed linearly modeled potential, it is anticipated to deliver accurate predictions of energy and force for untrained configurations.

### **3.4 Validation of the newly generated potential**

Once the potential is established, the subsequent task is to validate it through diverse validation methods. One effective way to validate the new MLIP is to conduct error checks on the newly created potentials using the same structure used in the training phase and examine the differences between the results predicted by new potential and the actual results obtained from the AIMD simulations. To visualize and evaluate the discrepancy between predicted and actual results, several metrics are used, including the coefficient of determination (R<sup>2</sup>), mean absolute error (MAE) and root mean square error (RMSE) [26]. The equations (2), (3), and (4) depict the formulas for calculating RMSE values for energy, force, and stress, respectively.

$$RMSE (E)^{2} = \frac{1}{K} \sum_{k=1}^{K} \left( \frac{E^{MTP}(cfg_{k};\theta)}{N^{(k)}} - \frac{E^{AIMD}(cfg_{k})}{N^{(k)}} \right)^{2}$$
(2)

$$RMSE (f)^{2} = \frac{1}{K} \sum_{k=1}^{K} \frac{1}{3N^{(k)}} \sum_{i=1}^{N_{k}} \left| f_{i}^{MTP}(cfg_{k};\theta) - f_{i}^{AIMD}(cfg_{k})) \right|^{2}$$
(3)

$$RMSE (\sigma)^{2} = \frac{1}{K} \sum_{k=1}^{K} \frac{1}{9} |\sigma^{MTP}(cfg_{k};\theta) - \sigma^{AIMD}(cfg_{k})|^{2}$$

$$(4)$$

Nonetheless, the new potential must exhibit robustness to be deemed effective. To further validate the robustness of the new MLIP. We conduct an error check on a validation dataset comprising configurations not included in the training process. This evaluation helps verify whether the model is overfitting or performing acceptably. Evaluation of the MAE and RMSE values of both training and validation data enables determination of the MLIPs performance and need for additional training.

## Chapter 4 : Determination of mechanical properties of BN sheets using new MLIP in LAMMPS

After confirming satisfactory performance, the new MLIP can be utilized as a potential file for CMD simulations, leading to enhanced accuracy in results for large scale simulations. In order to obtain precise mechanical property for large hBN structures at different environmental conditions, we used the newly developed MLIP and performed multiple simulations on large–scale atomic/molecular massively parallel simulator (LAMMPS) software package [58].

## 4.1 Data file generation for CMD simulations

Moreover, to analyze the effect of size on mechanical properties, multiple nanosheets were constructed via VMD (visual molecular dynamics software (VMD) package. Multiple sheets having dimensions:  $30\times30$ ,  $50\times50$ ,  $80\times80$ ,  $100\times100$ , and  $150\times150$  Å<sup>2</sup>, all with a uniform thickness of 3.3 Å were modeled. The number of atoms in the sheets were 336, 960, 2432, 3840, and 8400 respectively. A hBN sheet of  $50\times50$  Å<sup>2</sup> is shown in Figure 11 depicting the loading direction.



Figure 11: Graphic depiction of a  $50 \times 50$  Å<sup>2</sup> hBN sheet, highlighting the orientation of loading and the interlayer spacing between adjacent bilayer sheet.

## 4.2 How LAMMPS Work: Step-by-Step process

The LAMMPS program mainly need 3 necessary files: the data file (mainly consisting of geometry of atoms and box dimensions), potential file (calculates interatomic energy) the input file (contains the code for running the simulation) and the output file (contains trajectory of atoms and required data at each timesteps such as pressure, temperature etc.).

The data file can be created independently, as demonstrated in the previous section (4.1) and based on the different atoms and molecules one can directly download the potential file from the interatomic potentials repository site [https://www.ctcms.nist.gov/potentials/].

The input file is like a script written in C++ language, where all the important steps of the simulation need to be mentioned. By following this script, LAMMPS can get detailed results from the simulation. It's crucial for the code within the input file to adhere to a structured sequence, which is primarily segmented into four key stages: initiation, atom definition, setting, and execution. Each of these steps must be meticulously followed in their designated order to ensure the seamless execution of the simulation, as illustrated in Figure 12.



Figure 12: Sequence of Actions in Creating a LAMMPS Input File

*Initiation – Units –* LAMMPS allow user to users to utilize various unit styles including *lj, real, metal, si, cgs, electron, micro and nano*. Each

style defines different units to mass, length, time, energy, pressure, etc. In our case we employed metal units.

*Initiation – Timestep –* In any MD simulations, the timestep plays a key role in result frequency and affects accuracy, simulation time, and trajectory visualization. Selecting the appropriate timestep is difficult due to its trade-offs with accuracy, cost, and simulation time as shown in the Figure 13. Within our study, we implemented a timestep of 0.0005 femtoseconds. Which implies that at every 0.0005 femtoseconds the system gets updated and produce the required output.



Figure 13: Demonstrating the reciprocal correlation between timestep and metrics such as accuracy, cost, and computation time.

*Initiation – Boundary conditions* - There are multiple styles to define boundary conditions. User can employ periodic, or non-periodic including fixed or shrink-wrapped boundary conditions in all direction. When fixed conditions is specified, atoms departing from the unit cell boundary are eliminated. While in shrink-wrapped conditions, boundary adjusts to enclose all atoms continuously. Periodic boundary condition (PBC) are selected to approximate a large or infinite system by using a small part of the structure called a unit cell. In PBS the atoms can't escape the simulation box, if an atom move out of the simulation box the same atom enters the simulation box but through opposite side as shown in the Figure 14. In our case we opted periodic boundary conditions in all directions to approximate that the sheet is to large.



Figure 14: Schematic representation of the idea of periodic boundary conditions.

Atom definition – In this step, you have the option to either directly import a data file or manually create the structure by defining atom types, interatomic distances, arrangements, etc. In this step we also need to define the potential style and import the potential file to study the dynamic interactions of atoms.

*Setting* - In this step we write all the required setting and to the simulation box and atoms using energy minimization, ensembles (NVE, NVT and NPT), and some fixes. Each fix must be assigned a new ID to avoid simulation errors. To balance energy and force in a system we need to apply energy minimization technique, for BN nanosheet we employed conjugate gradient method, with energy and force tolerance values maintained at 10-10 eV and 10-12 eV/Å, respectively [59].

Similarly, to control the environment inside a unit cell LAMMPS provide different types of ensembles, some of the popular ensembles are NVE, NVT and NPT. A short details of this ensembles is given below [60].

*NVT*: This ensemble is used to keep the number of atoms, volume, and temperature constant in unit cell. Typically used when temperature and volume both need to be kept constant

*NPT*: It keeps number of atoms, pressure , and temperature constant, mainly used when pressure and temperature need to be kept constant.

*NVE*: It is used when the number of particle, volume and energy need to be kept constant.

In the case of hBN nanosheets we employed NPT ensemble by Nose– Hoover thermostat and barostat to equilibrate system at specified temperature and pressure [61]. A uniaxial tensile strain in the armchair direction at a steady strain rate of  $1 \times 10^9$  s<sup>-1</sup> [62] is applied to study the mechanical properties. Additionally, users need to define the format of the output, specifying the required data and the location for storing the results.

Run – In the last segment of the MD simulation, we define the number of iterations to be conducted. This is done by considering the timestep and the amount of parametric variation necessary.

### **4.3 Working principle of CMD simulations (LAMMPS)**

The main goal of performing simulation is to capture the trajectory or position (r<sub>ij</sub>) of each atom within a system over time. This iterative process begins by calculating the total interatomic energy (U) based on atom pair distances, encompassing various interactions such as Coulombic, van der Waals, and electronic repulsion. To integrate the contributions of each interaction we use potential such as Tersoff, ARIBO, MLIP etc. This potentials are a mathematical expression which provides the total energy of each atom based on interatomic distance. Subsequently, utilizing Newtonian mechanics, the differentiation of energy (U) with respect to interatomic distance (r) facilitates the computation of forces between atoms.

$$F_{(i,j)} = -\frac{\partial U}{\partial r}$$

This leads to determining acceleration and velocity for each atom. The velocity verlet algorithm was chosen for numerical integration, despite the Even though the algorithm's accuracy diminishes with larger time steps, considering smaller timesteps demonstrates excellent accuracy. The algorithm has shown itself to be quick, straightforward, stable, time-reversible, and memory efficient.

$$\mathcal{A}_{(i)} = \frac{F}{m}$$
  
v(i) (t+dt) = v(t) +  $\frac{dV(i)}{dt}$  dt + ....  
r(i) (t+dt) = r(t) +  $\frac{dr(i)}{dt}$  dt + ....

This cyclic progression, carried out at every timestep, effectively captures the trajectory, as depicted in Figure 15



Figure 15: The working principle and the math behind molecular dynamics simulation

The energy between a pair of atoms depends upon various interatomic interactions, including Coulombic, van der Waals, and electronic

repulsion forces between atoms. Calculating the total energy involves summing up these interactions. Next, the energy is differentiated with respect to the interatomic distance to derive the force acting on each atom. This force is then further differentiated with respect to the mass of each atom to determine the acceleration. By integrating the acceleration over a defined timestep, the velocity of each atom is updated. Finally, multiplying this velocity by the timestep yields the new position of each atom in the system, allowing for the dynamic simulation of molecular behavior.

## Chapter 5 : Result and discussion through new MLIP

# 5.1 Validation of newly developed MLIP through error metrics

The precision and efficacy of our newly developed MLIP have been quantitatively assessed by comparing its predictions with those from DFT based AIMD simulations. The performance of MLIP, especially in predicting the energy of atomic configurations, is pivotal as it serves as the basis for estimating other mechanical properties through CMD simulations.

Our MLIP demonstrated outstanding predictive capabilities, as illustrated in Figure 16. Upon rigorous examination of the energy predictions, based on the training and validation datasets, our model vielded an R-squared value of 0.99. This value signifies a near-perfect linear correlation between the energies estimated by MLIP and those obtained from AIMD simulations, thus confirming a 99% accuracy rate in energy prediction. The mean absolute error (MAE) and root mean square error (RMSE) serve as critical indicators of the potential's predictive accuracy. For the training set, the MAE was found to be 2.03  $\times$  10<sup>-3</sup> eV/atom, and the RMSE was 4.77  $\times$  10<sup>-3</sup> eV/atom. These low error margins suggest a high degree of precision in the potential's training phase. Comparatively, the validation set results, which are essential in establishing the model's generalizability, also reported minimal error with an MAE of  $1.6 \times 10^{-3}$  eV/atom and an RMSE of 1.35  $\times 10^{-3}$  eV/atom. The reduction in error rates from training to validation datasets is indicative of the robustness of the MLIP, ensuring its reliability when extrapolated to new, unseen data sets.



Figure 16: Comparing the energies derived from MLIP and ab initio simulation (a) training data and (b) validation data, with data points represented in blue and the identity line in black.

The performance of our MLIP is further validated when compared with the findings from previous research, particularly the study by author Novikov in 2020 [57]. Our training data achieved lower error outcomes compared to those reported in the earlier study, particularly demonstrating a lower energy error as illustrated in Table 2. The force and stress errors for the training phase were higher compared to Novikov's results. However, this is counterbalanced by the validation errors where our model demonstrated superior performance with significantly lower energy and stress errors of 1.35 meV/atom and 2.32 GPa, respectively, compared to the earlier study's validation results of  $5.10 \pm 1.12$  meV/atom and  $0.46 \pm 0.09$  GPa, respectively.

Table 2: A comparative assessment of training and validation errors, focusing on energy, force, and stress, compared to results from previous studies.

	Energy error	Force error	Stress error
	meV/atom	meV/ Å	GPa
Training	4.77	222	6.61
Training [57]	$5.54\pm2.10$	$175 \pm 4$	$0.46\pm0.04$
Validation	1.35	139	2.32
Validation [57]	$5.10\pm1.12$	$180 \pm 6$	$0.46\pm0.09$

## 5.2 Validation of MLIP through energy prediction

Figure 17 provides a visual comparison of energy predictions between the newly developed MLIP and AIMD simulations over time, effectively illustrating the dynamical evolution of the system's energy per atom during CMD simulations conducted under the NVT ensemble (constant number of particles, volume, and temperature). Initially, we observe that the CMD energy quickly converges to a range of fluctuation that closely tracks the AIMD energy. Despite the rapid initial changes, which could be attributed to the system equilibrating under the NVT conditions, both energies soon settle into a consistent pattern of fluctuation, suggesting that the CMD simulations are capturing the thermal motions and interactions within the structure with high fidelity. The overlay of the CMD and AIMD lines throughout the simulation time suggests that the MLIP is accurately predicting the same energetic trends as the AIMD simulations.



Figure 17: Fluctuation in energy prediction obtained through CMD and AIMD simulations.

The slight deviations between the CMD and AIMD energies that do appear are minimal, emphasizing the precision of the MLIP in replicating DFT-level accuracy. The amplitude of the energy variations in both lines is quite similar, indicating that the MLIP has effectively learned the potential energy surface as calculated by DFT to a degree that allows for near-equivalent predictions of system energy over time. Importantly, the ability of the MLIP to closely follow the AIMD results across all time steps both in the equilibration phase and during the established fluctuations supports the MLIP's robustness and transferability to different conditions, which are essential characteristics for reliable predictions in varied simulation scenarios.

# **5.3** Mechanical properties of monolayer BN sheet under uniaxial tension through new MLIP

#### 5.3.1 Impact of sheet size on mechanical property variation

Upon achieving satisfactory validation results for the newly developed potential, we employed it to investigate the correlation between sheets dimension/temperature and mechanical properties under uniaxial strain as shown in Figure 18.



Figure 18: Stress response in monolayer hBN sheet under uniaxial tensile loading with varying sheet size at 300 K

We observe that mechanical properties of the hBN sheets remain consistent across different dimensions as shown in Figure 18: Stress response in monolayer hBN sheet under uniaxial tensile loading with varying sheet size at 300 K. Sheets of varying dimensions exhibited a consistent trend, with the breaking stress of the majority of hBN sheets remaining close to 106 GPa. A similar pattern was observed for the breaking strain, which was around 16%, suggesting that the mechanical robustness of hBN is largely size-independent within the tested parameters.

Moreover, employing the linear fit method to the data collected at 1 K, we determined a Young's modulus of 994 GPa, which closely matches the results obtained through first-principle techniques in previous investigations [24]. Table 3 provides a detailed summary of structural performance with varying sheet size from  $30 \times 30$  Å<sup>2</sup> to  $150 \times 150$  Å<sup>2</sup>. The Young's modulus is relatively consistent across all sizes, indicating material uniformity. Breaking stress and breaking strain also shows minimal variation. The findings indicate that the mechanical characteristics are not influenced by sheet sizes.

Sheet size	Young's modulus	Breaking stress	Breaking strain
(Å <sup>2</sup> )	(GPa)	(GPa)	
30×30	995.5	110.237	0.1828
50×50	994.6	110.466	0.1796
80×80	994.8	110.391	0.179
100×100	992	110.495	0.1995
120×120	994.5	110.466	0.1796
150×150	994.6	110.548	0.1792

Table 3: Mechanical properties of hexagonal boron nitride (h-BN) sheets of varying sizes at 1 K

## 5.3.2 Impact of temperature on mechanical property variation

Following the confirmation of uniform mechanical properties across size variations, we proceeded to investigate temperature effects through additional simulations, with a constant sheet size chosen as  $100 \times 100$  Å<sup>2</sup>. We reported that with an increase in temperature both breaking stress as well as breaking strain reduces, as shown in Figure 19



Figure 19: Stress response in monolayer hBN sheet under uniaxial tensile loading with varying temperature.

With the increase in temperature from 1 K to 1500 K the breaking stress reduced from 110 GPa to 85.8 GPa whereas breaking strain reduced from 18% to 12.4% respectively. This negative correlation between temperature and mechanical strength is the result of the increase in kinetic energy of atoms with the increasing temperature. At lower temperatures, the reduced atomic vibrations correlate with a higher resistance to deformation, as evidenced by the higher breaking stress and strain. This requires more stress for displacement, strengthening the nanosheets. Conversely, at higher temperatures, increased atomic vibration allows large movement among atoms even with lower applied stress, resulting in weaker nanosheets. The temperature-induced softening of the hBN sheets can be attributed to the enhanced kinetic energy of the atoms, which lowers the energy barrier for bond rearrangement and leads to premature structural failure.

## 5.4 Variation of potential energy under uniaxial strain in monolayer hBN nanosheet

In addition, Figure 20 illustrates the variation in potential energy per atom in h-BN sheets of different sizes under uniaxial strain. Applying strain to a sheet results in an increase in the potential energy stored within the atoms of the sheet. As atoms are separated from their stable positions, the bond energy rises, resulting in gradual increase in the potential energy. As the potential energy reaches a peak, further strain on the sheet leads to crack initiation. The sudden growth of the crack leads to a rapid drop in potential energy as the atomic bond breaks. This is depicted by the sharp decline in potential energy, indicating that the sheet has fractured, and the atoms are rapidly returning to a lower energy state. The sudden failure of the sheet also indicates that the h-BN sheet shows brittle failure behavior [20].



Figure 20:Variation of the potential energy of different sheet sizes under uniaxial strain.





Figure 21: Stress distribution in monolayer hBN nanosheet experiencing uniaxial strain in the armchair direction.

Figure 21 illustrates the dynamic response of a monolayer hBN sheet as it is subjected to incremental uniaxial tensile strain, revealing the evolution of internal stress concentrations leading to fracture. The color gradient transitions from blue (low stress) to red (high stress) vividly illustrate the stress heterogeneity across the sheet, providing insight into the failure mechanism. Initially, in Figure 21 (a), the pristine lattice displays a uniform stress profile indicative of an equilibrated state with no loading condition. With the application of tensile strain, Figure 21 (b) illustrates the emergence of localized stress at atomic sites, signaling the initiation of the crack as the strain reaches 15.9%. These high-stress regions, depicted in red colors, highlight zones where atomic bonds are nearing their tensile limits. Figure 21 (c) showcases the rapid propagation of the crack as the strain approaches 16.1%, evident by the expansion of high-stress regions. The crack's propagation path appears to follow crystallographic directions, indicating the influence of lattice orientation on fracture behavior. The final phase of rupture is demonstrated in Figure 21 (d) at a strain of 16.3%, where the material exhibits a clean separation, confirming the brittle failure of hBN under the given conditions. This brittle fracture behavior is characteristic of hBN and underlines the material's high strength and low ductility under the tested conditions.

# 5.6 Mechanical response of bi-layer hBN through new MLIP

Upon witnessing the exceptional performance of the newly developed potential at elevated temperatures, we aimed to assess its effectiveness by subjecting it to an additional layer on the sheet. We developed a bilayer nanosheet mimicking the monolayer in AA' stacking manner with an interlayer distance of 3.4 Å and performed mechanical strain analysis under the same boundary conditions as shown in the Figure 22.



Figure 22: Multilayer BN sheet with AA' stacking

This additional analysis aimed to evaluate the versatility and reliability of the potential when applied to layered structures. Our findings demonstrate that the new potential performs exceptionally well, even in the context of bilayer.



5.6.1 Impact of sheet size on mechanical property variation

Figure 23: Stress response in bilayer hBN sheet under uniaxial tensile loading with varying sheet size,

Figure 23 shows the mechanical properties of hBN under uniaxial tension. On observing the graph, we can see that the same trend as followed in monolayer, i.e. the sheet strength remains independent of dimensions. With breaking stress and breaking strain at a 107.269 GPa and 15.9% respectively, thereby reinforcing the dimensional independence of the sheet's mechanical properties and mirroring the monolayer's behavior. This finding suggests a negligible influence of interlayer interactions on the intrinsic tensile properties of hBN, aligning with previous research conducted by Falin et al. in 2017 [63].

#### 5.6.2 Impact of temperature on mechanical property variation

Correspondingly, increase in temperature result in a gradual weakening of strength as depicted in Figure 24, demonstrating a trend similar to that seen in monolayer configurations. The consistent trend across monolayer and bilayer systems indicates that the newly developed potential captures the essential physics governing the mechanical response of hBN, thereby proving its efficacy for simulations that span a wide range of structural configurations and thermal environments.



Figure 24: Stress response in bilayer hBN sheet under uniaxial tensile loading with varying temperature.

### 5.7 Dynamic response of bi-layer hBN

Figure 25 portrays the stress response and fracture progression of a bilayer hBN sheet under uniaxial tensile strain. Initially, Fig. 25 (a) presents the bilayer sheet in an unstressed state, serving as a baseline for subsequent comparison. As the strain reaches 15.7%, the onset of stress concentration primarily in the top layer, visualized through the color gradient with red indicating areas of highest stress as shown in Fig. 25(b). This gradient reflects the non-uniform distribution of tensile forces within the sheet. With further increases in strain to 15.84%, critical stress levels lead to the initiation of microcracks in the top layer as seen in Fig. 25(c), marking the beginning of the material's failure process. Upon reaching 15.87% strain, these microcracks coalesce into larger fractures, leading to the separation of the top layer, as shown in Fig. 25(d). This phase highlights the brittle fracture mechanism,

characterized by the rapid propagation of cracks without significant plastic deformation.



Figure 25. Stress distribution in bilayer hBN nanosheet experiencing uniaxial strain in the armchair direction.

Once the top layer fractures, the load redistribution to the bottom layer results in renewed stress accumulation. This is evident in Fig. 25(e), where the bottom layer begins to exhibit similar stress patterns to those initially observed in the top layer. The fracture process culminates in Fig. 25(f) with the complete separation of the bottom layer at a strain of 15.9%, mirroring the failure mode of the top layer. Throughout this process, the stress distribution visualized in the bilayer hBN sheet provides valuable insights into the layer-by-layer failure mechanism. This detailed depiction confirms that while individual layers may fail sequentially, the overall material retains its brittle fracture characteristics irrespective of mono or bilayer configurations.

## **Chapter 6 : Conclusion and Future Scope**

#### **6.1** Conclusion to the current work

This study extensively examine various methods for investigating the structural properties of nanostructures, particularly emphasizing the DFT based AIMD, and EIP based CMD, while also discussing their respective strengths and limitations. To bridge the gap between these approaches, a novel method is introduced known as MLIP, a potential that is developed on small set of accurate outcomes using machine learning. We constructed MLIP for investigating the mechanical properties of hBN nanosheets, examining how these properties vary with temperature, size configurations, and layer additions. Our discussions also comprehensively covers the entire procedure for constructing MLIP, elaborating on every step from dataset creation, subsampling, training, to its validation. For MLIP development, we acquired accurate dataset by executing numerous AIMD simulations on a compact unit cell of hBN. To validate the newly developed potential we evaluated its predictive performance through various validation approaches. The outcomes of the validation process highlighted that the predictions of energy and atomic forces closely resemble DFT accuracy, indicating the MLIPs reliability.

Subsequently, we employed the new MLIP as an interatomic potential in our CMD simulations to study the structural properties of large-sized hBN sheets. The result from the CMD simulations on hBN nanosheets with varying dimensions, temperature, and layer addition shows consistency with the previous studies for properties such as Young's modulus, breaking stress, and breaking strain. This study highlights that the mechanical strength of hBN sheet is independent of sheet dimensions and the addition of layers. However, it reduces gradually with increasing temperature, emphasizing the robustness of MLIP across different environmental conditions. The MLIPs also offers opportunities to investigate the properties of complex nanomaterials containing defects, thereby broadening their scope for application.

## **6.2 Future Scope**

Our study sets the pathway for the development of new flexible interatomic potentials for emerging nano structures, facilitating the examination of structural properties in diverse environments with enhanced precision, while also reducing computational time and costs. Furthermore, training the current potential with polarization data performed on AIMD will enhance its ability to accurately predict the piezoelectric and flexoelectric effects in hBN nanosheets.

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