

Development and application of machine-learned interatomic potentials for the study of high-temperature TiAlNb alloys

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Abstract

The lack of accurate and efficient interatomic potentials for technologically vital TiAlNb alloys has hindered their computational design and optimization for aerospace and energy applications. The thesis addresses the gap through a hierarchical investigation that begins by establishing the limitations of a conventional classical interatomic potential for the system. To address these limitations, the work then details the development and application of two state of the art machine learning interatomic potentials (ML-IAPs), the neural network-based deep potential molecular dynamics (DeePMD) and the linear regression-based moment tensor potentials (MTP), to investigate the influence of Niobium on the thermomechanical properties of TiAlNb alloys. Furthermore, the efficiency of the interatomic potential development process itself is systematically evaluated by contrasting a traditional passive learning approach with a data-efficient active learning strategy. The results demonstrate that the developed ML-IAPs achieve near-*ab initio* accuracy, revealing a clear trade-off between the higher fidelity of DeePMD and the greater computational and data efficiency of MTP. Critically, the active learning strategy is shown to reduce the required number of *ab initio* calculations by an order of magnitude without sacrificing model accuracy. Application of these potentials confirms that Nb addition enhances ductility while decreasing the strength of TiAl phases, providing a clear mechanistic link through simulated tension tests and the analysis of stacking fault energies. This research thus makes three principal contributions: it delivers a quantitative and mechanistic understanding of Niobium’s influence on the thermomechanical properties of TiAl-based alloys; it provides a set of rigorously validated, high-fidelity machine learning (ML) potentials for the TiAlNb system; and it presents a detailed methodological guide for the efficient construction of such potentials via active learning. The benchmark datasets are made publicly available to accelerate the data-driven design of high-performance materials.

Contents

List of figures	vii
List of abbreviations	ix
1 Introduction	1
1.1 Background and motivation	1
1.2 The interatomic potential dilemma	2
1.3 The machine learning approach to interatomic potentials	3
1.4 Research gaps and open questions	3
1.5 Outline of the thesis	4
2 Fundamentals of material modeling	7
2.1 Overview of computational framework	7
2.2 Density functional theory	7
2.3 <i>Ab initio</i> molecular dynamics	10
2.4 Classical molecular dynamics and force fields	12
2.5 Machine learning based interatomic potentials	16
2.5.1 Deep potential molecular dynamics	18
2.5.2 Moment tensor potentials	22
2.6 Active learning	24
2.6.1 Active learning for DeePMD	26
2.6.2 Active learning for MTP	27
3 Thermomechanical properties of TiAlNb using Farkas’s interatomic potential	31
3.1 Introduction and motivation	31
3.2 Key findings and contribution of Article A.1	34
3.3 Results and discussion	34
3.3.1 Validation of the Farkas potential in binary Ti-Al systems	34
3.3.2 First-principles insights into Nb alloying effects	35
3.3.3 Thermomechanical behavior of single-lamellar interfaces	36
3.3.4 Deformation mechanisms in microstructure-informed models	37
3.3.5 Summary	38

Contents

4	Moment tensor vs deep learning: a comparison	41
4.1	Introduction and motivation	41
4.2	Key findings and contribution of Article A.2	43
4.3	Results and discussion	43
4.3.1	Model parameterization and initial error analysis	43
4.3.2	Prediction of equilibrium, elastic and thermal properties	44
4.3.3	Alloying effects on mechanical behavior	45
4.3.4	Computational efficiency	46
4.3.5	Summary	46
5	Improvement through active learning	49
5.1	Introduction and motivation	49
5.2	Key findings and contribution of Article A.3	50
5.3	Results and discussion	51
5.3.1	Data requirements and error analysis: Active versus passive learning . .	52
5.3.2	Prediction of equilibrium and elastic properties	53
5.3.3	Structural fidelity at finite temperatures	54
5.3.4	Influence of Nb on thermomechanical behavior	54
5.3.5	Summary	55
6	Conclusion	57
	Bibliography	60
A	Peer reviewed and published articles	71
A.1	Publication 1	71
A.2	Publication 2	91
A.3	Publication 3	117

List of figures

2.1	Schematic of self-consistent field cycle in Kohn–Sham density functional theory	9
2.2	Schematic of the <i>ab initio</i> molecular dynamics simulation loop	12
2.3	Lennard–Jones 12–6 potential energy $E(r) = V(r) = 4\varepsilon[(\sigma/r)^{12} - (\sigma/r)^6]$ for argon-like parameters $\varepsilon = 100 \text{ cm}^{-1}$ and $\sigma = 3.4 \text{ \AA}$. The repulsive $+A/r^{12}$ (blue dashed), attractive $-B/r^6$ (orange dash-dot), and total potential energy (red solid) are plotted as functions of interatomic distance $r = 3.0\text{--}8.0 \text{ \AA}$. The vertical double-headed arrow indicates the well depth ε , and the horizontal marker at $r = \sigma$ shows the distance where $E(r) = 0$ (collision diameter).	15
2.4	Comparison of deep potential molecular dynamics and moment tensor potential architectures	18
2.5	Schematic of the active learning cycle	26
3.1	Atomistic models of the Titanium–Aluminium–Niobium (TiAlNb) system at multiple length scales	32
3.2	Effect of Niobium concentration and temperature on microstructure-informed atomistic models (MIAMs) deformation	38
4.1	Comparison of accuracy and performance of moment tensor potential and deep potential molecular dynamics	44
5.1	Comparison of active and passive learning strategies for moment tensor potential and deep potential molecular dynamics	51

List of abbreviations

Abbreviation	Description
3D	Three-dimensional
ABOP	Analytical bond-order potential
AIMD	<i>Ab initio</i> molecular dynamics
ASCF	Atom-centered symmetry function
at. %	Atomic percentage
B	Boron
BCC	Body-centered cubic
BOMD	Born–Oppenheimer molecular dynamics
C	Carbon
CPMD	Car–Parrinello molecular dynamics
CSF	Complex stacking fault
CSP	Centrosymmetry parameter
Cr	Chromium
DeePMD	Deep potential molecular dynamics
DFT	Density functional theory
DMFT	Dynamical mean-field theory
DP	Deep potential
DXA	Dislocation extraction algorithm
EAM	Embedded atom method
ELF	Electron localization function

Continued on next page

List of abbreviations

Abbreviation	Description
GAP	Gaussian approximation potential
GGA	Generalized gradient approximation
GPU	Graphics processing unit
GSFE	Generalized stacking fault energy
HCP	Hexagonal close-packed
LDA	Local density approximation
LPT	Low-pressure turbine
MD	Molecular dynamics
MEAM	Modified embedded atom method
MIAMs	Microstructure-informed atomistic models
ML	Machine learning
ML-IAPs	Machine learning interatomic potentials
Mn	Manganese
Mo	Molybdenum
MTP	Moment tensor potential
Nb	Niobium
PBC	Periodic boundary condition
PDOS	Partial density of states
PES	Potential energy surface
RDF	Radial distribution function
RMSE	Root mean square error
SCF	Self-consistent field
Si	Silicon
SISF	Superlattice intrinsic stacking fault
SOAP	Smooth overlap of atomic positions
Ta	Tantalum

Continued on next page

List of abbreviations

Abbreviation	Description
TDDFT	Time-dependent density functional theory
TiAlNb	Titanium–aluminium–niobium
UTS	Ultimate tensile strength
V	Vanadium
W	Tungsten
γ -TiAl	Gamma-titanium aluminide

Chapter 1

Introduction

1.1 Background and motivation

The continued advancement of technologies in the aerospace and power generation industries is intrinsically linked to the development of new high-performance materials. The demand for lighter, stronger, and more efficient systems requires materials capable of withstanding severe conditions of high temperature and mechanical stress. This pursuit has driven extensive research into novel material systems that can exceed the performance limits of conventional alloys at high temperatures.

Among the most promising candidates for these demanding applications are intermetallic alloys based on gamma titanium aluminide (γ -TiAl). These materials are widely regarded as excellent structural materials due to their low density, good oxidation resistance, high strength, and creep resistance at elevated temperatures^{1,2}. They offer a good combination of properties that make them uniquely suited for high-temperature structural components. The development and application of TiAl-based alloys accelerated significantly following General Electric's* pioneering decision to use cast TiAl blades in the low-pressure turbine (LPT) stages of their GENx jet engine³. Beyond aerospace, the automotive industry has also shown interest, exploring the use of TiAl in turbocharger turbine wheels to improve engine response and meet future environmental standards⁴⁻⁶.

Despite their immense potential and successful implementation, the broader application of binary TiAl alloys has been historically hindered by their significant drawback of poor ductility and fracture toughness at room temperature. This inherent brittleness complicates manufacturing processes and raises concerns about the damage tolerance and reliability of components during service. To overcome these limitations, a primary materials design strategy is the introduction of ternary and quaternary alloying elements to binary TiAl alloys^{1,5,7}.

The addition of Niobium (Nb), particularly in the concentration range of 5-10 at.%, has been identified as a highly effective strategy for enhancing the thermomechanical properties of TiAl-based systems^{1,2,5,8}. The resulting ternary TiAlNb (Titanium-Aluminium-Niobium)

*General Electric Company, often abbreviated as GE, is an American multinational company. Its aerospace division is a leading producer of jet engines for commercial and military aircraft.

alloys thus represent a more viable and robust material system for industrial applications. However, the thermomechanical properties of these alloys are highly sensitive to their precise elemental composition and the complex microstructures that form during processing. Therefore, identifying the optimal composition to achieve the desired balance of strength, ductility, and high-temperature stability constitutes a major and ongoing challenge in modern materials science, and provides the central motivation for this thesis.

1.2 The interatomic potential dilemma

The traditional, experiment-driven approach to materials development, is often a slow, costly, and laborious process. The synthesis and characterization of numerous alloy compositions through physical trial-and-error can take years, representing a significant bottleneck in the innovation cycle. To overcome these limitations and accelerate the design of new materials like the TiAlNb alloys described previously, the field of materials science has increasingly turned to computational modeling and simulation^{9,10}. This digital revolution allows researchers to investigate material behavior in a virtual environment, providing insights that are often difficult or impossible to obtain through direct experimentation.

Among the most powerful tools in computational materials science is molecular dynamics (MD) simulation^{11–14}. In essence, MD serves as a virtual microscope, enabling us to observe the collective behavior of thousands to millions of atoms as they move and interact over time. By analyzing the collective motion and interaction of atoms in an MD simulation, it is possible to compute a wide range of macroscopic thermomechanical properties that are directly comparable to experimental measurements. This approach allows for the rapid virtual screening of many candidate compositions, significantly narrowing the experimental search space and guiding the design of new, optimized alloys. However, the predictive accuracy of any MD simulation depends entirely on its interatomic potential, which acts as a rulebook governing the forces between atoms. The choice of this potential represents a fundamental compromise in materials science, often called the ‘accuracy versus speed’ dilemma. This trade-off forces a choice between models that lie at opposite ends of a wide spectrum of computational cost and physical fidelity.

At one end of the spectrum are the methods derived from first-principles quantum mechanics techniques such as density functional theory (DFT)^{15–19}, which provide a highly accurate description of atomic interactions by solving approximations to the Schrödinger equation. By explicitly modeling the behavior of electrons, DFT can capture the complex nature of chemical bonding, including the metallic and covalent characteristics present in intermetallic alloys like TiAlNb. The complexity of solving the electronic structure problem limits such first-principle based calculations to systems of only a few hundred atoms for timescales of mere picoseconds. This scale is insufficient for studying many critical material phenomena, such as the evolution of microstructures or the interaction of extended defects, which require millions of atoms and nanoseconds of simulation time.

At the opposite end of the spectrum are classical interatomic potentials. These models employ relatively simple, empirically-fitted analytical functions to describe the atomic inter-

actions, forgoing any explicit treatment of electrons. This simplification makes them incredibly fast, enabling simulations of millions or even billions of atoms for long timescales^{20,21}. However, this computational efficiency is achieved at the cost of accuracy and transferability. The rigid functional forms of classical potentials are often unable to capture the nuanced and environment-dependent nature of bonding in complex, multi-component alloys.

This dilemma has historically forced researchers into a difficult compromise. For decades, the study of complex, technologically relevant materials has been caught between the quantum mechanical methods that are too slow and the classical potentials that are too inaccurate^{22,23}. This significant gap has been a major bottleneck, limiting the ability of simulations to provide truly predictive insights for real-world engineering problems. Overcoming this fundamental trade-off is the primary motivation for the development of the new generation of interatomic potentials that are the focus of this thesis.

1.3 The machine learning approach to interatomic potentials

In recent years, a new approach has emerged^{22,24,25} that promises to resolve the long-standing ‘accuracy versus speed’ dilemma in atomistic simulations which is the development of machine learning interatomic potentials (ML-IAPs). This approach fundamentally changes how interatomic potentials are constructed. Instead of relying on human-intuited, fixed functional forms, ML-IAPs employ highly flexible machine learning (ML) algorithms to learn the complex, high-dimensional relationship between the local arrangement of atoms and the system’s potential energy.

The underlying concept is to combine the best of both worlds. A database of atomic configurations is first generated, for which the energies and forces are calculated with a high-accuracy but computationally expensive first-principle based method. This high-quality dataset then serves as the training data for the ML model. The model learns to reproduce the potential energy surface of the quantum mechanical method, effectively acting as a highly accurate surrogate model. Once trained, the ML-IAP can predict energies and forces with near-DFT accuracy but at a computational cost that is orders of magnitude lower, approaching that of classical potentials.

1.4 Research gaps and open questions

Despite the extensive research on TiAl-based alloys, a review of the current literature^{1,2,5,8,26,27} reveals several critical gaps in understanding, particularly concerning the specific role of Nb. Most studies on advanced TiAl alloys involve complex, multicomponent systems containing not only Niobium (Nb) but also other alloying elements such as Molybdenum (Mo), Boron (B), Carbon (C), Silicon (Si), Chromium (Cr), Manganese (Mn), Vanadium (V), Tungsten (W), and Tantalum (Ta), among others. While these investigations are vital for developing engineering alloys, the synergistic effects of multiple elements make it difficult to distinguish and isolate the fundamental influence of Nb on the underlying thermomechanical properties. A systematic

investigation focused on the ternary TiAlNb system is therefore needed to provide a baseline understanding that can inform the design of more complex alloys.

Furthermore, a significant challenge for performing such an investigation with atomistic simulations has been the lack of a suitable interatomic potential. While MD is an ideal tool for this problem, its predictive power is entirely dependent on the accuracy of the potential used to describe the TiAlNb interactions. Initial work²⁸, detailed in Chapter 3, employed an existing classical potential for this system. However, this potential was found to have significant limitations in the composition range of interest (5-10 at.% Nb), highlighting a critical need for a more accurate and robust model. The development of a high-fidelity interatomic potential for the TiAlNb system is therefore a primary prerequisite for making reliable predictive simulations.

This necessity directly motivates the central focus of this thesis, which is to leverage the power of ML-IAPs to overcome the limitations of classical models. As introduced previously, ML-IAPs offer a pathway to achieving near-quantum mechanical accuracy at a manageable computational cost. This work focuses on the development, validation, and comparison of two state-of-the-art ML-IAP frameworks. The first is deep potential molecular dynamics (DeePMD), a method that employs deep neural networks to learn the atomic interactions. The second framework is moment tensor potentials (MTP), which represents the atomic environment using a systematic, physics-inspired basis set and fits the potential using linear regression.

Finally, a key challenge in creating ML-IAPs is the generation of the extensive first-principle based training data they require. This thesis therefore also investigates the use of active learning, an advanced training strategy that can reduce the computational expense of data generation. By intelligently and iteratively selecting only the most informative configurations, active learning addresses a major bottleneck in the ML-IAP development workflow. By combining these cutting-edge techniques, this research aims to create robust and accurate models for the TiAlNb system, enabling the large-scale, predictive simulations necessary to resolve the open questions regarding Nb’s influence on this technologically vital class of materials.

1.5 Outline of the thesis

This thesis is structured into the following chapters:

- **Chapter 2** provides the theoretical and computational background for the methods employed throughout this work. It begins with an overview of fundamental simulation techniques, including MD, and first-principles methods such as DFT and *ab initio* molecular dynamics (AIMD). The chapter then details the two ML-IAP frameworks investigated: DeePMD and MTP. The section concludes with a detailed discussion of the active learning strategies used to construct these advanced interatomic potentials.
- **Chapter 3** presents an initial investigation into the influence of Nb on the thermomechanical properties of TiAl-based alloys using an existing classical interatomic potential, the Farkas ternary interatomic potential²⁹. This study serves as a baseline and critically evaluates the capabilities and limitations of this classical potential for the TiAlNb system. The findings

from this chapter highlight the deficiencies of the classical model and provide the primary motivation for developing more accurate ML-based potentials.

- **Chapter 4** details the development and validation of new interatomic potentials for the TiAlNb system using both the MTP and DeePMD frameworks. A comprehensive comparative analysis is performed to evaluate the potentials based on their predictive accuracy for a range of fundamental material properties, including lattice parameters, elastic constants, equilibrium volume, and key defect energies such as stacking-fault energies, as well as ultimate tensile strength. The computational cost associated with training and deploying each potential is also systematically assessed.
- **Chapter 5** presents a comparative study of learning strategies by implementing and evaluating active learning for both the MTP and DeePMD frameworks. This chapter contrasts the data efficiency and final model performance of potentials trained via active learning with those trained using a traditional passive learning approach. The potentials developed using both learning strategies are rigorously compared to assess the impact of the training methodology on predictive accuracy and the overall balance between computational cost and model fidelity.
- **Chapter 6** summarizes the principal findings of this thesis, synthesizes the conclusions drawn from the comparative studies, and provides an outlook on potential future research directions in the field of ML-driven materials design for high-temperature alloys.

Chapter 2

Fundamentals of modeling

2.1 Overview of computational framework

This chapter provides a detailed overview of the computational methods for material modeling employed in this work, which span a range of time and length scales. A primary focus is to establish the connection between these methods, illustrating how they serve as essential components in the development of machine learning interatomic potentials. The discussion explains how each method offers complementary insights, from quantum mechanical accuracy and long timescale dynamics to data-driven modeling, which ultimately contribute to the creation of a robust potential model for TiAlNb alloys.

2.2 Density functional theory

The many-body Schrödinger equation, which governs a system of interacting electrons, is given by

$$\hat{H} \Psi(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_N) = E \Psi(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_N), \quad (2.1)$$

where $\hat{H}$ is the Hamiltonian operator, E is the total energy of the system, and $\Psi(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_N)$ is the many-electron wavefunction. The wavefunction is a complex-valued function that depends on the 3D spatial coordinates $\mathbf{r}_i$ of each of the N electrons in the system. The Hamiltonian includes the kinetic energy of the electrons, their interaction with an external potential (typically arising from atomic nuclei), and the electron-electron Coulomb repulsion. Although this equation, in principle, yields the exact ground-state wavefunction containing all information about the system, its solution is computationally intractable for large systems because Ψ is defined in a $3N$ -dimensional space, causing an exponential scaling in complexity.

To address these challenges, DFT was developed. DFT bypasses the need for the full many-body wavefunction by establishing that the ground-state properties of a system can instead be uniquely determined from the electron density, $\rho(\mathbf{r})$. Unlike the wavefunction, which depends on the individual coordinates of all N electrons, the electron density is a function of a single 3D position vector $\mathbf{r}$. It describes the probability of finding any of the N electrons at a given point $\mathbf{r}$ in space. Early work by Thomas and Fermi in 1927^{30,31} emphasized the

importance of the total electron density in describing many-body systems; however, the effects of exchange and correlation, arising respectively from the antisymmetry of the many electron wavefunction (Pauli exclusion principle) and the Coulomb interactions between electrons, were not fully understood at that time. The seminal work of Hohenberg, Kohn, and Sham in the 1960s^{17,18} laid the foundation of modern DFT. The Hohenberg–Kohn theorems demonstrated that the ground-state electron density uniquely determines all properties of a quantum system in the absence of a magnetic field. The first Hohenberg–Kohn theorem establishes a one-to-one correspondence between the external potential $V_{\text{ext}}(\mathbf{r})$ and the ground-state electron density $\rho(\mathbf{r})$, meaning that $\rho(\mathbf{r})$ encodes all the information of the many-body system. The second theorem provides a variational principle: for any physically acceptable trial density $\tilde{\rho}(\mathbf{r})$ (one that is non-negative and integrates to the correct number of electrons), the corresponding energy functional $E[\tilde{\rho}]$ will always be greater than or equal to the true ground-state energy E_0 ,

$$E_0 \leq E[\tilde{\rho}], \quad (2.2)$$

with equality achieved only when $\tilde{\rho}(\mathbf{r})$ is the exact ground-state density.

Although the Hohenberg–Kohn theorems provide the theoretical basis for DFT, they do not prescribe a practical method for computing the electron density. This gap is bridged by the Kohn–Sham approach, which reformulates the complex many-electron problem into a more manageable set of independent, single-particle equations. The many-body Schrödinger equation is thereby replaced by the Kohn–Sham equation¹⁸:

$$\left[-\frac{\hbar^2}{2m} \nabla^2 + v_{\text{KS}}(\mathbf{r}) \right] \psi_i(\mathbf{r}) = \epsilon_i \psi_i(\mathbf{r}) \quad (2.3)$$

In this equation, $\psi_i(\mathbf{r})$ are the single-particle Kohn–Sham orbitals, which represent the wavefunctions of a fictitious system of non-interacting electrons that are constrained to have the same ground-state density as the real, interacting system. The term $\hbar$ is the reduced Planck constant, m is the mass of the electron, ∇^2 is the Laplacian operator representing the kinetic energy, and ϵ_i are the corresponding orbital energies. The central challenge of this approach lies in defining the effective Kohn–Sham potential, $v_{\text{KS}}(\mathbf{r})$. It is composed of three distinct terms: the external potential $v_{\text{ext}}(\mathbf{r})$ (typically from the atomic nuclei), the Hartree potential $v_{\text{H}}[\rho](\mathbf{r})$ representing the classical electrostatic repulsion between electrons, and the exchange-correlation potential $v_{\text{xc}}[\rho](\mathbf{r})$. The latter term encapsulates all the complex many-body quantum mechanical effects not covered by the other terms. The full potential is thus expressed as:

$$v_{\text{KS}}(\mathbf{r}) = v_{\text{ext}}(\mathbf{r}) + v_{\text{H}}[\rho](\mathbf{r}) + v_{\text{xc}}[\rho](\mathbf{r}) \quad (2.4)$$

The total electron density is reconstructed from the N occupied orbitals as

$$\rho(\mathbf{r}) = \sum_{i=1}^N |\psi_i(\mathbf{r})|^2. \quad (2.5)$$

Because $v_{\text{KS}}(\mathbf{r})$ depends on $\rho(\mathbf{r})$ and vice versa, the Kohn–Sham equations are solved self-

consistently via an iterative procedure: starting with an initial guess for $\rho(\mathbf{r})$, one computes $v_{\text{KS}}(\mathbf{r})$, solves for the orbitals $\psi_i(\mathbf{r})$, updates $\rho(\mathbf{r})$, and repeats this cycle until convergence is achieved. Figure 2.1 shows the self consistent cycle of DFT.

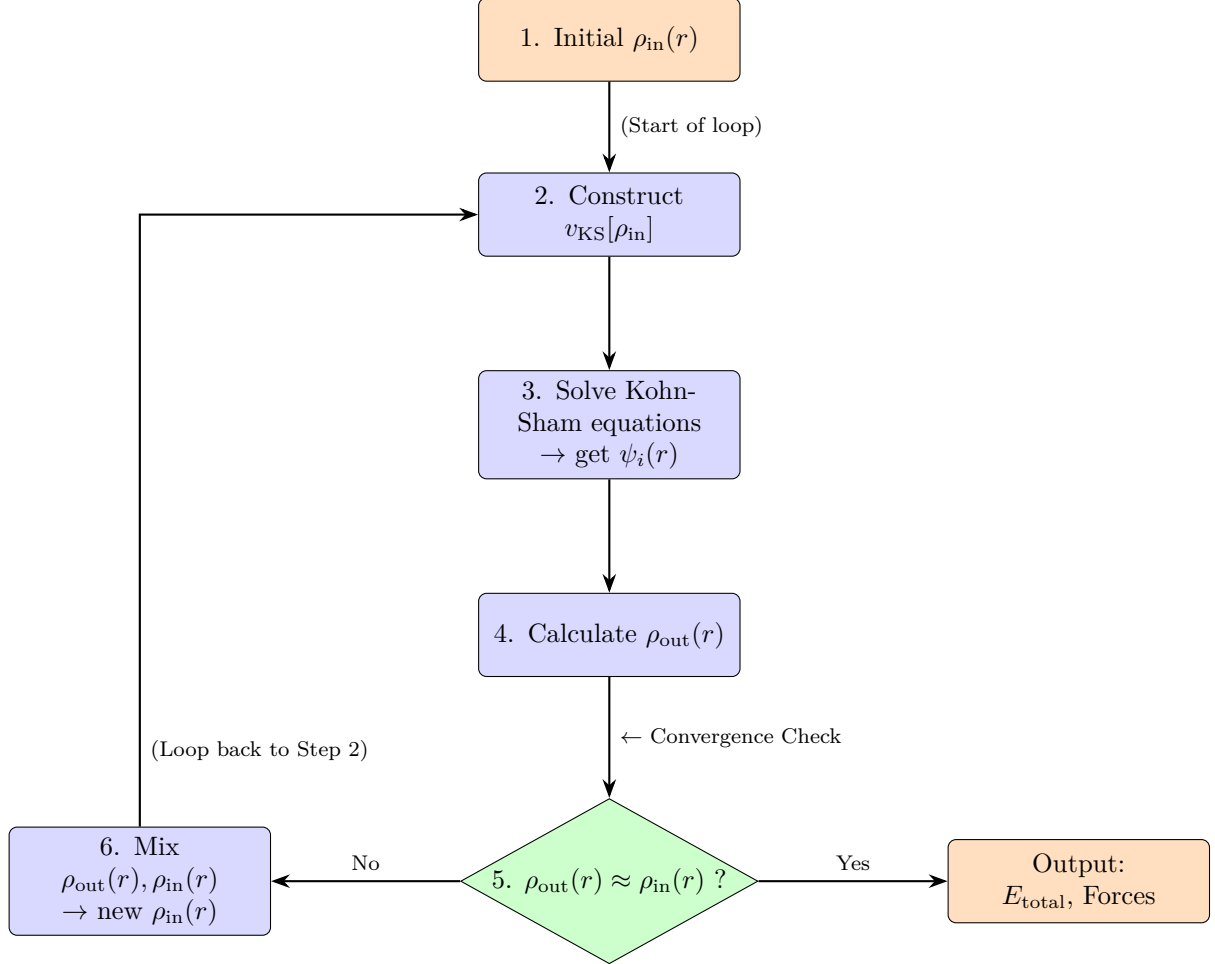


Figure 2.1: Schematic of the iterative self-consistent field (SCF) cycle in Kohn-Sham DFT. The calculation refines an initial guess for the electron density, ρ_{in} , by repeatedly constructing the Kohn-Sham potential, solving for the orbitals, and calculating a new output density, ρ_{out} . The cycle continues until self-consistency is reached ($\rho_{\text{in}} \approx \rho_{\text{out}}$), at which point the converged solution yields the ground-state energy and atomic forces.

The total ground-state energy within the Kohn-Sham formalism is then expressed as

$$E_{\text{KS}}[\rho] = T_s[\rho] + E_{\text{H}}[\rho] + E_{\text{ext}}[\rho] + E_{\text{xc}}[\rho], \quad (2.6)$$

where $T_s[\rho]$ is the kinetic energy of the non-interacting electrons, $E_{\text{H}}[\rho]$ is the Hartree energy, $E_{\text{ext}}[\rho]$ is the energy due to the external potential, and $E_{\text{xc}}[\rho]$ is the exchange-correlation energy. The latter term, $E_{\text{xc}}[\rho]$, is particularly significant as it accounts for both the electron

correlation effects, describing how each electron is influenced by all others, and the exchange interaction, which arises from the antisymmetry of the many-electron wavefunction (related to the Pauli exclusion principle). Since its exact form is unknown, $E_{xc}[\rho]$ is approximated in practice. The simplest approximation is the local density approximation (LDA)¹⁸, where E_{xc} depends only on the local value of the electron density. More refined approaches, such as the generalized gradient approximation (GGA)³², include the density gradient to better capture spatial variations in $\rho(\mathbf{r})$. Further enhancements involve meta-GGAs^{33,34}, which consider the kinetic energy density or the second derivative of the density, and hybrid functionals that incorporate a fraction of exact exchange from Hartree–Fock theory³⁵.

In addition to these theoretical considerations, DFT faces several computational challenges. The $\mathcal{O}(N^3)$ scaling of most DFT methods limits their applicability to very large systems. Convergence issues in the iterative solution of the Kohn–Sham equations and the choice of basis sets or grids can also significantly impact both the accuracy and efficiency of simulations. Moreover, standard DFT functionals can struggle with weak van der Waals interactions and finite-temperature effects, often necessitating further corrections or more sophisticated methodologies such as DFT+U³⁶, dynamical mean-field theory (DMFT)³⁷, or time-dependent DFT (TDDFT)³⁸ for excited-state or dynamic simulations.

Overall, DFT offers a robust framework for accurately calculating the ground-state properties of many-electron systems by reducing the complex, many-body problem to the determination of a three-dimensional electron density. This foundational approach has revolutionized computational materials science, chemistry, and condensed matter physics, providing essential insights into the behavior of complex systems while guiding the development of more efficient and scalable methods for studying both static and dynamic phenomena.

2.3 *Ab initio* molecular dynamics

While DFT has been instrumental in providing accurate ground state properties of materials by focusing on the electron density instead of the full many electron wavefunction, its static nature prevents it from capturing the time dependent evolution of atomic systems. Many material properties and phenomena such as thermal fluctuations, phase transitions, diffusion, and chemical reactions are inherently dynamic and thus require a time dependent treatment. In this context, *Ab initio* molecular dynamics (AIMD) emerges as a powerful approach that extends DFT by incorporating the temporal evolution of atomic configurations.

AIMD addresses the limitations of static DFT by performing DFT calculations “on the fly” during the simulation. At each time step, the electronic ground state is determined through the self-consistent solution of the DFT equations for the current atomic arrangement. The force on the i -th atom, $\mathbf{F}_i$, is then computed as the negative gradient of the total DFT energy with respect to its atomic position:

$$\mathbf{F}_i = -\frac{\partial E_{\text{DFT}}}{\partial \mathbf{R}_i}, \quad (2.7)$$

where E_{DFT} is the total energy obtained from DFT and $\mathbf{R}_i$ denotes the position of the i -th atom. This on-the-fly force evaluation ensures that the instantaneous electronic response to structural changes is accurately captured, thereby providing high-fidelity data for each configuration.

Once the forces are available, the atomic positions and velocities are updated using classical Newtonian dynamics:

$$m_i \frac{d^2 \mathbf{R}_i}{dt^2} = \mathbf{F}_i, \quad (2.8)$$

with m_i representing the mass of the i -th atom. By integrating these equations of motion, AIMD simulates the dynamic behavior of the system, enabling the study of finite-temperature effects, transient states, and reaction mechanisms that are inaccessible through static DFT alone.

A key aspect of AIMD is its iterative, self-consistent field (SCF) procedure at every time step as observed in the case of DFT (as depicted in Figure 2.2). For each new atomic configuration, an initial guess for the electron density is generated, followed by iterative DFT calculations until convergence is reached. This converged electron density is then used to compute the forces, ensuring that the electronic and ionic degrees of freedom are tightly coupled throughout the simulation.

Within AIMD, two primary variants have evolved to balance computational efficiency and accuracy. In Born–Oppenheimer molecular dynamics (BOMD)³⁹, the electronic structure is fully converged at each time step before the forces are evaluated. This method treats electrons and nuclei separately and tends to yield highly accurate results; however, it is computationally demanding since a full self-consistent DFT calculation is required for every time step. Alternatively, Car–Parrinello molecular dynamics (CPMD)⁴⁰ adopts an extended Lagrangian approach in which electronic orbitals are treated as additional dynamical variables alongside the nuclear positions. By evolving the electronic and ionic degrees of freedom concurrently, CPMD avoids full SCF convergence at each step, resulting in enhanced computational efficiency. However, CPMD requires careful tuning of parameters such as the fictitious electron mass to ensure the adiabatic separation between electronic and nuclear motions.

In essence, AIMD leverages the accuracy of DFT for electronic structure determination while simultaneously incorporating classical molecular dynamics to simulate the time evolution of the system. Although AIMD is significantly more computationally intensive than static DFT, its ability to capture realistic dynamics renders it indispensable for studying time-dependent phenomena. Moreover, high-fidelity AIMD data are often used to develop machine learning interatomic potentials²² and to benchmark classical force fields^{41,42}, bridging the gap between quantum mechanical accuracy and the computational efficiency required for large-scale simulations.

By incorporating the temporal dimension into material simulations, AIMD not only overcomes the static limitations of DFT but also provides insights into the interplay between electronic and ionic motions under realistic operational conditions.

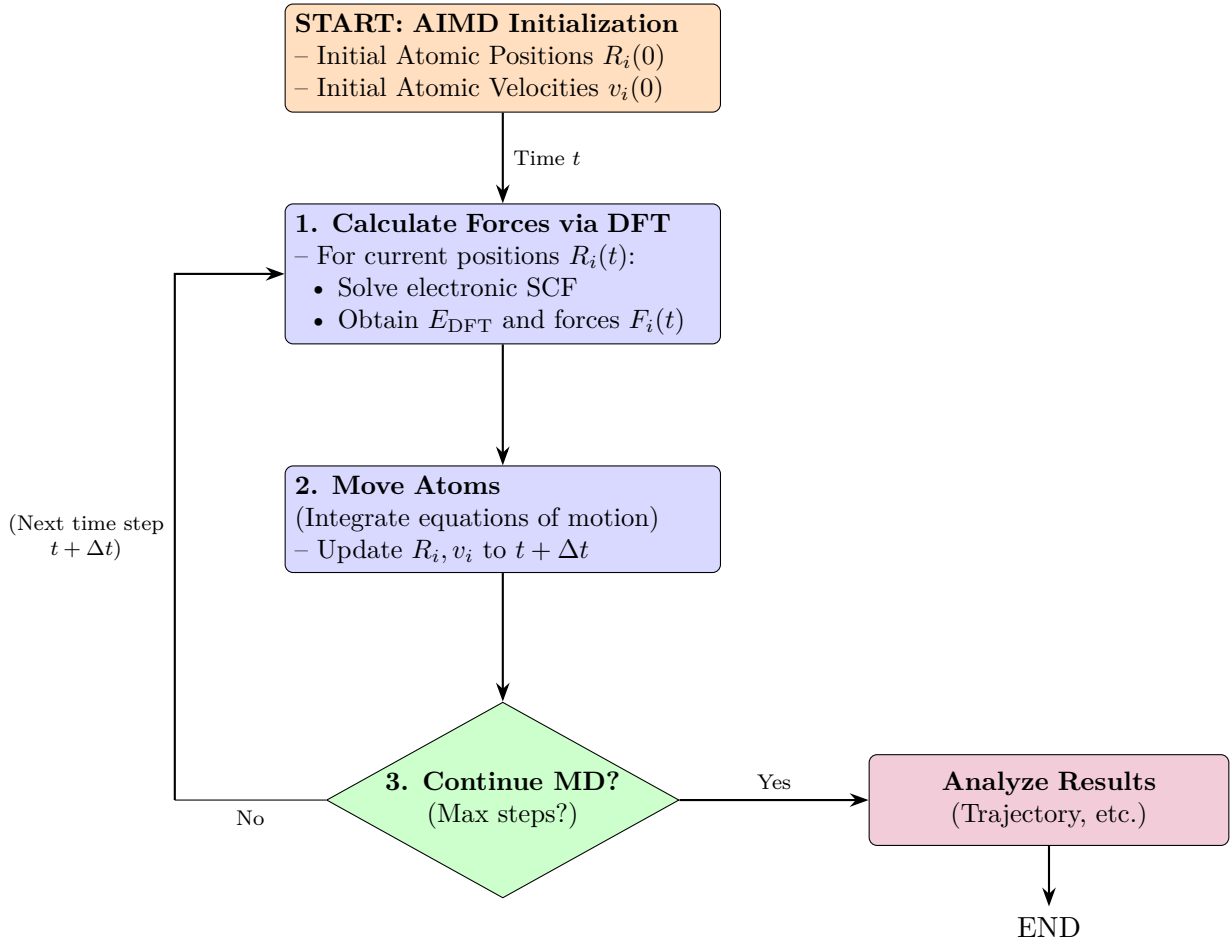


Figure 2.2: Schematic of the AIMD simulation loop. At each time step (Δt), a full Kohn-Sham DFT calculation is performed to obtain the forces on the atoms from first principles. These forces are then used to integrate Newton’s equations of motion to update the atomic positions and velocities. This cycle of force calculation and atomic movement is repeated until the simulation ends.

2.4 Classical molecular dynamics and force fields

Classical MD is a computational technique designed to simulate the time evolution of atomic systems by treating atoms as classical particles with well-defined positions and momenta. In contrast to *ab initio* MD, where electronic degrees of freedom are explicitly considered using quantum mechanics, classical MD relies on force fields to approximate the potential energy surface (PES) without having to recalculate electronic interactions at every time step.

The potential energy surface represents the system’s energy as a function of the nuclear positions R . In force-field-based MD, the total potential energy $V(R)$ is typically expressed as a sum over interactions, often beginning with pair potentials. For a system of N particles, a

common representation is

$$V(R_1, R_2, \dots, R_N) = \sum_{i=1}^N \sum_{j=i+1}^N U_{ij}(r_{ij}), \quad (2.9)$$

where $U_{ij}(r_{ij})$ is the interaction energy between particles i and j and $r_{ij} = \|R_j - R_i\|$ is the interparticle distance. The forces acting on the particles are then derived as the negative gradient of the potential energy with respect to the atomic coordinates,

$$F_i = -\nabla_{R_i} V(R_1, R_2, \dots, R_N). \quad (2.10)$$

These forces, along with the mass m_i of each particle i , are employed in Newton's second equation of motion,

$$m_i \frac{d^2 R_i}{dt^2} = F_i, \quad (2.11)$$

which is integrated over time to generate the atomic trajectories. Numerical integration algorithms, such as the Verlet or velocity Verlet methods, are favored for their stability and energy conserving properties. For example, the velocity Verlet algorithm updates positions and velocities at each time step Δt as follows:

$$R_i(t + \Delta t) = R_i(t) + v_i(t)\Delta t + \frac{F_i(t)}{2m_i}\Delta t^2, \quad (2.12)$$

$$v_i(t + \Delta t) = v_i(t) + \frac{F_i(t) + F_i(t + \Delta t)}{2m_i}\Delta t. \quad (2.13)$$

where $R_i(t)$ and $v_i(t)$ are the position and velocity of particle i at time t . Typical MD simulations consist of L time steps (with L ranging from 10^4 to 10^8) and a time step size of approximately 10^{-15} s, allowing simulations to capture dynamics over intervals up to about 1–10 nanoseconds.

To mimic bulk systems and minimize edge effects, periodic boundary conditions (PBC) are commonly applied. Under PBC, when a particle leaves one side of the simulation box, it re-enters from the opposite side, effectively creating an infinite lattice. In molecular dynamics, different statistical ensembles can be used to control conserved quantities. In the microcanonical ensemble (NVE), the number of particles N , volume V , and total energy E are conserved, and the system evolves without any external temperature or pressure control. In the canonical ensemble (NVT), thermostats such as Nosé–Hoover, Berendsen, or Langevin adjust particle velocities to maintain a target temperature T . In the isothermal–isobaric ensemble (NPT), a thermostat and a barostat (e.g., Parrinello–Rahman or Berendsen) work together to regulate both temperature T and pressure P , allowing the simulation box volume to fluctuate so as to achieve the desired pressure.

Force fields play a central role in classical MD by providing an analytical approximation for the PES. They allow for efficient force computations without resorting to electronic structure calculations at every time step. A wide variety of force fields exist, each tailored to capture specific interactions. Common pair potentials include the Lennard-Jones potential,

$$U_{ij}(r_{ij}) = 4\epsilon \left(\left(\frac{\sigma}{r_{ij}} \right)^{12} - \left(\frac{\sigma}{r_{ij}} \right)^6 \right), \quad (2.14)$$

where $U_{ij}(r_{ij})$ is the interaction energy between particles i and j separated by a distance r_{ij} , ϵ represents the depth of the potential well, and σ is the distance at which the inter-particle potential is zero. This form captures the balance between long-range attractive van der Waals forces, which are proportional to r_{ij}^{-6} , and short-range Pauli repulsion, which is proportional to r_{ij}^{-12} (refer Figure 2.3).

Another common form of pair potential is the Coulomb potential,

$$U_{ij}(r_{ij}) = \frac{q_i q_j}{4\pi\epsilon_0 r_{ij}}, \quad (2.15)$$

where q_i and q_j are the electric charges of particles i and j , and ϵ_0 is the vacuum permittivity. This potential describes the electrostatic interaction between charged particles, which decays inversely with their separation.

Among various other models of force fields, the embedded atom method (EAM)⁴³ is especially prominent for simulating metals and alloys. In EAM, the potential energy is expressed in terms of an electron density and pair interactions. The electron density $\rho(\mathbf{r})$ at a given position $\mathbf{r}$ is obtained as a superposition of contributions from individual atoms:

$$\rho(\mathbf{r}) = \sum_j \rho_j(\mathbf{r}), \quad (2.16)$$

where $\rho_j(\mathbf{r})$ represents the electron density contribution from atom j . The total energy of the system is then given by:

$$E = \sum_i F(\rho_i) + \frac{1}{2} \sum_{i \neq j} \phi(r_{ij}). \quad (2.17)$$

Here, the first summation runs over all atoms i in the system. The second summation runs over all unique pairs of distinct atoms i and j , with the prefactor of $\frac{1}{2}$ correcting for double-counting each interaction. In this equation, $F(\rho_i)$ is the embedding function that represents the energy required to insert atom i into the local electron density ρ_i , and $\phi(r_{ij})$ is the pair potential between atoms i and j . This formulation successfully captures the many-body nature of metallic bonding by linking the energy of an atom to its local environment. However, EAM is inherently limited by its isotropic treatment; since it lacks angular dependence, it is best suited for systems where the bonding is predominantly non-directional.

To address systems where directional bonding is significant, modifications of the standard

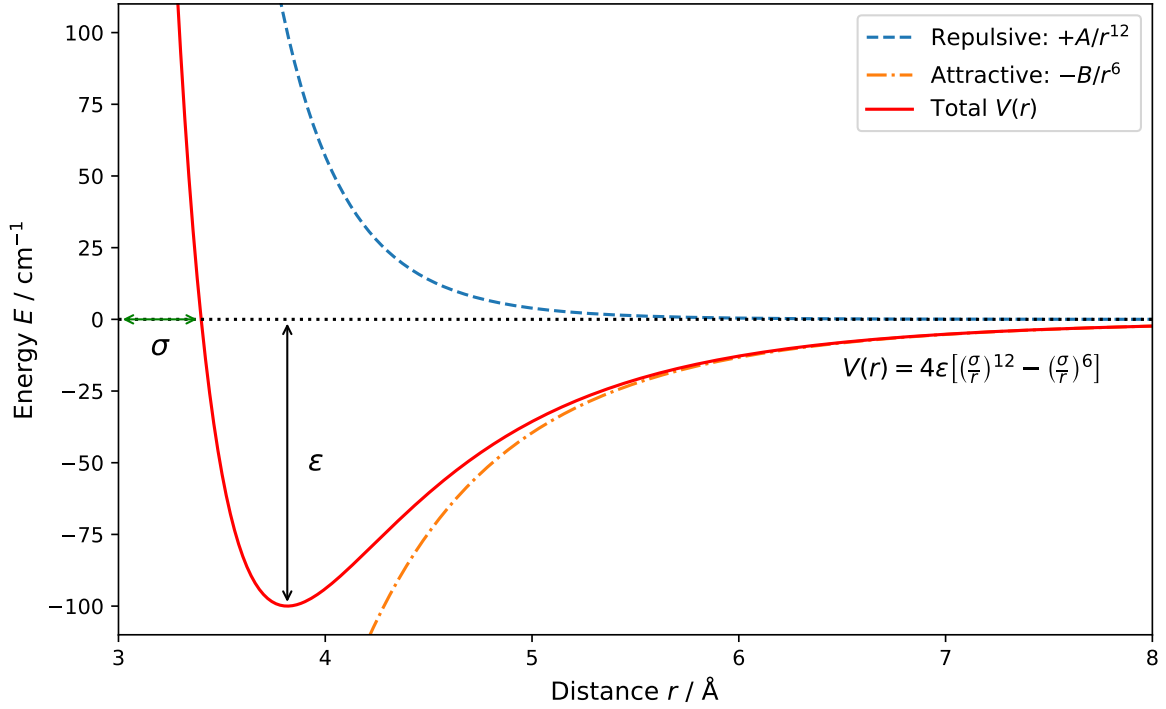


Figure 2.3: Lennard-Jones 12-6 potential energy $E(r) = V(r) = 4\epsilon[(\sigma/r)^{12} - (\sigma/r)^6]$ for argon-like parameters $\epsilon = 100 \text{ cm}^{-1}$ and $\sigma = 3.4 \text{ Å}$. The repulsive $+A/r^{12}$ (blue dashed), attractive $-B/r^6$ (orange dash-dot), and total potential energy (red solid) are plotted as functions of interatomic distance $r = 3.0\text{--}8.0 \text{ Å}$. The vertical double-headed arrow indicates the well depth ϵ , and the horizontal marker at $r = \sigma$ shows the distance where $E(r) = 0$ (collision diameter).

EAM have been developed. For instance, the modified embedded atom method (MEAM)⁴⁴ incorporates angular dependencies by introducing additional terms that capture covalent character and directional bonding. Likewise, the angular embedded atom method⁴⁵ and Tersoff like bond order potentials⁴⁶ such as the analytical bond order potential (ABOP)⁴⁷ modify the interatomic potential further by explicitly including bond angle effects and coordination dependent bond strengths.

In summary, classical MD simulations rely on force fields, which are analytical functions that approximate the potential energy surface. The development of these potentials has progressed from simple pair potentials to more sophisticated many body formalisms like EAM and angular dependent models such as MEAM. Each of these models represents a human designed, physically motivated attempt to capture the essential nature of atomic interactions for a particular class of materials. While computationally efficient, these classical potentials, often referred to as interatomic potentials, share a fundamental limitation. Their accuracy is constrained by the rigidity of their predefined mathematical forms. Capturing the complex,

environment dependent, and quantum mechanical nature of bonding in multicomponent alloys like TiAlNb often proves to be beyond the reach of these fixed functional forms. This challenge, which is the need for a model with the accuracy of quantum mechanics but the efficiency of a classical potential, directly motivates the development of the next generation of data driven models.

2.5 Machine learning based interatomic potentials

As computational capabilities continue to advance, allowing for more precise MD simulations, the development of interatomic potentials stands at a crossroads. One option is to enhance these potentials by incorporating additional physical principles, striving for rigorous derivations from first principles with minimal approximations. Alternatively, a more recent approach leverages basic physical symmetries combined with ML techniques to approximate interatomic interactions. At its core, machine learning is a computational paradigm that constructs a model by learning patterns directly from data, rather than being programmed with explicit, physics-derived rules. In the context of interatomic potentials, this involves training a highly flexible, data-driven model on a large set of labeled examples, where each atomic configuration is labeled with its corresponding quantum mechanical energy and forces. The success of ML-based potentials in the last decade indicates that this latter strategy, while unconventional from a traditional physics viewpoint, holds significant promise for the future of interatomic potential development.

An ML potential can be characterized by the following²²:

- It employs machine learning to directly construct a functional relationship between atomic configurations and their corresponding energies.
- It relies exclusively on the chosen reference electronic structure method, without additional physical approximations.
- It is trained on a consistent dataset derived from electronic structure calculations.

The initial ML-based interatomic potentials appeared in 1995, utilizing artificial neural networks⁴⁸ to model potential energy surfaces. However, ML potentials gained broader attention after the introduction of neural-network potentials by Behler and Parrinello in 2007²⁵, followed by the Gaussian approximation potential (GAP) framework by Bartók and Csányi in 2010²⁴. Since then, various ML potentials have emerged, generally categorized into two groups based on their underlying ML approach: (1) neural networks and (2) other regression techniques, such as kernel-based or linear regression. This thesis primarily focuses on two state-of-the-art frameworks that exemplify these categories: deep potential molecular dynamics (DeePMD), which is based on neural networks, and moment tensor potentials (MTP), which utilize linear regression.

A neural network, the core of the DeePMD model, is a highly flexible, non-linear model inspired by the structure of the human brain. It consists of interconnected “neurons” organized

in layers. The strength of the connection between any two neurons is defined by a numerical parameter called a weight. The collection of all such weights in the network constitutes its trainable parameters. During training, these weights are systematically optimized to minimize the difference between the model’s predictions and the reference DFT data. This optimization is typically performed using stochastic gradient descent, guided by a learning rate, which controls the magnitude of the weight adjustments at each step. To improve training stability, the learning rate is often decayed over time according to a schedule defined by a decay rate and decay steps. The power of neural networks lies in their ability to learn extremely complex and high-dimensional functions without a predefined mathematical form.

The linear regression model, which is the foundation of the MTP framework, is based on a more structured and transparent approach. This method assumes that the energy of an atom can be expressed as a simple weighted sum of a set of pre-defined, physically-motivated basis functions that represent the atomic environment. The model’s only trainable parameters are the linear coefficients (or weights) for each basis function. The training process involves solving a system of linear equations to find the set of coefficients that best reproduces the reference data, a procedure that is typically much faster and more deterministic than the iterative optimization required for neural networks.

In ML-based interatomic potentials, a key approximation assumes that the total energy (E_{tot}) of a system can be expressed as a sum of atomic contributions, each influenced only by its local atomic environment:

$$E_{\text{tot}} = \sum_{i=1}^N E(q_i),$$

where N is the number of atoms, and q_i represents the descriptor for the local atomic environment around atom i . This descriptor effectively captures the spatial configuration surrounding each atom. For an ML potential to be both accurate and robust, it is essential to use a well-crafted descriptor that accurately reflects local atomic geometry. In practice, ML potentials are trained on a dataset of atomic structures, with each structure annotated by quantum-mechanical energies, forces, and optionally, stresses obtained from high-accuracy calculations such as DFT.

In ML applications for interatomic potentials, atomic structures must be encoded as “descriptors” or “features”, which represent the atomic configuration or other relevant system information²³. Descriptors focused on local environments capture information centered on each atom and its neighbors, ranging from simple two- or three-body terms to complex many-body functions. Descriptors must yield identical values for equivalent atomic configurations: those related by translation, rotation, permutation, or symmetry operations²². This ensures the ML model assigns the same energy to each representation of a given structure. Descriptors are typically confined to a local environment within a cutoff radius (usually 5-6 Å). Avoiding artificial symmetries in descriptors is crucial to prevent distinct structures from being mapped identically. Ideally, each structure would correspond uniquely to a specific set of descriptor values, though achieving this would require descriptors per degree of freedom, which is impractical for general ML potentials²². While descriptor transformations need not be invertible,

they should be efficient to compute, as they are recalculated for every configuration, and differentiable to allow analytical gradient calculations for forces. High-accuracy ML potentials require descriptors that capture detailed structural information, often using multidimensional functions to fit complex many-body interactions seen in electronic structure calculations.

Some of the prominent descriptors used include, atom-centered symmetry functions (ASCFs)²⁵, smooth overlap of atomic position (SOAP)⁴⁹, moment tensors^{50,51}, bispectrum⁵². However, this thesis focuses exclusively on the descriptors employed in DeePMD (Section 2.5.1) and MTP (Section 2.5.2).

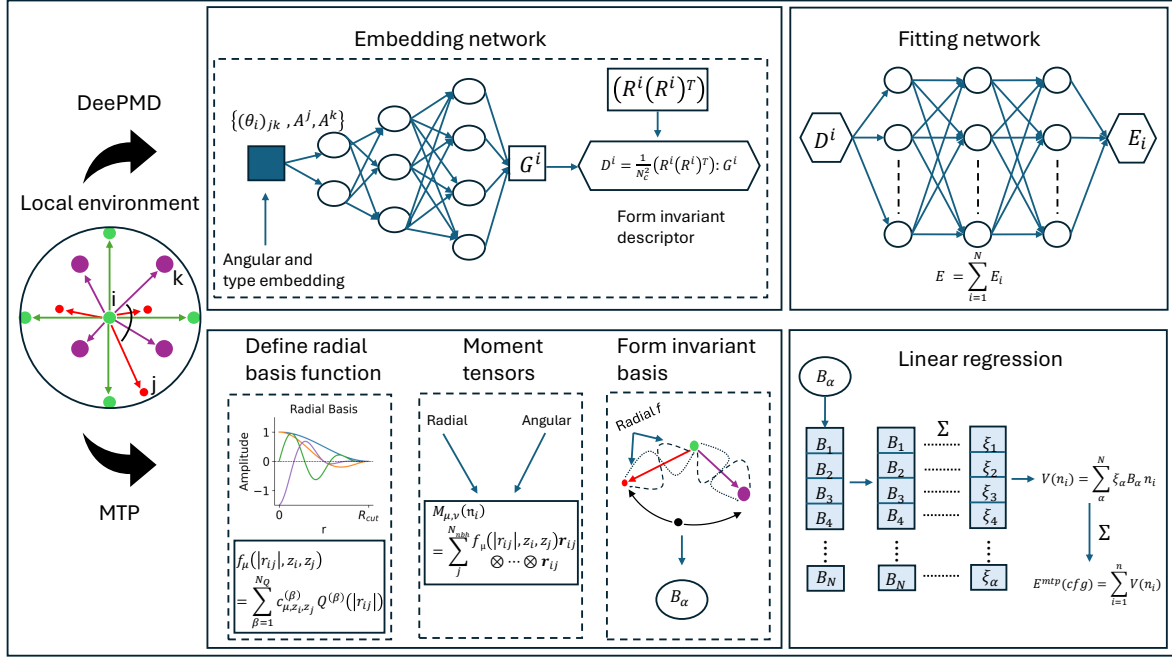


Figure 2.4: Schematic comparison of the architectural workflows for the deep potential molecular dynamics (DeePMD) and moment tensor potential (MTP) frameworks. (Top) The DeePMD model utilizes two neural networks in sequence: an “Embedding network” learns a feature representation which is contracted with a geometric matrix to form an invariant descriptor, D^i , followed by a “Fitting network” that performs a non-linear mapping from this descriptor to the atomic energy, E_i . (Bottom) In contrast, the MTP framework relies on a predefined, non-learnable basis. It systematically constructs a vector of invariant basis functions, B_α , from moment tensors, and the final energy is obtained via a simple linear regression on this basis.

2.5.1 Deep potential molecular dynamics

Deep potential molecular dynamics (DeePMD)⁵³ models provide a bridge between the accuracy of *ab initio* methods and the efficiency of empirical force fields by learning atomic interactions

directly from energies and forces. The framework is founded on the principle of locality, asserting that the energy of an atom i is determined solely by the configuration of its neighbors within a cutoff radius r_c . The relative positions and chemical identities of these neighbors are encoded into a fixed-length vector known as a descriptor, $\mathbf{D}^i$. This descriptor is systematically constructed to be invariant under translation, rotation, and the permutation of identical atoms, thereby ensuring that the learned potential respects fundamental physical symmetries.

As depicted in the final stage of the workflow in Figure 2.4, this invariant descriptor $\mathbf{D}^i$ is then processed by a deep neural network, the fitting network $\mathcal{F}$, to predict a scalar atomic energy E_i given by

$$E_i = \mathcal{F}(\mathbf{D}^i; \theta), \quad (2.18)$$

where θ represents all trainable parameters of the model. The total potential energy E of the system is expressed as the sum of atomic contributions,

$$E = \sum_{i=1}^N E_i, \quad (2.19)$$

with N denoting the total number of atoms in the configuration. Forces and stress tensors are obtained analytically through automatic differentiation of the total energy with respect to atomic coordinates and cell vectors, respectively. This fully differentiable architecture allows the model parameters to be optimized against reference energies, forces, and virials, yielding a potential that is both accurate and physically consistent.

The DeePMD-kit framework⁵³ supports several descriptor formulations, each offering a different balance of computational cost and descriptive power. A widely used and powerful choice is the DeepPot-SE (Smooth Edition)⁵³ descriptor. It addresses the discontinuities present in earlier descriptor models by employing a smooth switching function $s(r)$ that ensures all features decay smoothly to zero at the cutoff radius. The function is defined as:

$$s(r) = \begin{cases} \frac{1}{r} & r < r_s \\ \frac{1}{r} [x^3(-6x^2 + 15x - 10) + 1] & r_s \leq r < r_c \\ 0 & r \geq r_c \end{cases} \quad (2.20)$$

where $x = (r - r_s)/(r_c - r_s)$ is a variable that varies linearly from 0 to 1 between the start of the switching region, r_s , and the cutoff r_c . The DeepPot-SE framework has variants based on the primary geometric information used.

The two-body embedding DeepPot-SE descriptor provides a multi-body representation of the local environment. It is termed “two-body embedding” because the input to its neural network, $\mathcal{N}_{e,2}$, is solely a function of the pairwise distance, $s(r_{ij})$, between the central atom i and each neighbor j . The final descriptor is then constructed by combining the learned output of this network with a geometric matrix $\mathbf{R}^i$ and its transpose. The matrix product $\mathbf{R}^i(\mathbf{R}^i)^T$ inherently encodes angular information about the environment, and by contracting this with the learned features, the resulting descriptor becomes sensitive to multi-body correlations.

To more directly and accurately model these angular dependencies, the three-body embedding DeepPot-SE descriptor was developed. This formulation, which was employed in this work and is detailed in Figure 2.4, modifies the feature-learning process. Instead of learning from distances, the neural network $\mathcal{N}_{e,3}$ learns directly from a representation of the bond angles. For each triplet consisting of a central atom i and a pair of its neighbors (j, k) , the network’s output forms an entry in the embedding matrix $\mathbf{G}^i$:

$$(\mathbf{G}^i)_{jk} = \mathcal{N}_{e,3}((\theta_i)_{jk}) \quad (2.21)$$

where $(\theta_i)_{jk}$ represents the angle formed by the triplet of atoms (i, j, k) with atom i as the vertex.

As illustrated in Figure 2.4, the final invariant descriptor is then formed by contracting the learned embedding tensor $\mathbf{G}^i$ with the geometric matrix product $\mathbf{R}^i(\mathbf{R}^i)^T$:

$$\mathbf{D}^i = \frac{1}{N_c^2} (\mathbf{R}^i(\mathbf{R}^i)^T) : \mathbf{G}^i. \quad (2.22)$$

Here, the prefactor $\frac{1}{N_c^2}$, where N_c is the number of neighbors, serves as a normalization constant that makes the descriptor invariant to the total number of neighbor pairs. The embedding $\mathbf{G}^i$ produced by the neural network is a rank-3 tensor, where each element $(\mathbf{G}^i)_{jk}$ is the feature vector for the neighbor pair (j, k) . The notation “:” in this context indicates a specific tensor contraction between the matrix $\mathbf{R}^i(\mathbf{R}^i)^T$ and the first two dimensions (the neighbor indices j and k) of the tensor $\mathbf{G}^i$. This operation effectively computes a weighted sum over all neighbor pairs to produce the final descriptor vector $\mathbf{D}^i$, elegantly combining the explicit physical geometry with the learned, chemistry-aware angular embedding.

Beyond the DeepPot-SE variants, the DeePMD framework continues to evolve with more advanced techniques. To efficiently model systems with many chemical species, a type-embedding scheme⁵³ is introduced. Rather than training a separate network for each interaction type, a single network is shared, and it is provided with learnable vectors that encode the chemical identity of the interacting atoms. For the three-body descriptor used in this work, the inputs to the $\mathcal{N}_{e,3}$ network are therefore the angle $(\theta_i)_{jk}$ and the type-embedding vectors for atoms j and k .

Further extensions include the attention-based descriptor, which uses a self-attention mechanism to allow the model to dynamically weight the importance of different neighbors, and the hybrid descriptor, which combines multiple descriptor types to leverage their complementary strengths. Additionally, techniques for model compression have been developed to reduce the computational cost during inference by merging and tabulating network operations, which is particularly beneficial for large-scale simulations on hardware like graphics processing units (GPUs). For a comprehensive overview of these advanced features, see Zeng et al.⁵³.

A key advantage of the DeePMD framework is that its end-to-end architecture is fully differentiable. This allows physical observables beyond the total energy to be computed analytically and included in the training process. The force on each atom i is obtained directly as

the negative gradient of the total potential energy with respect to its position:

$$\mathbf{F}_i = -\frac{\partial E}{\partial \mathbf{r}_i}. \quad (2.23)$$

Similarly, the stress tensor Ξ is computed from the derivative of the total energy with respect to the simulation cell. Let $\mathbf{h}$ be a 3×3 matrix whose columns are the lattice vectors $\mathbf{a}_1, \mathbf{a}_2, \mathbf{a}_3$ defining the periodic simulation box:

$$\mathbf{h} = \begin{bmatrix} \mathbf{a}_1 & \mathbf{a}_2 & \mathbf{a}_3 \end{bmatrix}. \quad (2.24)$$

The transpose of this matrix, denoted $\mathbf{h}^\top$, has the same vectors as rows instead of columns. In the DeePMD framework, the stress tensor is then calculated as:

$$\Xi = -\frac{\partial E}{\partial \mathbf{h}} \cdot \mathbf{h}^\top. \quad (2.25)$$

This formulation ensures correct dimensionality and coordinate consistency when transforming the energy derivatives into a Cartesian stress tensor. It also allows the model to maintain physical consistency between energy, forces, and stresses during training.

The training process optimizes the network parameters θ by minimizing a loss function L that quantifies the deviation between the model's predictions and the reference *ab initio* data. The total loss is defined as a sum of the mean square errors in the predicted energy, forces, and virial, weighted by tunable prefactors:

$$L(p_\epsilon, p_f, p_\xi) = \frac{p_\epsilon}{N} \Delta E^2 + \frac{p_f}{3N} \sum_i |\Delta \mathbf{F}_i|^2 + \frac{p_\xi}{9N} \|\Delta \Xi\|^2 \quad (2.26)$$

Here, ΔE , $\Delta \mathbf{F}_i$, and $\Delta \Xi$ denote the root mean square errors (RMSEs) between the predicted and reference values for the energy, forces, and virial, respectively. The prefactors p_ϵ , p_f , and p_ξ control the relative importance of each physical observable in the total loss.

In the DeePMD-kit implementation, these prefactors are often dynamically adjusted during training. A common strategy is to link the prefactor $p(t)$ at a given training step t to the learning rate schedule, allowing the model to prioritize different observables as training progresses. The prefactor varies from a starting value p^{start} to a limit value p^{limit} according to:

$$p(t) = p^{\text{limit}} \left[1 - \frac{r_l(t)}{r_l^0} \right] + p^{\text{start}} \left[\frac{r_l(t)}{r_l^0} \right] \quad (2.27)$$

where $r_l(t)$ is the learning rate at step t and r_l^0 is the initial learning rate.

The learning rate itself typically follows an exponential decay schedule to allow for fine-tuning of the parameters in later stages of training. The learning rate at step t is given by:

$$r_l(t) = r_l^0 \cdot d_r^{t/d_s} \quad (2.28)$$

where d_r is the decay rate (required to be less than 1) and d_s is the number of decay steps. This coupled evolution of the learning rate and loss function prefactors provides a flexible and powerful scheme for optimizing the potential. The model is trained by minimizing the total loss function using stochastic gradient descent, typically with an adaptive optimizer like Adam⁵⁴, to learn a potential energy surface that is both accurate and physically consistent.

2.5.2 Moment tensor potentials

Moment tensor potentials (MTP) are a class of machine-learning interatomic potentials implemented in the MLIP software package⁵¹. The MTP framework expresses the total potential energy of an atomic configuration, *cfg*, as a sum of local energy contributions, where each contribution $V(\eta_i)$ depends on the environment η_i of a central atom i . For a system of N atoms, the total energy is

$$E^{\text{MTP}}(\text{cfg}) = \sum_{i=1}^N V(\eta_i), \quad (2.29)$$

where η_i encapsulates the local environment, including the chemical types and relative positions of neighboring atoms within a cutoff radius. As illustrated in the workflow of Figure 2.4, MTPs are constructed from a set of pre-defined, non-learnable basis functions.

The local energy contribution $V(\eta_i)$ is expressed as a linear combination of these basis functions, $B_\alpha(\eta_i)$:

$$V(\eta_i) = \sum_{\alpha=1}^{N_{\text{lin}}} \xi_\alpha B_\alpha(\eta_i). \quad (2.30)$$

Here, N_{lin} is the total number of basis functions included in the linear expansion, and $\xi = \{\xi_\alpha\}$ are the trainable linear fitting coefficients determined by fitting the model to a reference dataset. The basis functions themselves are built from moment tensor descriptors. A moment tensor M of order (μ, ν) for atom i is defined as a sum over its neighbors:

$$M_{\mu,\nu}(\eta_i) = \sum_{j \in \text{nbh}(i)} f_\mu(|r_{ij}|, z_i, z_j) \underbrace{\mathbf{r}_{ij} \otimes \cdots \otimes \mathbf{r}_{ij}}_{\nu \text{ times}} \quad (2.31)$$

where $\text{nbh}(i)$ is the set of neighbors and f_μ is a radial function expanded in a polynomial basis. The variables z_i and z_j denote the chemical types (atomic species) of the central atom i and its neighbor j , respectively, allowing the radial function to depend explicitly on the element types involved. The expression $\mathbf{r}_{ij} \otimes \cdots \otimes \mathbf{r}_{ij}$ denotes the ν -fold outer product of the relative position vector $\mathbf{r}_{ij}$. This outer product results in a tensor of rank ν ; for example, $\nu = 0$ (the product is omitted, leaving only f_μ) gives a scalar, $\nu = 1$ (the vector $\mathbf{r}_{ij}$ itself) gives a vector, and $\nu = 2$ gives a matrix.

The moment tensors $M_{\mu,\nu}$ are not themselves rotationally invariant (except when their

rank $\nu = 0$). To form the scalar, invariant basis functions B_α , one must combine the moment tensors according to a set of predefined "recipes". These recipes are systematically generated based on invariant theory to form a complete and non-redundant basis set. Each recipe involves taking products of one or more moment tensors and contracting them to a scalar, for example by using the Frobenius inner product, defined as $A : B = \text{tr}(A^T B)$, where $\text{tr}(\cdot)$ denotes the trace of a matrix.

The complexity of the MTP model is systematically controlled by a single hyperparameter, $\text{lev}_{\max}$. This parameter acts as a filter on the predefined list of recipes that form the basis functions. Each moment tensor is first assigned a "complexity score" or level, which is a function of its radial index μ and its angular rank ν :

$$\text{lev}(M_{\mu,\nu}) = 2 + 4\mu + \nu \quad (2.32)$$

Here, the radial index μ relates to the complexity of the radial part of the function, while the angular rank ν specifies the tensorial order of the angular part. The integer coefficients 2, 4, and 1 in this formula were empirically found to be optimal across a number of tests and are fixed within the MLIP package⁵⁵.

The level of a basis function B_α is calculated as the sum of the levels of all the individual moment tensors that are multiplied and contracted to form it. If a basis function is constructed from a combination of P such moment tensors (indexed by p from 1 to P), its total level is given by:

$$\text{lev}(B_\alpha) = \sum_{p=1}^P \text{lev}(M_{\mu_p,\nu_p}) \quad (2.33)$$

Only those basis functions B_α are included for which $\text{lev}(B_\alpha) \leq \text{lev}_{\max}$. For example, with $\text{lev}_{\max} = 6$, the basis includes simple functions such as $B_1 = M_{0,0}$ (with $P = 1$ and $\text{lev} = 2$). It also contains more complex invariants formed by tensor contractions. For instance, B_4 is obtained by contracting the moment tensor $\mathbf{M}_{0,1}$ with itself via the Frobenius inner product, $(\mathbf{M}_{0,1} : \mathbf{M}_{0,1})$. For rank-1 moment tensors like $\mathbf{M}_{0,1}$, this operation is equivalent to the standard dot product. Here, two moment tensors are combined ($P = 2$), and the total level is $\text{lev}(M_{0,1}) + \text{lev}(M_{0,1}) = 3 + 3 = 6$. The full set of these selected invariant functions $\{B_\alpha\}$ forms the final descriptor vector for the environment η_i .

Training an MTP involves determining the optimal set of linear coefficients $\{\xi_\alpha\}$ by solving a weighted linear least-squares problem. The objective is to minimize a loss function over a set of K reference configurations. In the following expression, E_k^{MTP} and E_k^{QM} are the MTP-predicted and quantum-mechanical reference energies for configuration k , respectively. The same notation applies to the forces $\mathbf{f}_{i,k}$ and stress tensors $\boldsymbol{\sigma}_k$. The loss function is:

$$\min_{\{\xi_\alpha\}} \sum_{k=1}^K \left[w_E (E_k^{\text{MTP}} - E_k^{\text{QM}})^2 + w_F \sum_{i=1}^{N_k} \|\mathbf{f}_{i,k}^{\text{MTP}} - \mathbf{f}_{i,k}^{\text{QM}}\|^2 + w_S \|\boldsymbol{\sigma}_k^{\text{MTP}} - \boldsymbol{\sigma}_k^{\text{QM}}\|^2 \right] \quad (2.34)$$

The parameters w_E , w_F , and w_S are user-defined weights that balance the relative importance

of fitting each property.

The performance of the fitted potential is assessed by computing the RMSE for each property on a held-out test set not used during training. This provides an unbiased measure of the model’s predictive accuracy. The RMSEs for energy per atom, forces, and stresses are defined as:

$$\text{RMSE}(E) = \sqrt{\frac{1}{K} \sum_{k=1}^K \left(\frac{E_k^{\text{MTP}} - E_k^{\text{QM}}}{N_k} \right)^2} \quad (2.35)$$

$$\text{RMSE}(F) = \sqrt{\frac{1}{K} \sum_{k=1}^K \frac{1}{3N_k} \sum_{i,\alpha} (f_{i,\alpha,k}^{\text{MTP}} - f_{i,\alpha,k}^{\text{QM}})^2} \quad (2.36)$$

$$\text{RMSE}(\sigma) = \sqrt{\frac{1}{K} \sum_{k=1}^K \frac{1}{9} \sum_{\alpha,\beta} (\sigma_{\alpha\beta,k}^{\text{MTP}} - \sigma_{\alpha\beta,k}^{\text{QM}})^2} \quad (2.37)$$

Here, N_k is the number of atoms in configuration k . The index i runs over atoms, while α and β denote Cartesian components (x, y, z). These metrics are essential for validating the potential and ensuring its robustness.

2.6 Active learning

The generation of a robust training set that covers all relevant regions of the potential energy surface is a critical challenge in developing machine learned interatomic potentials. A traditional approach, often termed passive learning, involves pre-compiling a large and diverse, but ultimately static, dataset of atomic configurations. While straightforward, this method can be inefficient, as it may require a vast number of expensive *ab initio* calculations to ensure all necessary configurations are sampled, many of which may be redundant or irrelevant to the target simulation.

In contrast, active learning provides a more dynamic and efficient paradigm for constructing training sets⁵⁶. As illustrated in the workflow of Figure 2.5, the training set is iteratively and automatically augmented during the course of a molecular dynamics run. The core idea is to use the machine learned potential itself to explore the configuration space and to identify novel configurations where the model is most likely to be inaccurate. This procedure operates as a feedback loop where the simulation proceeds using the current potential, and a metric is evaluated at each step to estimate the model’s error without performing a new quantum mechanical calculation. If this error indicator exceeds a predefined threshold, the configuration is labeled with a high-fidelity *ab initio* model, added to the training set, and the potential is retrained.

The strategy used to estimate the model’s error on-the-fly is known in the field of machine learning as a query strategy⁵⁷. A prominent and widely adopted query strategy for neural network potentials was first implemented by Artrith and Behler, known as query-by-committee⁵⁸.

In this approach, an ensemble of several neural network potentials are trained independently on the same data. During the simulation, the disagreement among the predictions of the committee members is taken as the indicator of the model’s uncertainty. Configurations where the committee members significantly disagree are then selected for addition to the training set. This strategy has become a common choice for many neural network based potentials^{59,60}. Other machine learning frameworks offer alternative query strategies; for instance, Gaussian Process based potentials can naturally use their predictive variance as a built-in uncertainty metric^{61,62}.

The active learning workflow for both MTP and DeePMD consists of two main stages: MD sampling and first-principles labeling. The process is initiated with a very small, limited training set, on which an initial potential is trained. This preliminary potential is then used to run an MD simulation. During this sampling stage, configurations that are poorly described by the current potential are identified using the respective query strategy, either D-optimality for MTP or model deviation for DeePMD. Once a configuration is selected, it proceeds to the labeling stage. A full first-principles calculation, using DFT, is performed to compute its reference energy, forces, and stress tensor. These newly labeled data points are then added to the existing training set. The potential is subsequently retrained on this augmented dataset, resulting in a more accurate and robust model. This iterative cycle of sampling, labeling, and retraining continues until a convergence criterion is met.

The primary advantage of this iterative approach is its remarkable efficiency. By selectively adding only the most informative configurations to the training set, active learning can construct a compact yet comprehensive dataset. This significantly reduces the number of expensive *ab initio* calculations required, often by several orders of magnitude. In this thesis, the specific active learning strategies implemented in the MTP and DeePMD frameworks are detailed below.

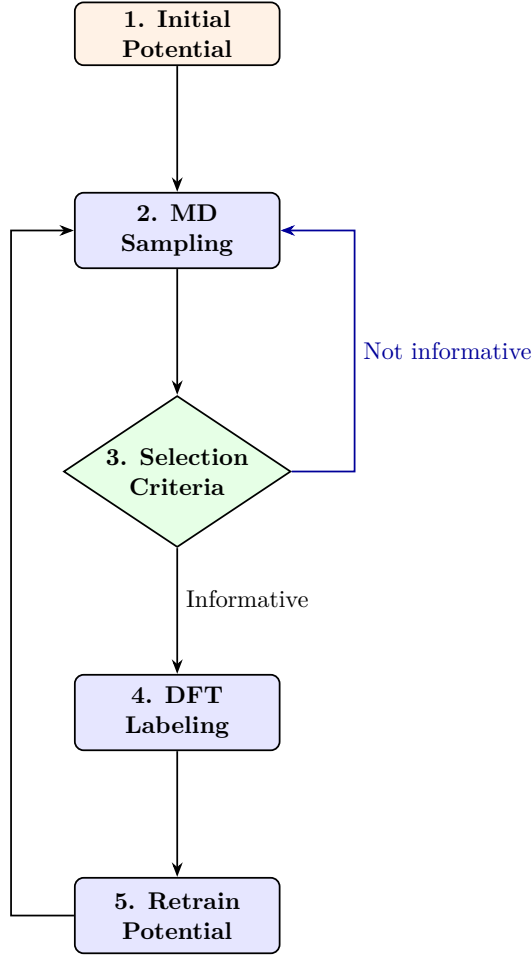


Figure 2.5: Schematic of the active learning cycle. The process begins with an initial potential and is iteratively improved. During MD sampling, a selection criterion identifies novel configurations. If a configuration is deemed informative, it is labeled via DFT and used to retrain the potential. Otherwise, the MD sampling continues.

2.6.1 Active learning for DeePMD

For DeePMD, the active learning strategy is based on the concept of model deviation, $\mathcal{E}$, which employs the query-by-committee methodology⁵⁹. This approach requires maintaining an ensemble of multiple DeePMD potentials, typically four to eight, where each model is trained independently on the same dataset but with different random weight initializations. The disagreement among the predictions of this committee of potentials is then used as a robust indicator of the model’s uncertainty.

The query strategy for DeePMD specifically uses the deviation in atomic forces, rather than total energy or virials, as the primary uncertainty metric. This choice is based on findings

that the maximum model deviation of forces serves as the most effective error indicator for active learning algorithms^{59,63}. A large, erroneous force on a single atom can destabilize a molecular dynamics trajectory, even if its effect on the total energy of the system is negligible.

During a simulation, the force $\mathbf{f}_i^{(k)}$ on each atom i is predicted by every model k in the ensemble. The mean force $\bar{\mathbf{f}}_i$ is calculated as the average over these predictions:

$$\bar{\mathbf{f}}_i = \frac{1}{N_{\text{models}}} \sum_{k=1}^{N_{\text{models}}} \mathbf{f}_i^{(k)}$$

The standard deviation of the force predictions for each atom, σ_i , is then computed as a measure of the committee’s disagreement:

$$\sigma_i = \sqrt{\frac{1}{N_{\text{models}}} \sum_{k=1}^{N_{\text{models}}} \|\mathbf{f}_i^{(k)} - \bar{\mathbf{f}}_i\|^2}$$

The model deviation for the entire configuration, $\mathcal{E}(\text{cfg})$, is defined as the maximum per-atom standard deviation found within the system:

$$\mathcal{E}(\text{cfg}) = \max_i \sigma_i$$

By focusing on the maximum per-atom force deviation, this metric is especially sensitive to the rare but large localized inconsistencies in predicted forces that can cause simulations to fail. The selection criterion is then a simple threshold test. If the model deviation for a configuration exceeds a user-defined tolerance ϵ_{tol} ,

$$\mathcal{E}(\text{cfg}) > \epsilon_{\text{tol}},$$

the configuration is flagged for *ab initio* evaluation. It is subsequently labeled with DFT and added to the training set for retraining the potential ensemble. The value of ϵ_{tol} is chosen to balance the computational cost of labeling new configurations against the desired safety margin for the simulation, with typical values lying in the range of 0.05 to 0.20 eV/Å⁶³.

2.6.2 Active learning for MTP

The active learning strategy for MTP is based on a D-optimality criterion designed to efficiently select the most informative configurations for the training set⁶⁴. This is achieved by calculating a metric for each new configuration known as the extrapolation grade, $\gamma(\text{cfg})$. This scalar value measures how far the configuration lies from the span of the current training set in the descriptor space. A configuration is considered interpolative and its prediction trusted if $\gamma(\text{cfg}) \lesssim 1$, whereas a value of $\gamma(\text{cfg}) > 1$ indicates an extrapolative and potentially unreliable prediction.

In this context, $E^{\text{MTP}}(\text{cfg}_k; \bar{\xi})$ denotes the total energy of configuration k predicted by

the current MTP with parameters $\bar{\xi} = \{\xi_\alpha\}$. The extrapolation grade is derived from the Jacobian matrix, $\mathbf{B}$, which represents the sensitivity of the MTP-predicted energies to changes in the m model parameters, $\{\xi_\alpha\}$. The Jacobian is a $K \times m$ matrix where K is the number of configurations in the current training set. Each element of this matrix, $B_{k\alpha}$, is the partial derivative of the predicted energy for configuration k with respect to the parameter ξ_α :

$$B_{k\alpha} = \frac{\partial E^{\text{MTP}}(\text{cfg}_k; \bar{\xi})}{\partial \xi_\alpha} \quad (2.38)$$

The full Jacobian matrix is then constructed with rows corresponding to the K configurations and columns corresponding to the m parameters:

$$\mathbf{B} = \begin{pmatrix} B_{1,1} & \cdots & B_{1,m} \\ \vdots & \ddots & \vdots \\ B_{K,1} & \cdots & B_{K,m} \end{pmatrix} = \begin{pmatrix} \frac{\partial E^{\text{MTP}}(\text{cfg}_1; \bar{\xi})}{\partial \xi_1} & \cdots & \frac{\partial E^{\text{MTP}}(\text{cfg}_1; \bar{\xi})}{\partial \xi_m} \\ \vdots & \ddots & \vdots \\ \frac{\partial E^{\text{MTP}}(\text{cfg}_K; \bar{\xi})}{\partial \xi_1} & \cdots & \frac{\partial E^{\text{MTP}}(\text{cfg}_K; \bar{\xi})}{\partial \xi_m} \end{pmatrix} \quad (2.39)$$

The underlying principle is the D-optimality criterion, which seeks to find a subset of m configurations from the training set, known as the active set, that are most informative. This corresponds to finding an $m \times m$ submatrix of $\mathbf{B}$ with the maximal determinant.

To efficiently evaluate the novelty of a new configuration, the MaxVol algorithm⁶⁴ is employed. Given the current active submatrix $\mathbf{A}$ (of size $m \times m$) and the Jacobian row $\mathbf{b}$ for a new configuration, one first computes the expansion coefficients $\mathbf{c}$ as $\mathbf{c} = \mathbf{b}\mathbf{A}^{-1}$. The extrapolation grade is then the maximum absolute value of the components of this coefficient vector:

$$\gamma(\text{cfg}) = \max_{1 \leq \alpha \leq m} |c_\alpha| \quad (2.40)$$

If $\gamma(\text{cfg}) > 1$, it indicates that replacing one of the rows in the active set with the new row $\mathbf{b}$ would increase the determinant of the active submatrix, meaning the new configuration is highly informative. It is therefore classified as extrapolative and is selected for *ab initio* labeling.

In practice, a two-threshold scheme is used to manage the active learning workflow. Two thresholds, γ_{select} and γ_{break} , are defined with typical values in the range of $1.1 \lesssim \gamma_{\text{select}} \lesssim 3$ and $3 \lesssim \gamma_{\text{break}} \lesssim 20$, respectively. The following actions are taken based on the calculated grade:

- If $1 < \gamma(\text{cfg}) < \gamma_{\text{select}}$, the configuration triggers an *ab initio* calculation and is added to the training set for future retraining, but the MD simulation continues uninterrupted with the current potential.

- If $\gamma(\text{cfg}) \geq \gamma_{\text{break}}$, the extrapolation is considered too severe to trust the simulation trajectory. The run is terminated, the potential is immediately retrained with the newly added data, and the simulation is restarted with the improved potential.

This two-threshold strategy ensures that the potential is continuously enriched with novel configurations while preventing the simulation from evolving into unphysical states.

Chapter 3

Thermomechanical properties of TiAlNb using Farkas's interatomic potential

3.1 Introduction and motivation

The primary intermetallic phases of TiAl-based alloys with engineering potential are γ -TiAl (ordered face-centered tetragonal, L1₀ prototype, space group P4/mmm) and α_2 -Ti₃Al (ordered hexagonal, D0₁₉ prototype, space group P6₃/mmc). In recent decades, experimental advances have led to the development of new generations of γ -TiAl alloys capable of withstanding higher service temperatures^{1,5,65}. Among these, the TNB (TiAl with Nb and B) and TNM (TiAl with Nb and Mo) alloy systems have demonstrated significant improvements in mechanical properties^{1,5}. These alloys generally follow the stoichiometric formula: Ti-(42–48)Al-(0–10)X-(0–3)Y-(0–1)Z-(0–0.5)RE, where X = Cr, Mn, Nb, Ta; Y = Mo, W, Hf, Zr; Z = C, B, Si; and RE represents rare earth elements. Typically, TNB alloys contain 5–10 at.% Nb with minor additions of B and C, while TNM alloys incorporate similar concentrations of Nb and Mo. In particular, Nb is critical for enhancing the mechanical properties of binary γ -TiAl alloys.

Among various ternary additions, Nb is especially notable for its substantial influence on the mechanical properties of binary γ -TiAl-based alloys^{1,2,5,8}. Experimental studies on TiAlNb systems have examined the combined effects of microstructural boundaries and alloying elements (e.g., Nb, Mo, B, and C) on the TiAl system. However, despite these contributions, the isolated effects of Nb on the strength and ductility of these alloys remain unclear. This gap motivated an investigation into the intrinsic effects of Nb on the thermo-mechanical behavior of γ -TiAl-based alloys using atomistic simulations, aiming to bridge the gap between experimental and modeling studies.

The chemical accuracy and physical reliability of MD simulations depend on the choice of interatomic potentials (or force fields) and appropriate boundary conditions. Farkas's ternary potential²⁹ was developed by combining three binary potentials: one for Nb-Al (fitted for A15 Nb₃Al and DO₂₂ NbAl₃), one for Nb-Ti (for the disordered body-centered cubic (BCC)

phase), and one for Ti-Al⁶⁶. The resulting ternary potential (NbTiAl) has been shown to accurately predict Nb-rich phases such as orthorhombic Ti₂AlNb and the ternary B₂ phase. In the present chapter, this potential is employed to investigate the role of Nb in the thermomechanical properties of TiAl-based alloys.

To comprehensively investigate Nb's contribution, the TiAl atomistic models were classified into three categories: (1) single-phase models, both with and without Nb; (2) single-lamellar interface models, including Nb-rich phases/bulk, Nb-rich interfaces, or homogeneously distributed Nb; and (3) microstructure-informed atomistic models (MIAMs) featuring nanopolycolonies, as illustrated in Fig. 3.1.

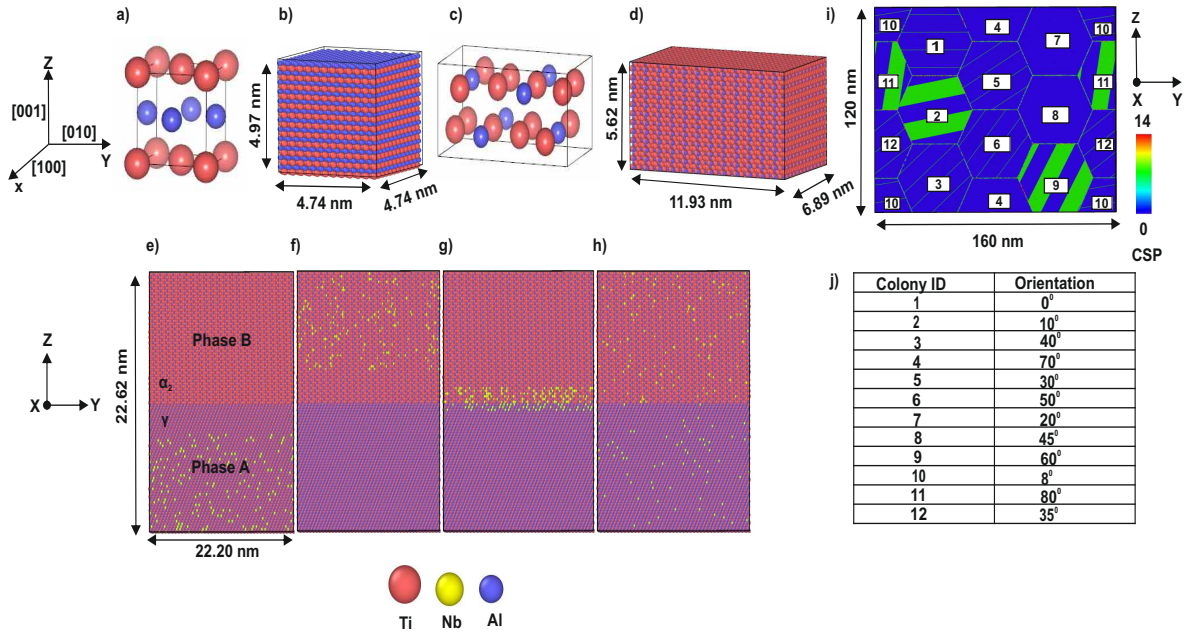


Figure 3.1: Atomistic models used to investigate the TiAlNb system at multiple length scales. (a-d) Bulk single-phase models, showing the unit cells and corresponding supercells for the γ -TiAl and α_2 -Ti₃Al phases. These are used to compute fundamental material properties. (e-h) Single-lamellar interface models containing a single γ/α_2 lamellar interface. These models are used to study the effect of Nb segregation, showing different initial distributions of Nb atoms: (e) in the γ phase, (f) in the α_2 phase, (g) at the interface, and (h) distributed homogeneously. (i, j) A large-scale, microstructure-informed atomistic model (MIAM) containing 12 nano-colonies with orientations specified in the table (j). This model is used to simulate the behavior of a more realistic polycrystalline material. Adapted from Article A.1.

Single-phase models of γ -TiAl (lattice parameters $a = 3.950 \text{ \AA}$, $c/a = 1.048$) and α_2 -Ti₃Al ($a = 5.740 \text{ \AA}$, $c/a = 0.815$) were constructed using the Farkas binary potential⁶⁶ in simulation boxes of dimensions $4.74 \times 4.74 \times 4.97 \text{ nm}^3$ and $6.89 \times 11.93 \times 5.62 \text{ nm}^3$, respectively (refer

to Figure 3.1b and d). The crystallographic orientations of single-phase γ -TiAl and α_2 -Ti₃Al simulation boxes were [100], [010], and [001] in the X, Y, and Z directions, respectively. The primary purpose of these simulation models was to benchmark the transferability of the Farkas ternary potential²⁹ against other existing interatomic potentials like the Zope⁶⁷ and Kim⁶⁸ potentials.

Interface models comprised a single-lamellar γ/α_2 interface and three γ/γ variants, namely, pseudo twin (γ_{PT}), rotational boundary (γ_{RB}), and true twin (γ_{TT}), which were obtained by rotations of 60°, 120°, and 180° about the $\langle 111 \rangle$ axis, respectively (refer to Figure 3.1e–h). All these models had simulation box dimensions of $22.6 \times 22.2 \times 22.4 \text{ nm}^3$ containing 675,840 atoms. In this study, the γ -TiAl phase was modeled with crystallographic orientations of [1-10], [11-2], and [111] along the X-, Y-, and Z-axes, respectively. The α_2 -Ti₃Al phase was similarly modeled with orientations [1-10], [110], and [001] along the X-, Y-, and Z-axes, respectively. All these simulation boxes had Nb introduced at concentrations of 1.0 at.%, 2.0 at.%, 5.0 at.%, 6.5 at.%, and 7.5 at.% to understand the influence of Nb on thermo-mechanical properties.

Finally, MIAMs⁶⁹ comprised 12 nano-sized colonies in a $12 \times 160 \times 120 \text{ nm}^3$ periodic domain (14,612,466 atoms), with each colony containing one of the four interfaces, i.e., γ/α_2 , γ_{PT} , γ_{RB} , and γ_{TT} . MIAMs were constructed using experimental lamellar metrics such as lamellar thickness (λ^γ , λ^{α_2}), α_2 volume fraction (ϕ^{α_2}), and the distance between subsequent α_2 lamellae (d^{α_2}). Figure 3.1i shows the MIAM model with its colonies represented using the centrosymmetry parameter (CSP), and Figure 3.1j shows the colony ID with their corresponding orientation around the X-axis. The CSP is used to measure the local lattice distortion around an atom, which helps in identifying defects, interfaces, and colony boundaries within a simulation model. To study the influence of alloying, Nb atoms were introduced at the appropriate site locations at concentrations of 0.0 at.%, 0.5 at.%, and 1.0 at.%.

Initially, the suitability of the Farkas ternary potential for the relevant binary Ti–Al phases (γ -TiAl and α_2 -Ti₃Al) was assessed by comparing its predictions against those from the established Zope and Kim potentials. This involved computing fundamental properties including single-crystal elastic constants, point-defect formation energies (ΔE_f), cohesive energies (E_{coh}), melting and transition temperatures (via simulated heating curves), and mechanical response under uniaxial tension, where simulated tension tests were performed at a strain rate of 10^9 s^{-1} to evaluate the ultimate tensile strength (UTS), coupled with dislocation analysis using the dislocation extraction algorithm (DXA). The detailed simulation settings for these calculations are provided in Article A.1. Subsequently, DFT calculations were performed on these single phases to probe the intrinsic effects of Nb alloying, specifically examining its influence on lattice parameters, site preference energetics, and local electronic structure through electron localization function (ELF) and partial density of states (PDOS) analyses. The investigation then progressed to single-lamellar interface models, encompassing both γ/α_2 and three distinct γ/γ boundary variants. As mentioned earlier, Nb was introduced into these models at concentrations ranging from 1.0 to 7.5 at.%, utilizing three different spatial distributions: concentrated at the interface, distributed within the bulk phases away from the interface, or homogeneously dispersed. Simulated tension tests and DXA analyses were repeated for these configurations to evaluate the UTS and defect evolution as functions of Nb content and location. These

simulations were supplemented by nanomechanical analyses focused on understanding how Nb alloying affects the microstructural deformation mechanisms. Finally, the study was extended to simulations employing MIAMs. Uniaxial tension tests were conducted MIAMs with (0.5 and 1.0 at.% Nb) and without Nb introduction, to evaluate the impact of Nb on the thermomechanical response and defect evolution within a more realistic lamellar microstructural context. Similar to the interface studies, complementary nanomechanical investigations were employed here to provide detailed insight into the interplay between Nb alloying, thermomechanical loading, and the resulting atomic-scale microstructural changes within the MIAMs.

3.2 Key findings and contribution of Article A.1

- The DFT calculations demonstrated that Nb prefers Ti sites compared to Al sites in γ -TiAl and α_2 -Ti₃Al phases.
- Simulated tension tests on single-phase models, interface models, and MIAMs revealed that strength generally decreases with increasing Nb concentration. In the interface models, concentrating Nb near the interface region resulted in superior strength compared to a homogeneous distribution of Nb across the entire model or within the individual phases A or B (see Figure 3.1).
- Nb alloying led to the early onset of nanomechanical processes, such as dislocation glide and twinning, even at moderate temperatures, thereby enhancing ductility. This enhancement was supported by dislocation density calculations in both MIAMs and interface models, and further verified by DFT calculations on single-phase models, which confirmed that Nb addition increases ductility in both phases. Nanomechanical analysis also revealed evidence of stress-induced $\gamma \rightarrow \alpha_2$ phase transformation in nanocolonies containing twins.

3.3 Results and discussion

This section presents the findings from a series of atomistic simulations designed to systematically investigate the influence of Nb on the thermomechanical properties of TiAl-based alloys using a classical interatomic potential. The investigation follows a hierarchical approach, beginning with the validation of the chosen potential against known binary subsystems and complemented by first-principles calculations for fundamental insights. The study then proceeds to examine mechanical behavior in models of increasing complexity, from idealized single-lamellar interfaces to more realistic, large-scale microstructure-informed models.

3.3.1 Validation of the Farkas potential in binary Ti-Al systems

Before investigating the ternary TiAlNb system, it was essential to first establish the suitability of the chosen Farkas ternary potential for describing the fundamental binary systems. To this

end, a series of validation tests were performed on single-phase γ -TiAl and α_2 -Ti₃Al models. The performance of the Farkas potential was benchmarked against two well-established binary potentials developed by Zope⁶⁷ and Kim⁶⁸.

The initial assessment focused on fundamental energetic and structural properties. A comparison of the predicted elastic constants revealed that the Farkas potential consistently yielded marginally higher values for both phases (refer Table 1 in Article A.1). This slight overestimation can be attributed to two main factors. Firstly, the reference data used to parameterize each potential, whether from experiment or DFT, can have inherent variations that lead to different predictions of material stiffness. Secondly, the Farkas potential was primarily developed for the high Nb-content orthorhombic Ti₂AlNb phase, whereas the Zope and Kim potentials were specifically fitted for the binary TiAl system.

Despite these minor differences in stiffness, a crucial consistency was found in the energetics of point defects. All three potentials agreed that titanium vacancies (V_{Ti}) and antisite at Ti sublattice (Al_{Ti}), where an aluminum atom incorrectly occupies a titanium sublattice site, are the most energetically favorable defect types. This consistency is significant, as this vacancy-antisite mechanism is understood to be the primary driver for atomic diffusion in these phases. Further validation of the thermal properties showed that the Farkas potential uniquely reproduced the correct relative melting behavior, predicting that α_2 -Ti₃Al melts at a lower temperature than γ -TiAl, in agreement with experimental observations⁷⁰.

Finally, the mechanical response was compared through uniaxial tension simulations. For γ -TiAl, the Farkas potential yielded an UTS similar to that of the Zope potential. However, for the α_2 -Ti₃Al phase, the Farkas potential predicted a markedly lower UTS compared to both binary potentials. The deformation mechanisms were also investigated via dislocation analysis. Here, the dislocation evolution predicted by the Farkas potential was found to be in close agreement with the Zope potential for both phases. In contrast, the Kim potential showed different behavior, such as delayed dislocation nucleation, which is likely attributable to its MEAM formalism. Unlike standard EAM, the MEAM formulation includes explicit angular-dependent terms to better describe covalent bonding characteristics. Considering the overall performance, particularly the correct prediction of basic material properties, relative phase stability, and the reasonable agreement on defect energetics, the Farkas ternary potential was deemed sufficiently transferable for use in the subsequent ternary simulations.

3.3.2 First-principles insights into Nb alloying effects

To provide a fundamental, quantum-mechanical understanding of Nb’s role, a series of first-principles DFT calculations were performed on the single-phase γ -TiAl and α_2 -Ti₃Al structures. The first critical question addressed was the preferential site occupancy of Nb atoms within the TiAl lattice. The calculations revealed a strong energetic preference for Nb to substitute on the titanium sublattice sites in both phases. In γ -TiAl, the formation energy for Nb on a Ti site was calculated to be -1.5697 eV, compared to only -0.7908 eV on an Al site. A similar preference was found in α_2 -Ti₃Al, with formation energies of -0.7919 eV (on Ti) versus -0.5915 eV (on Al). This finding is in excellent agreement with numerous previous experimental and theoretical

studies^{71–76}, providing a robust physical basis for the modeling approach. Consequently, in all subsequent classical MD simulations, Nb atoms were exclusively placed on Ti sublattice sites.

Having established where the Nb atoms reside, the investigation then turned to how they alter the electronic structure and bonding. An analysis of the ELF highlighted that the presence of Nb leads to enhanced Nb(d)–Ti(d) and Nb(d)–Al(p) bonding. This finding is consistent with earlier reports showing that 4d transition metal dopants (X), such as Nb, strengthen both X(d)–Ti(d) and X(d)–Al(p) bonds⁷⁷. Further insight was gained from the PDOS, which suggested that Nb may also play a role in improving ductility. The calculations indicated that Nb potentially weakens the primary Ti(3d)–Al(3p) covalent interactions, a mechanism that has been previously proposed to enhance ductility in this class of alloys^{77,78}. Together, these first-principles results provide a clear electronic-level picture: Nb integrates into the Ti sublattice, strengthens the local bonding environment, and simultaneously modifies key electronic interactions in a way that is consistent with its known effect of improving the ductility of TiAl alloys.

3.3.3 Thermomechanical behavior of single-lamellar interfaces

Having established the behavior in bulk single phases, the investigation was extended to a more structurally complex system: a single-lamellar interface model containing both γ -TiAl and α_2 -Ti₃Al phases. This allowed for a systematic exploration of how the spatial distribution of Nb atoms influences the thermomechanical properties of the lamellar structure. To this end, a series of models were constructed with different distributions of Nb atoms: concentrated near the interface, distributed only within the bulk of the phases, or distributed homogeneously throughout the system, as illustrated in Figure 3.1(e-h).

The first key question addressed was whether the location of Nb atoms influenced the mechanical strength. Simulated tension tests on these interface models revealed a consistent and clear trend in the UTS. It was found that configurations with Nb concentrated at the interface exhibited the highest UTS, followed by those with Nb in the bulk phases, while the homogeneously distributed models were the weakest. To understand the atomic-scale origin of this behavior, the deformation was analyzed by tracking atomic shear strain. This analysis revealed that dislocations that nucleated in the α_2 phase and propagated towards the γ phase were obstructed upon reaching the interface region. This observation suggests that the hindering of dislocation propagation by Nb atoms located around the interface may be responsible for the higher UTS observed when Nb is segregated to the interface.

The investigation then turned to the effect of overall Nb concentration. For most configurations, a general trend was observed where the UTS decreased as the total Nb concentration increased. However, this trend showed some irregularities, particularly when Nb was confined to the bulk phases. In contrast to the trend in strength, an analysis of the dislocation density pointed towards a different effect on ductility. It was observed that the dislocation density generally increased at higher Nb concentrations, suggesting that the presence of Nb may enhance the material’s capacity for plastic deformation, although some irregularities in this trend were noted for the homogeneously distributed Nb cases.

During these simulations, a significant limitation of the underlying Farkas potential was identified. While Nb concentrations up to 7.5 at.% could be simulated in the bulk and homogeneous models, the simulations for the Nb-rich interface models were only feasible up to a concentration of 2.0 at.% Nb. Above this threshold, the potential struggled to accurately handle the high local concentration of Nb atoms at the sharp lamellar boundary. This finding is critical as it defines the domain of validity for the potential, which remains applicable for lower concentrations and more uniform distributions but may be less reliable for describing highly segregated Nb states. This limitation, along with the competing trends in strength and ductility, motivated the final stage of the investigation using more complex, realistic microstructures.

3.3.4 Deformation mechanisms in microstructure-informed models

To investigate more realistic microstructures, the study was extended to MIAMs. These large-scale models, alloyed with 0.5 at.% and 1.0 at.% Nb, were subjected to uniaxial tension simulations across a temperature range of 300 K to 900 K. The results from these simulations confirmed the trend observed in the simpler models: the addition of Nb led to a reduction in the UTS.

However, a more complex picture emerged when the deformation mechanisms were analyzed in detail. A qualitative and quantitative analysis of dislocation density revealed a significant increase in dislocation activity within the Nb-alloyed MIAMs, pointing to a key difference in the failure mechanism. This is visualized in the atomic snapshots shown in Figure 3.2. The Nb-free MIAM (Figure 3.2a-d) exhibits significant intergranular fracture along the colony boundaries, especially at lower temperatures. In contrast, the Nb-alloyed MIAMs (Figure 3.2e-l) show a marked reduction in this failure mode across the entire temperature range. This mitigation of intergranular fracture becomes more pronounced with increasing Nb concentration and at elevated temperatures. Instead of failing at the boundaries, the alloyed models exhibit enhanced intra-lamellar dislocation nucleation and glide. This observation suggests that the enhanced intra-lamellar plasticity is the mechanism by which Nb delays macroscopic failure, highlighting that Nb promotes dislocation mobility within the lamellae, hence the ductility, at the expense of overall strength.

In addition to dislocation activity, a competing deformation mechanism was observed in the form of a stress-induced $\gamma \rightarrow \alpha_2$ phase transformation. This is consistent with findings in various previous studies^{79–82}, although it is important to note that this stress-induced α_2 phase has different lattice parameters than the equilibrium ordered hexagonal α_2 (DO₁₉) phase. This transformation was found to be a general feature of the material, occurring even in the Nb-free models, particularly within colonies containing twins (γ/γ_{TT} and γ/γ_{PT}).

A final analysis was conducted to determine how Nb influences this phase transformation. The effect was quantified by tracking the volume fraction of the transformed phase, which has a hexagonal close-packed (HCP) crystal structure. It was observed that the HCP volume fraction decreased in the γ/γ_{RB} and γ/α_2 colonies as Nb concentration increased. In contrast, a reverse trend was observed for the twin-containing γ/γ_{TT} and γ/γ_{PT} colonies, where the transformation was enhanced by the presence of Nb. This complex, colony-dependent behavior

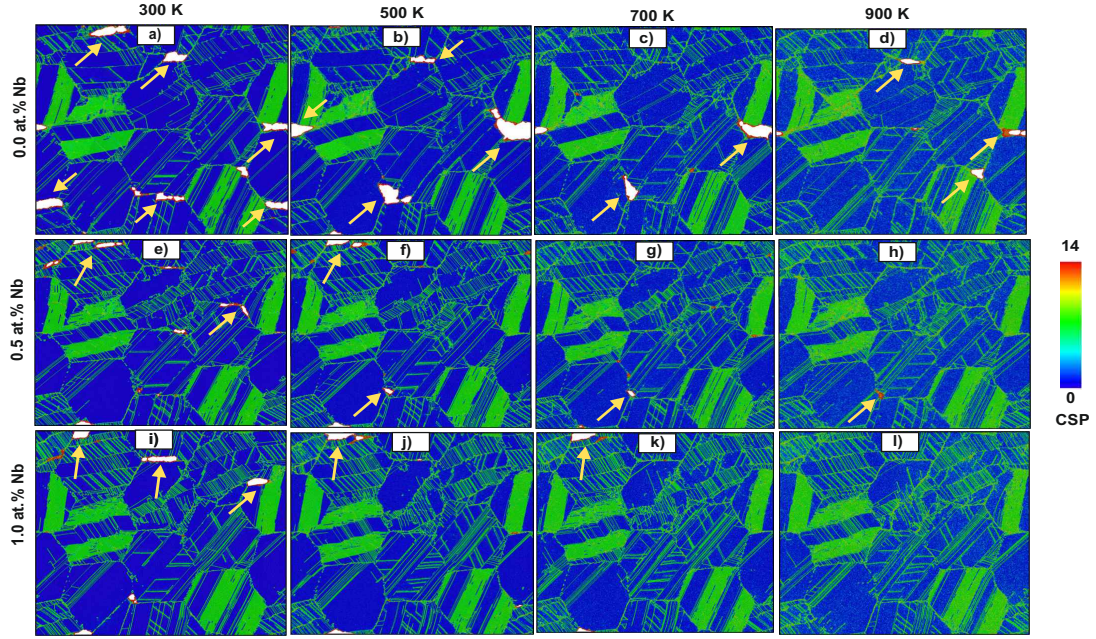


Figure 3.2: MIAMs with Nb concentrations of 0.0 at.% (top), 0.5 at.% (middle), and 1.0 at.% (bottom), deformed at various temperatures after 200 ps. The columns correspond to temperatures of 300 K, 500 K, 700 K, and 900 K, respectively. The snapshots, colored by the centrosymmetry parameter (CSP), show that intergranular fracture (highlighted by yellow arrows) is prevalent in the Nb-free case but is substantially suppressed by the addition of Nb. Adapted from Article A.1.

highlights the nuanced role of alloying elements in governing deformation pathways and is consistent with similar complex dependencies reported previously⁷⁹.

In addition to these mechanistic insights, the simulations also revealed a critical limitation concerning the transferability of the Farkas potential. While the models were stable for Nb concentrations of 0.5 at.% and 1.0 at.%, attempts to simulate systems within the technologically more relevant 5–10 at.% range were unsuccessful due to instabilities in the potential. This finding, consistent with the limitations observed in the single-lamellar interface models, confirms that the Farkas interatomic potential²⁹ is not robust enough for exploring the full range of Nb concentrations required to optimize the alloy's properties.

3.3.5 Summary

Across the hierarchy of models, from single phases to single-lamellar interfaces and finally to complex MIAMs, a consistent physical picture of Nb's influence emerges. The addition of Nb generally reduces the UTS but enhances ductility, primarily by promoting intra-lamellar dislocation activity and influencing stress-induced phase transformation pathways.

However, a recurring and significant limitation was identified throughout this investigation: the inability of the Farkas ternary potential to reliably model the technologically crucial Nb concentration range of 5–10 at.%. The potential exhibited clear transferability issues at these higher concentrations, particularly in the presence of complex microstructures and interfaces. This critical deficiency prevents the use of the existing classical Farkas ternary potential²⁹ to computationally explore and optimize the alloy composition for maximum ductility enhancement, as suggested by experimental findings¹.

Therefore, a crucial next step is the development of a new, more robust interatomic potential specifically designed for the TiAlNb system across a wider range of compositions. The limitations encountered with the current classical Farkas ternary potential²⁹ thus strongly motivate the work presented in the subsequent chapters on the development of advanced machine learning based interatomic potentials.

Chapter 4

Moment tensor vs deep learning: a comparison

4.1 Introduction and motivation

The primary aim of Article A.1 was to elucidate the effect of Nb on the thermo-mechanical properties of TiAl-based alloys, an investigation conducted using Farkas’s ternary interatomic potential²⁹. A critical finding, however, was the significant limitations of this potential. The intended focus was the technologically relevant Nb concentration range of 5 at.% to 10 at.%. Unfortunately, the Farkas potential proved inadequate above 1 at.% Nb in MIAMs and above 2 at.% Nb in specific single-lamellar interface scenarios. This inability to accurately model higher Nb concentrations motivated the development of new interatomic potentials for TiAlNb, employing machine learning methodologies to optimize for the desired 5 at.% to 10 at.% Nb window.

In recent years, the integration of machine learning with molecular dynamics simulations has emerged as a rapidly evolving field, primarily focusing on modeling interatomic PES using reference data from *ab initio* simulations. Various machine learning methodologies have significantly contributed to the study of condensed matter systems. These techniques include linear regression methods (e.g., moment tensor potentials (MTP)^{50,55,83}), kernel methods (e.g., Gaussian approximation potential^{49,84,85} and spectral neighbor analysis^{52,86–88}), and deep neural network-based approaches^{25,89,90}.

From these options, two machine learning interatomic potential methods were selected for detailed investigation: deep potential (DP) and MTP. This decision was based on their demonstrated strengths in modeling both ordered and disordered systems. Specifically, DP’s highly flexible descriptors make it particularly suitable for accurately representing systems with diverse atomic environments and varying concentrations. MTP stands out for its superior computational efficiency relative to many alternatives, while still offering comparable accuracy in physical system modeling, thus providing an excellent trade-off between computational cost and predictive power.

Article A.2 presents a comparative study of these two approaches (MTP and DeePMD, an

implementation of DP) for TiAlNb alloys. In this work, the potentials were evaluated through thorough error analysis and performance metrics. Their ability to predict material properties such as elastic constants, equilibrium volume, and lattice constants was also assessed, along with the influence of Nb on thermo-mechanical properties. The goal of this comprehensive comparison was to identify the scenarios where each model performs optimally, thereby serving as a guideline for readers in choosing between these potentials for their own use cases.

With this objective, a comprehensive dataset was curated to cover a wide range of Nb concentrations in γ -TiAl and α_2 -Ti₃Al alloys, from 1 at.% to 14 at.%. The dataset features 28 distinct structural configurations of two types: γ -TiAl structures containing 108 atoms and α_2 -Ti₃Al structures containing 128 atoms. The primary focus was on Nb concentrations between 1 at.% and 10 at.%; however, the dataset was extended up to 14 at.% Nb to improve the potential training process. Nb atoms were introduced into titanium lattice sites in both phases, with careful attention to their preferential site occupancy based on Article A.1 and other studies^{72,75,76,91,92}. AIMD simulations were then conducted on these structures using both NVT and NPT ensembles for nearly 10 ps; detailed simulation settings for these runs are discussed in Article A.1. To effectively analyze thermal behavior, simulations were performed at temperatures of 1 K, 300 K, 500 K, 700 K, and 900 K. In the NVT ensemble, frames were carefully selected after sufficient equilibration for analysis. The collection of snapshots from these AIMD simulations created a varied dataset suitable for training both the MTP and DeePMD potentials. This dataset was then split, with 80% used for training and 20% reserved for testing. The TiAlNb dataset generated in the course of this research has been made publicly available⁹³.

Using these datasets, a comparative analysis of error metrics and performance was conducted for both MTP and DeePMD interatomic potentials. The study began with a convergence test for MTP to determine the optimal set of parameters for training. Specifically, key MTP settings were systematically varied: the cutoff radius (r_{cut}), which defines the range of atomic interactions considered, and the MTP levels, which determine the complexity of the basis functions. During this process, the RMSE for both energy and force predictions was monitored as these parameters were adjusted. Based on these observations, an appropriate cutoff radius and MTP level were selected for training; the specific training parameters for MTP are detailed in Article A.2. Similarly, the training parameters for DeePMD are also provided in Article A.2.

Following the convergence tests, an error analysis was performed by comparing the errors from the training and test data for both MTP and DeePMD. Once the error profiles had been established, molecular dynamics simulations were carried out using the developed potentials. The study was further extended by computing various material properties, such as elastic constants, equilibrium volume, and lattice constants. Furthermore, the study explored finite-temperature properties, specifically specific heat and thermal expansion, using both potentials. The analysis also included evaluating the radial distribution functions (RDFs) to assess the ability of the potentials to capture atomic arrangements at finite temperature. Mechanical behaviors, such as uniaxial tension and the calculation of generalized stacking fault energy (GSFE), were investigated to determine the impact of Nb alloying on TiAl-based alloys.

4.2 Key findings and contribution of Article A.2

- The DeePMD potential yielded lower error values for forces and energies compared to the MTP. However, DeePMD required a significantly larger training sample size. Moreover, the computational cost of MTP is lower than that of DeePMD, making MTP a better choice when computational resources and sample sizes are limited; however, this involves some compromise on accuracy. Conversely, if high accuracy is of high importance and computational resources and large sample sizes are available, DeePMD is the preferred option.
- Both the MTP and DeePMD potentials performed well in predicting fundamental material properties such as elastic constants, lattice parameters, and equilibrium volume. Although there were occasional outliers, the predicted values were sufficiently close to the AIMD reference values. Additionally, both potentials demonstrated reliable performance in finite-temperature property analyses, making them suitable for MD simulations at elevated temperatures.
- Overall, both potentials effectively captured the impact of Nb addition, showing a decrease in strength and an increase in ductility of TiAl alloys, despite some minor discrepancies.

4.3 Results and discussion

This section details the development and rigorous comparative validation of MTP and DeePMD potentials for the TiAlNb system. A schematic overview of the complete workflow, from dataset generation to the final analysis, is presented in Figure 4.1. The investigation systematically evaluates the trade-offs between the two frameworks in terms of accuracy, computational cost, and data requirements.

4.3.1 Model parameterization and initial error analysis

The development process for the MTP potential began with a crucial first step: a convergence test to determine the optimal model hyperparameters for the TiAlNb system. To this end, 1800 samples were used in a grid search that varied the MTP level from 6 to 24 and the r_{cut} from 5 Å to 8 Å in increments of 1 Å. It was observed that the energy per atom (energy/atom) began converging at level 16, with a similar trend noted for the RMSE of the forces, affirming the reliability of the potential from this level onward. Based on these findings, a configuration of level 18 and a cutoff radius of 7 Å was selected to ensure robustness while maintaining computational efficiency.

Following the training of both potentials, a detailed error analysis was conducted. The MTP model was trained on 40,000 samples, while the DeePMD model was trained on 333,340 samples. The DeePMD model exhibited lower RMSEs, with an energy error of 1.1×10^{-3} eV/atom and force component errors of 7.83×10^{-2} eV/Å (f_x), 7.95×10^{-2} eV/Å (f_y), and

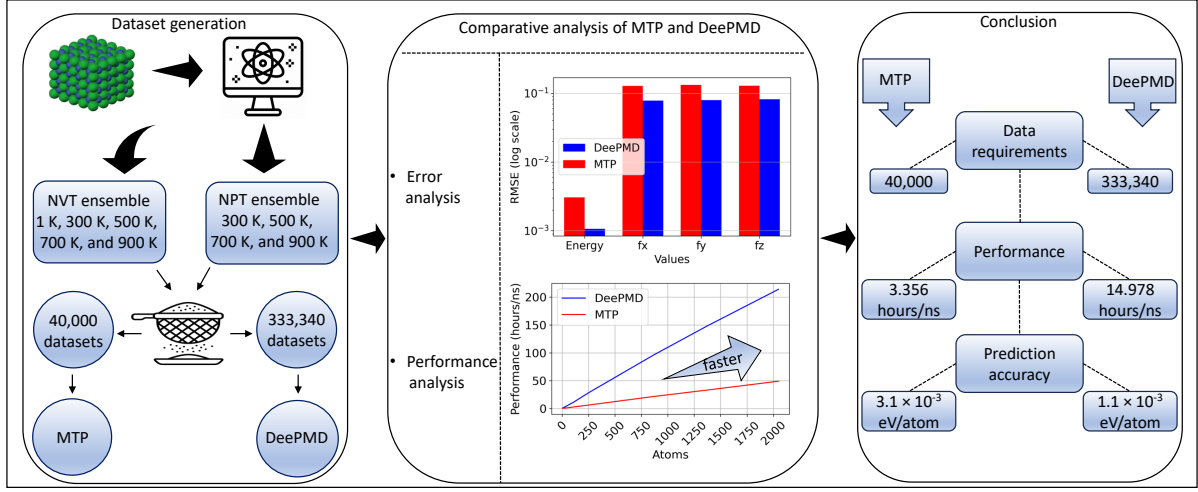


Figure 4.1: Comparative analysis of the MTP and DeePMD frameworks for TiAlNb interatomic potentials. The center panel presents a direct comparison of the models’ predictive accuracy (RMSE) and computational performance (simulation speed). The right panel summarizes the overall findings, illustrating the fundamental trade-offs between the two methods regarding their training data requirements, simulation speed, and final accuracy. The results indicate that while DeePMD achieves higher accuracy, MTP offers significantly better computational performance and requires a smaller training dataset. Adapted from Article A.2.

8.20×10^{-2} eV/Å (f_z). In comparison, the MTP model yielded an energy RMSE of 3.1×10^{-3} eV/atom and force component errors of 1.285×10^{-1} eV/Å (f_x), 1.329×10^{-1} eV/Å (f_y), and 1.292×10^{-1} eV/Å (f_z).

In addition to these overall RMSE values, the predictive quality was assessed visually using parity plots. For both the MTP and DeePMD potentials, the predicted energies and forces showed a strong linear correlation with the reference DFT values, with data points closely aligned along the ideal $y = x$ line (where predicted equals reference). This alignment confirms the absence of systematic bias and demonstrates the general capacity of both potentials to accurately reproduce the reference data across the tested temperature range.

4.3.2 Prediction of equilibrium, elastic and thermal properties

With the models trained and their initial errors quantified, the investigation proceeded to validate their ability to predict fundamental material properties. Energy–volume curves were

first computed for the γ -TiAl and α_2 -Ti₃Al phases to determine equilibrium properties. For the γ -TiAl phase, the lattice parameters predicted by MTP were found to be in closer agreement with the DFT reference values than those predicted by DeePMD. In contrast, for the α_2 -Ti₃Al phase, both the MTP and DeePMD potentials yielded predictions in close agreement with the DFT values, with comparable accuracy between the two models. The elastic constants of both phases, which describe a material’s stiffness and response to mechanical strain, were also calculated (see Table 2 in Article A.2). These constants, denoted C_{ij} , are critical for accurately modeling mechanical behavior, and agreement with reference DFT values is a key indicator of a potential’s quality. For γ -TiAl, the results were mixed: MTP’s predictions for C_{13} and C_{66} were more accurate, whereas DeePMD provided better accuracy for C_{12} and C_{44} . Both potentials predicted identical values for C_{11} , which were also in good agreement with DFT. For the more complex α_2 -Ti₃Al phase, the predictions from both models were less accurate overall, though they provided reasonable estimates for elastic constants.

To evaluate the ability of the models to capture correct atomic arrangements at finite temperatures, RDFs were compared with AIMD curves. Both potentials accurately reproduced the RDFs for the γ -TiAl phase across all tested temperatures (300 to 900 K). However, capturing the RDFs for the α_2 -Ti₃Al phase proved more challenging for both models, especially at elevated temperatures.

Finally, thermal properties including the specific heat capacity at constant volume (C_V) and the thermal expansion coefficients for the lattice parameters a and c (denoted α_a and α_c , respectively) were computed. The specific heat capacity was observed to increase with temperature for all methods. For the α_2 -Ti₃Al phase, the C_V results from DeePMD closely matched the DFT outcomes. In contrast, MTP showed minor differences at lower temperatures but demonstrated better agreement as temperatures increased. For the γ -TiAl phase, while the C_V predictions from both models initially agreed well, slight differences appeared at higher temperatures. Furthermore, the thermal expansion coefficients, α_a and α_c , as predicted by both models, generally increased with temperature before stabilizing. In these calculations, DeePMD’s predictions were closer to the reference values, while MTP tended to slightly underestimate the coefficients for both phases. It is important to note that low-temperature behaviors, often influenced by quantum effects, are not typically well captured by machine learned interatomic potentials.

4.3.3 Alloying effects on mechanical behavior

The central part of the investigation focused on how Nb alloying influences thermomechanical properties. This was assessed by examining the UTS from simulated stress-strain curves and the GSFE, which describe the energy barriers associated with the slip of atomic planes, a fundamental process of plastic deformation. In the simulated tensile tests, both potentials predicted a decrease in UTS with increasing Nb concentration in both the γ -TiAl and α_2 -Ti₃Al phases, with the notable exception of an outlier in the DeePMD results for γ -TiAl.

To understand the atomic-scale origin of this mechanical behavior, GSFEs were computed for both phases and benchmarked against reference values from DFT and other MD simula-

tions^{94–99}. The validation revealed a nuanced performance for both models. For the γ -TiAl phase, MTP’s predictions for both the complex stacking fault (CSF) and the superlattice intrinsic stacking fault (SISF) were in good agreement with the MD reference values, whereas DeePMD’s prediction for the SISF showed better agreement with both the DFT and MD reference data. For the α_2 -Ti₃Al phase, the strengths were distributed differently: MTP provided a closer prediction for the SISF with respect to the reference DFT value, whereas DeePMD’s CSF prediction was nearer to both the DFT and MD references.

Crucially, the GSFE values for most cases were found to decrease with increasing Nb concentration. Since a lower GSFE indicates a reduced energy barrier for dislocation slip, this trend is generally associated with an enhancement in ductility, while concurrently leading to a decrease in the material’s overall strength. This provides a clear mechanistic explanation for the known role of Nb in TiAl alloys, and the overall findings are consistent with previous reports¹⁰⁰ as well as those presented in Article A.1. A noteworthy exception was the anomalous increasing UTS trend observed for γ -TiAl with the DeePMD potential, which also correlated with a corresponding anomalous trend in its predicted GSFE values, demonstrating the internal consistency of that particular model.

4.3.4 Computational efficiency

A final, critical comparison was made of the computational efficiency of the two frameworks in conducting MD simulations. The results indicate that the MTP potential is approximately 4.5 times more computationally efficient than the DeePMD potential. This performance advantage is particularly noteworthy given that the MTP was trained on a significantly smaller dataset of 40,000 samples, compared to the 333,340 samples required for the DeePMD model.

4.3.5 Summary

This comparative analysis of MTP and DeePMD for the TiAlNb system reveals a clear trade-off between predictive accuracy and computational resource requirements. The DeePMD framework, leveraging a deep neural network and a large training set, consistently demonstrated higher fidelity in predicting energies, forces, and a range of material properties. It is therefore the recommended choice for applications where the highest accuracy is paramount and sufficient computational and data generation resources are available. Conversely, the MTP framework offers a compelling and viable alternative. Its strong computational performance and significantly lower data dependency make it a highly practical choice for exploratory studies or for large-scale simulations where a slight compromise in accuracy is acceptable in exchange for a substantial gain in speed.

However, a recurring challenge was identified throughout this work: both models struggled to accurately capture all the properties of the α_2 -Ti₃Al phase, highlighting its complex nature. Furthermore, the substantial dataset sizes required even for this passive learning approach (the traditional method of generating a large, static dataset before training begins) underscore a major bottleneck in the potential development workflow. These challenges directly motivate

the investigation presented in the next chapter, which explores the use of advanced active learning techniques to significantly improve model accuracy while simultaneously reducing the reliance on extensive datasets.

Chapter 5

Improvement through active learning

5.1 Introduction and motivation

The investigation presented in Article A.2 demonstrated that while both MTP and DeePMD frameworks can produce accurate interatomic potentials for the TiAlNb system, a clear trade-off exists. Specifically, the DeePMD model achieved higher accuracy but required a very large training set of 333,340 configurations, whereas the MTP model performed reasonably well with a much smaller set of 40,000 configurations. This significant data requirement, a common challenge for ML-IAPs, motivates the central investigation of Article A.3. In this work, the implementation of an active learning strategy is explored with the goal of developing potentials that can either improve upon the previous accuracy or attain equivalent performance with a substantially smaller and more efficiently generated training set.

Active learning is an advanced training paradigm that begins with a small initial dataset and iteratively improves the model by intelligently selecting only the most informative new data points to add to the training set. As detailed in Section 2.6, this “on-the-fly” approach can produce high-quality interatomic potentials without the need for massive, pre-computed datasets. The workflow consists of two main phases: sampling and labeling. During the sampling phase, MD simulations are run using an initial potential (trained on a limited sample size) to explore the configuration space. Configurations where the model fails to make accurate predictions are identified by a query strategy. In the labeling phase, these selected configurations are accurately annotated using DFT simulations, where their energy and force values are calculated. This new data is used to retrain and refine the potential. This strategy drastically reduces the computational expense of *ab initio* data generation and has been successfully implemented for a wide range of material systems using both the MTP^{55,64,101–106} and DeePMD frameworks^{107–115}.

The methodology for this study began with the generation of a suitable starting dataset for the active learning process. The initial structures used were the same 28 unique structural configurations as in Article A.2, created for both γ -TiAl (108 atoms) and α_2 -Ti₃Al (128 atoms)

with Nb concentrations spanning from 1 to 14 at.%. Short AIMD simulations were then conducted at 300 K using the NVT ensemble for 2.5 ps to provide the initial training data. The complete training and testing datasets generated through this active learning study have been made publicly available¹¹⁶. For the comparative analysis, the passive learning results are based on the datasets developed and published in the work presented in the previous chapter⁹³.

To assess the effectiveness of the active learning strategy, a two-type comparison with passive learning was performed. Comparison 1 was designed to isolate the impact of the learning strategy by evaluating the performance of potentials trained on an identical number of configurations. This was further subdivided into two scenarios: one using only 300 K training data, and a second using multi-temperature data to best match the initial conditions of the active learning runs. Comparison 2 was designed to assess data efficiency by allowing the dataset sizes to vary, with the goal of determining the number of configurations required by each approach to achieve a comparable RMSE.

During the investigation, it was found that a single DeePMD potential could be successfully trained to describe both the γ -TiAl and α_2 -Ti₃Al phases simultaneously. In contrast, for MTP, attempting to train a single active learning model for both phases yielded unsatisfactory results, necessitating a separate procedure for each phase. The validation of the final potentials was conducted in a sequence of increasing complexity. First, a detailed error analysis was performed, comparing the energy/atom and force RMSEs for the active and passive learning cases under both Comparison 1 and Comparison 2 scenarios. Subsequently, the potentials were used in MD simulations to predict fundamental material properties, including equilibrium volume, lattice parameters, and elastic constants. The ability of the potentials to capture finite-temperature atomic arrangements was then assessed by comparing their predicted RDFs to AIMD data at various temperatures (300 K, 500 K, 700 K, and 900 K). Finally, the influence of Nb on the thermomechanical properties was evaluated by computing stress-strain curves and generalized stacking fault energies.

5.2 Key findings and contribution of Article A.3

- The active learning strategy was demonstrated to be significantly more data-efficient than the passive approach. When models were trained to achieve comparable RMSE values, active learning required substantially fewer samples. For MTP, only 3,322 samples for γ -TiAl and 2,602 for α_2 -Ti₃Al were needed, compared to the 20,000 samples required by the passive approach. Similarly, for DeePMD, the active learning potential required 61,657 samples to reach a similar accuracy level as a passive potential trained on 127,212 samples.
- The active learning potentials consistently outperformed their passively trained counterparts in terms of predictive accuracy. The RMSE of energy/atom for the active learning cases was generally lower; in instances where the energy/atom RMSE of an active learning model was marginally higher than a passive one, the corresponding force RMSE for the active model was found to be lower, indicating a more robust overall fit.

- The developed active learning potentials were generally successful in predicting equilibrium properties like equilibrium volume and lattice parameters, often outperforming the passively trained models. However, some discrepancies were observed in the predicted elastic constants when compared with the DFT results. This is likely because these fundamental properties were not the primary focus during the active learning sample selection process, which can result in a bias toward higher-temperature configurations that differ from the 0 K base state.
- The actively trained potentials successfully reproduced the core scientific conclusions of the previous studies. A thermomechanical analysis confirmed that the introduction of Nb increases ductility while reducing strength in both the γ -TiAl and α_2 -Ti₃Al phases, a finding consistent with the results reported in Articles A.1 and A.2. This demonstrates that the data-efficient active learning strategy produces physically reliable potentials.

5.3 Results and discussion

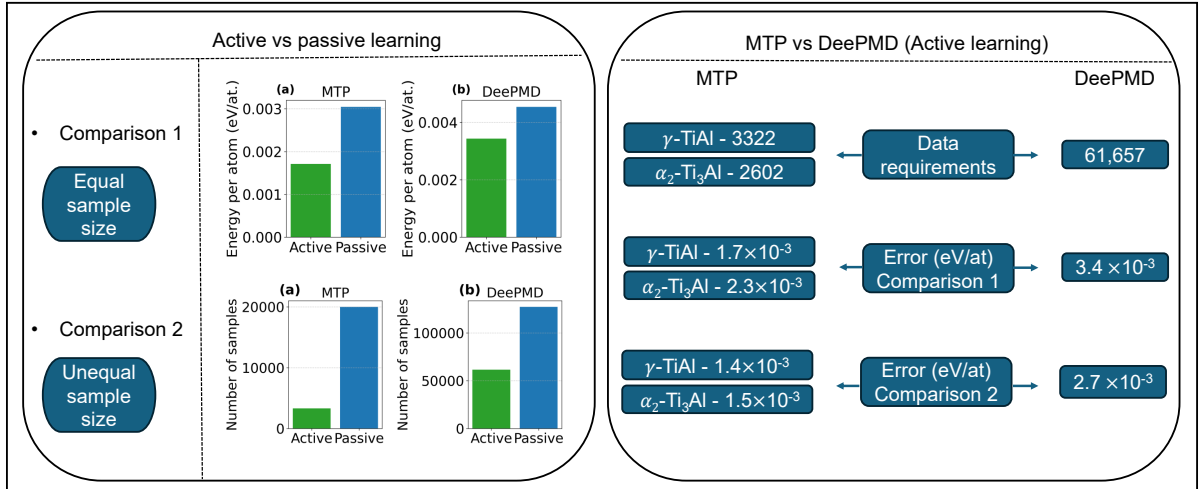


Figure 5.1: Overview of the comparative study of MTP and DeePMD potentials developed via active and passive learning. The analysis is performed at two levels: In Comparison 1, potentials are trained on datasets of equal size to directly compare performance. In Comparison 2, the dataset sizes are allowed to differ to achieve a comparable level of accuracy, thereby evaluating the data efficiency of each learning strategy. The final panel summarizes the key trade-offs for the converged, actively-trained MTP and DeePMD models in terms of their data requirements and predictive accuracy. Adapted from Article A.3.

This section presents a detailed comparative analysis of active versus passive learning

strategies for developing MTP and DeePMD potentials. Figure 5.1 provides a graphical summary of the results from this comparison, evaluating the active and passive learning strategies in terms of their final error metrics and training data requirements for both the MTP and DeePMD frameworks. The following subsections detail this analysis, beginning with a direct comparison of the errors and data efficiency of the two learning strategies. The discussion then proceeds to a comprehensive validation of the potentials’ ability to predict a range of fundamental material and thermomechanical properties.

5.3.1 Data requirements and error analysis: Active versus passive learning

The initial investigation focused on quantifying the effectiveness of the active learning strategy by directly comparing the errors of predicted energy/atom and force values with the passive learning approach. As introduced previously, this analysis was structured into two types of comparisons. Comparison 1 assessed performance on datasets of equal size, while Comparison 2 evaluated the number of samples required by each method to achieve comparable accuracy. It is also important to reiterate a key methodological finding: while a single DeePMD potential could be trained for both phases, the MTP framework required a separate active learning procedure for the γ -TiAl and α_2 -Ti₃Al phases to achieve satisfactory results.

The analysis for the MTP framework revealed the clear benefits of active learning. For the γ -TiAl phase under Comparison 1, where the same number of samples (3,094) was used, the actively trained MTP yielded a lower RMSE for energy/atom than the passive learning cases using either samples exclusively at 300 K or samples spanning multiple temperatures. In contrast, the RMSE for forces in the active learning case was slightly higher than that of the 300 K passive case, yet lower than that of the multi-temperature passive case; the increased RMSE in the latter is attributed to thermodynamic noise from high-temperature frames. In Comparison 2 for γ -TiAl, an actively trained model with 3,322 samples was benchmarked against a passive model trained on a larger dataset of 20,000 samples. The active learning model produced a lower RMSE for energy/atom, although its RMSE for forces was slightly higher.

For the α_2 -Ti₃Al phase under Comparison 1 (1,665 samples), the RMSE for energy/atom in active learning was slightly higher than that obtained using only 300 K samples in passive learning, but lower than that from the multi-temperature passive learning configuration. Moreover, the RMSE for forces in active learning was lower than those in both passive cases. In Comparison 2 for this phase, an actively trained model with 2,602 samples was compared to a passive model with 20,000 samples. Here, the RMSE for energy/atom in active learning was slightly higher than that from passive learning, while the RMSE for forces was lower.

A final critical test was conducted for both phases to assess the limits of the passive learning strategy. Although the passive learning models with 20,000 samples produced errors comparable to those from the much smaller active learning sets, further testing with a larger dataset of 80,000 samples revealed that this four-fold increase in data did not substantially decrease the error values. This suggests a saturation point for the effectiveness of passive learning

with MTP for this system, highlighting that simply adding more data does not guarantee a better potential.

For the DeePMD framework, active learning also proved highly effective. In Comparison 1 (57,180 samples), the actively trained model produced lower RMSEs for both energy/atom and forces than both of its passively trained counterparts (300 K and multi-temperature). In Comparison 2, the actively trained model (61,657 samples) was compared against a passive model with a larger dataset (127,212 samples). The RMSE for energy/atom was similar between the two, while the RMSE for forces was lower in the active learning case. Consistent with the MTP findings, this demonstrated that active learning could achieve comparable or better accuracy with significantly less data. However, unlike MTP, it was observed that further increasing the passive learning dataset to 333,340 samples did yield lower errors, indicating that DeePMD can leverage more data if available.

5.3.2 Prediction of equilibrium and elastic properties

With the error characteristics established, the investigation proceeded to validate the potentials' ability to predict fundamental 0 K physical properties. The validation focused on the equilibrium volume and lattice parameters derived from energy-volume curves, as well as the full set of elastic constants for both the γ -TiAl and α_2 -Ti₃Al phases.

The analysis for the MTP framework revealed a significant dependency on the learning strategy. In Comparison 1, the passively trained potential failed to identify a stable energy minimum in the energy-volume curves for either phase. In contrast, the actively trained MTP successfully predicted these equilibrium properties, with the predictions for the γ -TiAl phase being closer to DFT values than those for the α_2 -Ti₃Al phase, which were slightly overpredicted. In Comparison 2, where the passive potential was trained with a much larger dataset, it was able to predict the equilibrium properties more accurately than the active learning potential for both phases. Regarding the elastic constants, the actively trained MTP for γ -TiAl outperformed its passive counterpart in predicting several constants (C_{11} , C_{12} , C_{44} and C_{66}) with values closer to those from DFT in both Comparison 1 and 2. However, for the α_2 -Ti₃Al phase, the active learning predictions for the elastic constants deviated more from DFT values than the passive learning results in both Comparison 1 and 2.

A similar analysis for the DeePMD framework also highlighted the critical role of the learning strategy. As with MTP, the passively trained potential in Comparison 1 failed to find a minimum in the energy-volume curves for either phase, a failure rectified by the actively trained model. The active learning DeePMD potential yielded predictions for the α_2 -Ti₃Al phase that were closer to DFT than for the γ -TiAl phase. In Comparison 2, active learning yielded equilibrium volumes that were closer to the DFT results for both phases. However, for the equilibrium lattice parameters, passive learning performed marginally better than active learning. The elastic constants also showed improved predictions in Comparison 2 for both learning strategies. In Comparison 1 for the γ -TiAl phase, the passive learning approach exhibited significant deviations from DFT, most notably with a substantial overprediction of

C_{11} . Furthermore, compared to the MTP results, the active learning potentials developed with DeePMD provided predictions for α_2 -Ti₃Al that were closer to the DFT values.

Overall, active learning exhibited strong predictive performance for key material properties. Nevertheless, some outliers were observed, notably with the MTP active learning potential for α_2 -Ti₃Al, where the predictions for elastic constants and certain equilibrium volumes and lattice parameters deviated from expectations. This is likely because these fundamental properties were not the primary focus during the active learning sample selection process, resulting in a bias toward higher-temperature configurations that differ from the ground state, thereby impacting overall prediction accuracy.

5.3.3 Structural fidelity at finite temperatures

The ability of the potentials to capture correct atomic arrangements at finite temperatures (300 K, 500 K, 700 K, and 900 K) was assessed by comparing RDFs with AIMD data. For the γ -TiAl phase, the results were consistent and accurate across all models. In both Comparison 1 and Comparison 2, the RDFs derived from passive and active learning using MTP and DeePMD were found to be nearly identical and in excellent agreement with the reference data. This high level of performance is expected, as the structurally simpler γ -TiAl phase is relatively easy to model.

In contrast, the more complex α_2 -Ti₃Al phase highlighted a key difference between the learning strategies in the data-limited scenario of Comparison 1. Here, noticeable discrepancies were observed in the RDF curves from the passive learning models, particularly at 700 K, while such deviations were absent in the active learning results. However, in Comparison 2, where the passive learning models were trained on a much larger dataset, the RDFs for the α_2 -Ti₃Al phase from both passive and active learning were nearly identical to the AIMD reference. These differences show the difficulties the interatomic potentials encounter in capturing the complex structural details of α_2 -Ti₃Al, particularly at higher temperatures, compared to the γ phase.

5.3.4 Influence of Nb on thermomechanical behavior

Having validated the potentials' performance on fundamental properties, the investigation turned to the primary scientific question: the influence of Nb on the thermomechanical behavior of TiAl-based alloys. To this end, uniaxial tension tests were simulated on selected samples of γ -TiAl and α_2 -Ti₃Al with different Nb concentrations at a strain rate of 10^9 s^{-1} . A consistent trend was observed across the different models: a significant reduction in the UTS with increasing Nb concentration was found for both phases. This result was consistent for potentials developed using both MTP and DeePMD, across both active and passive learning cases, and aligns with the earlier findings in Article A.1 and with results reported in other studies^{94,95,100}.

To understand the atomic-scale mechanism responsible for this change in strength, GSFE computations were performed. For both the γ -TiAl and α_2 -Ti₃Al phases, it was observed

that the GSFE decreased with an increase in Nb concentration, a trend also noted in previous work¹⁰⁰. Generally, a higher GSFE indicates greater resistance to dislocation motion and reduced dislocation mobility, which directly enhances the strengthening behavior in nanomaterials. Conversely, a lower GSFE implies increased dislocation mobility and improved ductility. The combined results of the tension tests and GSFE calculations suggest that Nb addition generally decreases the strength of both phases while simultaneously increasing ductility. These findings are in full agreement with the conclusions from both Article A.1 and Article A.2, which demonstrated that Nb alloying enhances dislocation activity and ductility while reducing overall strength.

The combined results of the tension tests and GSFE calculations provide a clear, self-consistent mechanistic picture. The observed decrease in GSFE with increasing Nb content explains the concurrent reduction in UTS, as the energy barrier for plastic deformation is lowered. This same mechanism, by facilitating dislocation motion, also explains the known role of Nb in improving the material’s ductility. These findings are in full agreement with the conclusions from both Article A.1 and Article A.2, which demonstrated that Nb alloying enhances dislocation activity and ductility while reducing overall strength.

5.3.5 Summary

This work has demonstrated that the active learning strategy enables the development of robust ML-IAPs with a significant reduction in the required number of *ab initio* calculations. The efficiency of this approach was highlighted by the small initial datasets required, with MTP commencing with only 170 samples and DeePMD with 40,000 samples. This efficiency is further quantified by the total simulation time needed for data generation: the active learning approach required only 32 AIMD simulations of 2.5 ps duration, a substantial reduction compared to the 180 simulations of 10 ps each that were needed for the passive learning study.

The detailed comparisons presented in Article A.3 confirm the effectiveness of the resulting active learning potentials in predicting a wide range of material properties. However, a persistent challenge was identified: in both the passive and active learning studies, it was evident that accurately capturing the energetics of the α_2 -Ti₃Al phase remained difficult. This suggests that while active learning significantly improves the robustness of the potentials, further methodological refinements may be needed to fully describe this complex phase. As a future work, the methodology developed here can be extended to include more complex structures in the initial dataset, such as explicit interfaces between the γ -TiAl and α_2 -Ti₃Al phases. Although this would require more computational power to perform AIMD calculations on such systems, the use of active learning would be essential to manage the computational cost. By ensuring that only the most informative configurations are selected for labeling, this approach can continue the path toward highly accurate and efficient potentials for this complex alloy system.

Chapter 6

Conclusion

This thesis has presented a comprehensive investigation into the development and application of interatomic potentials for the technologically significant TiAlNb alloy system. The research commenced with an evaluation of a classical potential, which highlighted the limitations of traditional models for this complex alloy and established the need for more accurate methods. This motivated the core of the work: a rigorous comparative analysis of two state-of-the-art ML-IAP frameworks, DeePMD and MTP. Finally, the efficiency of the potential development process itself was addressed through a systematic comparison of passive and active learning strategies, demonstrating a path toward more rapid and data-efficient model creation.

Beyond the specific scientific findings of the three articles presented, a primary contribution of this thesis is to serve as a methodological guide for researchers aiming to apply similar techniques to other material systems. The methods for developing both MTP and DeePMD potentials, from descriptor construction to model training, have been discussed in detail. Furthermore, a thorough methodology has been provided for the implementation of active learning, a crucial technique for reducing the immense computational cost associated with *ab initio* data generation. In the interest of scientific transparency and to foster future research, the training and test datasets generated for the comparative studies in this work have been made publicly available^{93,116}. This ensures that the results presented are fully reproducible and provides the community with a high-quality benchmark dataset for the TiAlNb system, which can be used to validate and compare interatomic potentials developed using other ML frameworks.

Despite the advancements presented, it is important to acknowledge the limitations of the current work, which in turn point toward avenues for future research. In both the passive and active learning studies, it was evident that accurately capturing the energetics of the α_2 -Ti₃Al phase was a persistent challenge. While the active learning approach demonstrated improved performance over the passive case, further refinement of the training and query strategies is needed to fully resolve the subtle energy differences associated with this phase. A second limitation is that the training and validation datasets were primarily composed of simple bulk crystal structures, without complex microstructural features such as grain boundaries, interfaces, or dislocations. To effectively model the mechanical behavior of polycrystalline TiAlNb, these defects must be included in the training set. However, doing so drastically increases the computational cost of data generation. For passive learning, this would necessitate

an enormous number of computationally expensive *ab initio* calculations. While active learning substantially mitigates this cost, the on-the-fly labeling of these complex, high-atom-count configurations remains a significant computational hurdle.

Looking forward, several exciting research directions can build upon the foundations established in this thesis, particularly concerning the extension of these models to the large-scale simulations required for studying complex microstructural phenomena.

A primary challenge for large-scale simulations is the efficient exploration of rare events, such as dislocation motion or crack propagation, which may not be adequately sampled in standard active learning workflows. A promising approach to tackle this is demonstrated by recent work on in-operando active learning, where MTPs have been used to model systems with thousands of atoms containing dislocations and other defects¹¹⁷. This methodology could be combined with enhanced sampling techniques, such as metadynamics or temperature accelerated dynamics, to more effectively drive simulations toward discovering and learning the critical configurations that govern mechanical behavior.

Another major avenue for progress lies in the fundamental architecture of the machine learning models themselves. The DeePMD and MTP frameworks, as discussed in this thesis, first compute a set of rotationally invariant scalar features known as the descriptor. This descriptor is then fed into a standard neural network or linear model. This approach solves the symmetry problem by creating a representation of the environment that does not change upon rotation, effectively discarding the raw directional information. A newer and more powerful approach is found in equivariant neural networks. Instead of discarding this directional information, models such as NequIP, Allegro, and MACE process the full coordinate vectors of the atoms through specialized network layers^{118–120}. These layers are designed to be explicitly equivariant to translations, rotations, and permutations, meaning their internal vector and tensor representations transform predictably and correctly as the input coordinates are rotated. By processing the complete geometric information rather than just invariant scalars, these networks can learn a much richer and more detailed representation of the atomic interactions. The physics that is built into the architecture often leads to a dramatic improvement in data efficiency and accuracy, potentially allowing for the creation of robust potentials from even smaller *ab initio* datasets, which is a key requirement for modeling large, complex systems.

Beyond the model architecture, significant performance gains for large-scale simulations are being achieved through software and algorithmic optimization. The developers of DeePMD-kit, for example, have demonstrated near-linear scaling for a 100-million-atom system on the Summit supercomputer (a world-leading petascale system at Oak Ridge National Laboratory), utilizing over 27,000 GPUs by employing sophisticated parallelization and communication schemes¹²¹.

Finally, a significant future direction is the integration of the high-fidelity ML-IAPs developed in this thesis into multi-scale modeling frameworks. In such approaches, ML-IAPs can be used to compute atomic-scale parameters that serve as inputs for higher-level continuum models, a prospect highlighted in comparative assessments of machine learning interatomic potentials¹²². For example, the potentials developed here could be applied in large-scale MD

simulations to evaluate stacking fault energies and dislocation mobility parameters, which are essential inputs for predictive crystal plasticity simulations; the potential of ML-based methods for defect property calculations has been demonstrated in recent work¹²³. Likewise, these potentials may also provide interfacial free energies and kinetic coefficients that enable the coupling of fundamental atomic interactions into mesoscale phase-field simulations, as illustrated in recent efforts to link density-based phase-field models with atomistic potentials¹²⁴. This multi-scale linkage allows the atomic-scale accuracy of ML-IAPs to directly inform predictions of engineering-scale phenomena.

Ultimately, the methods used and potentials developed in this work represent a significant step toward the predictive, bottom-up design of next-generation high-temperature alloys. By providing a pathway to creating accurate, computationally efficient models for complex multi-component systems, this research contributes to the ongoing paradigm shift toward data-driven materials science, enabling a new level of physical realism in large-scale atomistic simulations.

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Appendix A

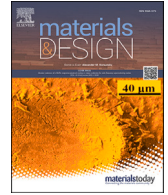
Peer reviewed and published articles

A.1 Publication 1

Studying the effects of Nb on high-temperature deformation in TiAl alloys using atomistic simulations

Anju Chandran, Hariprasath Ganesan and Christian J. Cyron

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Studying the effects of Nb on high-temperature deformation in TiAl alloys using atomistic simulations

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ABSTRACT

Intermetallic γ (TiAl)-based alloys find their application as high-temperature materials for aero engine and automotive components. Microstructure optimization and microalloying play key roles in optimizing these alloys. Several pioneering experimental works showed improved mechanical properties of γ (TiAl)-based alloys containing Niobium (Nb). Despite Nb being a key alloying element, its contribution remains debated, if not least understood, due to the TiAl microstructure's complexity with hierarchical interfaces. This work examines the effects of Nb on the high-temperature deformation behavior of TiAl alloys using atomistic simulations. These revealed that Nb alloying retarded the stress-induced phase transformation of $\gamma \rightarrow \alpha_2$, favoring a refined microstructure with the dislocation sources from microstructure boundaries and interfaces at high temperature and improving thus the ductility. Our microstructure-informed atomistic models reveal a comprehensive picture of the underlying nanomechanical events.

1. Introduction

Gamma titanium aluminide γ (TiAl)-based intermetallic alloys have various applications as lightweight structural materials due to their properties like low density, good oxidation resistance, high strength and creep resistance at high-temperature [1]. The interesting intermetallic phases for engineering applications are γ -TiAl (ordered face-centered tetragonal, $L1_0$, $P4/mmm$) and α_2 -Ti₃Al (ordered hexagonal, $D0_{19}$, $P6_3/mmc$). The past decades witnessed new generations of γ -TiAl based alloys with increasing service temperatures [1–4]. Among them, TNB (T: TiAl, N: Nb and B: B) and TNM (T: TiAl, N: Nb and M: Mo) alloys [1,2,5,6] made substantial improvements. The stoichiometry of these alloys read: Ti-(42–48)Al-(0–10)X-(0–3)Y-(0–1)Z-(0–0.5)RE where, X = Cr, Mn, Nb, Ta; Y = Mo, W, Hf, Zr; Z = C, B, Si; RE denotes rare earth elements. TNB alloys contain 5–10 at. % Nb and small additions of B and C, whereas TNM alloys have similar concentrations of Nb and Mo [2]. Against this background, Nb plays a crucial role in enhancing the mechanical properties of binary γ (TiAl)-based alloys.

Many experimental works [1,2,7,8] reported that Nb enhances the ductility and strength of TiAl. It reduces the stacking fault energy, favor-

ing the activation of mechanical twins resulting in enhanced strength [7,9] and improved low-temperature ductility [9] of γ (TiAl)-based alloys. The contribution of Nb to increased toughness, tensile strength, and yield strength of γ (TiAl)-based alloys was studied by various experimental works [10–19]. The site preference of Nb on Ti lattice sites was experimentally shown in [20–23]. It is worth emphasizing that the insights from the experimental studies of TiAlNb systems discussed the combined effects of microstructure boundaries and alloying elements, e.g., (Nb, Mo, B, and C) on the TiAl system. [1,2,6,8] reported improvement in mechanical properties of γ (TiAl)-based alloys, e.g., ductility and strength by Nb alloying. Despite several interesting contributions from previous experimental investigations and studies on improving TNB alloys, Nb's isolated contributions to the strength and ductility of these alloys remain unclear. This work aims to address this research question by investigating the effects of Nb on thermo-mechanical behavior of γ (TiAl)-based alloys using atomistic simulations.

Density Functional Theory (DFT) helped to understand the site preference of alloying elements in the γ -TiAl and α_2 -Ti₃Al phases [24–28]. Strong site preference of Nb at Ti sites was reported in single phases [24] and in bulk and interface regions [25] in a γ -TiAl/ α_2 -Ti₃Al inter-

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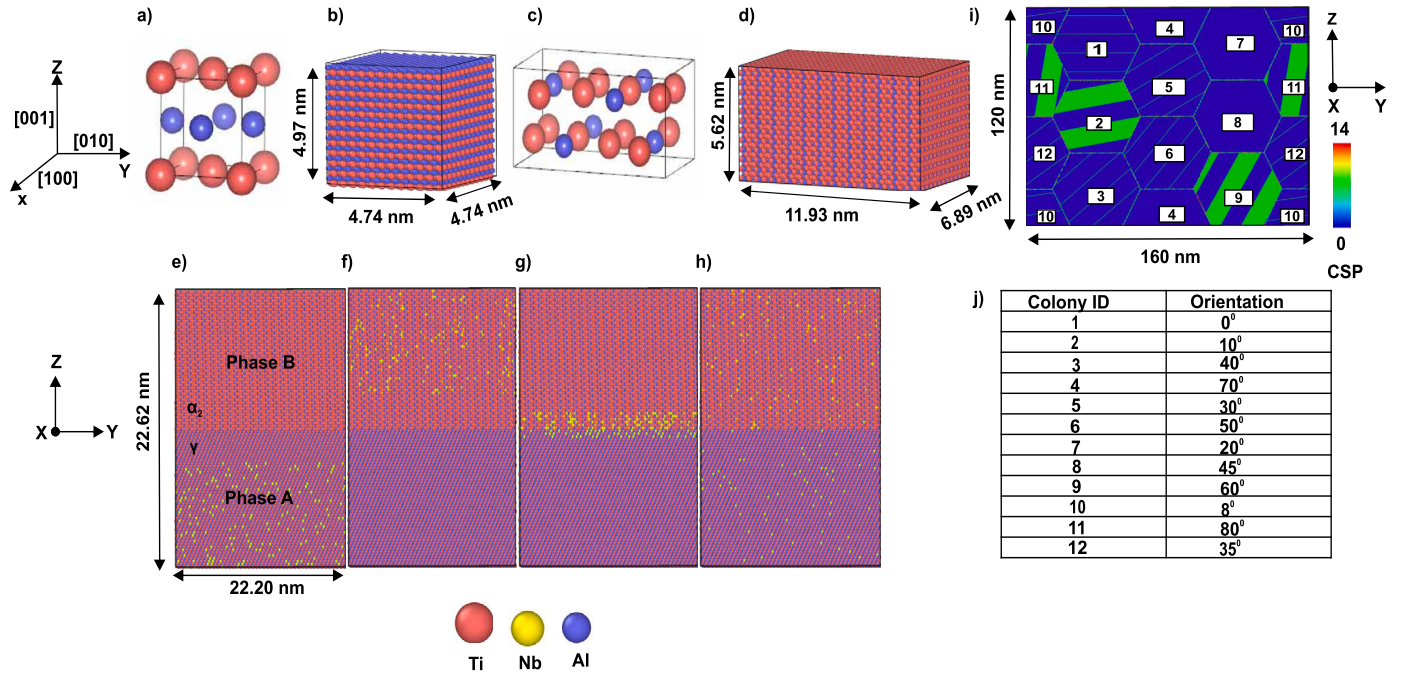


Fig. 1. Three categories of atomistic models used in this work: 1) Single phase models ((a) unit cells of γ -TiAl compose (b) γ -TiAl supercell; (c) unit cells of α_2 -Ti₃Al compose (d) α_2 -Ti₃Al supercell); 2) Single-lamellar interface models (e.g., γ/α_2) with (e) Nb distributed in phase A, (f) Nb distributed in phase B, (g) Nb distributed near lamellar interface, and (h) Nb homogeneously distributed. Phase A corresponds to γ and phase B to α_2 , γ_{PT} , γ_{TT} and γ_{RB} ; 3) (i) Microstructure-informed atomistic model (MIAM) [40] with 12 nano-colonies, where each colony has different lamella combinations (γ/α_2 , γ/γ_{PT} , γ/γ_{RB} , γ/γ_{TT}), (j) Table showing the colony ID (identification number) and orientations of colonies in the MIAM.

face model. The site preference of Nb was also studied using Density of States (DOS) calculation in [26]. However, DFT simulations are prohibitively expensive to investigate atomistic models containing $\geq 10^3$ atoms, thus, preventing us from incorporating complex microstructure features like grain/colony boundaries, interfaces and their contributions.

Molecular dynamics (MD) enables understanding the evolution of many-body atomic systems (routinely $10^3 - 10^9$ atoms) besides capturing the interplay between thermal, mechanical, chemical, and microstructure contributions. However, the chemical accuracy and physical reliability of MD simulations are limited by the choice of the underlying interatomic potentials or force fields and appropriate boundary conditions. To gain insight into γ (TiAl)-based alloys, recent MD studies used in particular the Zope potential [29], Farkas binary potential [30], Farkas ternary potential [31], and Kim potential [32]. Recently, [33] developed a new MEAM based Nb-Al-Ti ternary potential which remains to be explored. [34–49] studied TiAl systems using the Zope potential to understand the synthesis of phases, strength, ductility, deformation behavior, fracture mechanism, tensile and creep properties of the material. [50–53] used the Farkas binary potential to investigate point defect formation energy, defect concentration, diffusion mechanisms and nature of crack formation in TiAl systems. Creep mechanisms and self-diffusion in nanocrystalline TiAl alloys were studied in [54,55] using the Kim potential. The Farkas ternary potential was used in [56–59] to investigate high Nb-containing TiAlNb systems to study the B2 phase stability, ductile nature and the diffusion properties of Nb. Previous MD studies [56–61] of TiAlNb did not address the influence of Nb on the thermo-mechanical properties of the material. One major challenge in studying high Nb-containing TiAl phases using MD simulations is the availability of limited interatomic potentials targeting the intermetallic phases of engineering interest.

In this paper, we first assess the transferability of the Farkas potential [31] to different target scenarios and compared it with other established interatomic potentials like the Zope [29] and Kim [32] potentials for γ (TiAl)-based systems. Using the insights obtained this

way, we study the isolated effect of Nb atoms on the lattice parameters and neighboring chemical environment using MD simulations and compare them with chemically accurate DFT calculations where possible. Driven by the insights gained, we alter the Nb concentration in atomistic models with varying microstructure complexity to unravel Nb's contributions to the thermo-mechanical behavior of γ (TiAl)-based alloys.

2. Methods

2.1. Molecular dynamics (MD)

Intermetallic γ (TiAl)-based alloys of engineering interest consist of two key phases, namely γ -TiAl (ordered face-centered tetragonal L1₀ structure) and α_2 -Ti₃Al (ordered hexagonal D0₁₉ structure). The volume fraction of γ and α_2 depends on the alloy's heat treatments and thermo-mechanical processing [62]. To investigate Nb's contribution comprehensively, we considered three categories of TiAl atomistic models: 1) Single phase models (with and without Nb), 2) Single lamellar interface models (Nb-rich phase/bulk, Nb-rich interface, homogeneously distributed Nb), and 3) Microstructure-informed atomistic models (MIAMs) with nano-polycolonies, as shown in Fig. 1. In the following, we first discuss the interatomic potentials used in these three types of models (section 2.1.1), then their atomistic configuration (sections 2.1.2 - 2.1.4), the different ways we resembled alloying in them (section 2.1.5) and we mimicked tension tests with them (section 2.1.6) and how they were implemented computationally (section 2.1.7).

2.1.1. Interatomic potentials

Herein, we primarily used the ternary Farkas potential [31] to define the interatomic interactions of the TiAlNb system. To realize this potential, the developers combined three binary potentials, namely one for Nb-Al (fitted for A15 Nb₃Al and D0₂₂ NbAl₃), one for Nb-Ti (for disordered BCC phase), and one for Ti-Al [30]. The resulting ternary potential (NbTiAl) was reported to predict the Nb-rich phases

like the orthorhombic Ti_2AlNb and ternary B_2 phase. The Farkas potential [31] was used to study and predict the behavior of the ternary TiAlNb system [57], although no input pertaining to the ternary system was included in the fitting procedure of this potential. We examined also other secondary interatomic potentials, namely the Zope potential [29], Kim potential [32], and Xiang potential [33] for the TiAl system to investigate the scope and transferability of the Farkas potential. Among them, the Zope potential and Kim potential were extensively used previously [34–41], to study several aspects in binary $\gamma\text{-TiAl}$ systems. Most recently, Xiang et al. developed an interatomic potential for the TiAlNb system [33]. A systematic investigation of this new ternary potential is beyond the scope of this work. Therefore, this potential was used only for selected models (for more information, see A). For brevity, in this article, we refer Farkas ternary potential [31] as Farkas potential (otherwise as Farkas binary potential [30] explicitly).

2.1.2. Single phase models

The single-phase models of $\gamma\text{-TiAl}$ ($a = 3.950 \text{ \AA}$, $c/a = 1.048$) and $\alpha_2\text{-Ti}_3\text{Al}$ ($a = 5.740 \text{ \AA}$, $c/a = 0.815$) were modeled with the Farkas binary potential [30], where such single phases represent the lamellae of the constituent intermetallic phase. The single-phase $\gamma\text{-TiAl}$ and $\alpha_2\text{-Ti}_3\text{Al}$ models were realized with simulation box dimensions of $4.74 \text{ nm} \times 4.74 \text{ nm} \times 4.97 \text{ nm}$ and $6.89 \text{ nm} \times 11.93 \text{ nm} \times 5.62 \text{ nm}$, respectively. Both of them were modeled with crystallographic orientation of X-direction [1 0 0], Y-direction [0 1 0], and Z-direction [0 0 1]. These single-phase models aid in understanding the transferability of Farkas potential [31] by comparing it with the Zope and Kim potentials. We computed the formation energy (ΔE_f) of point defects, elastic constants, and cohesive energy (E_{coh}) at $T = 0 \text{ K}$ as the Farkas potential was fitted to predict some of these properties. Furthermore, we computed the phase transition temperature to evaluate the Farkas potential's predictive abilities at elevated temperatures (2400 K). Some tension tests were performed at ground state and room temperature to assess the mechanical property predictions.

2.1.3. Interface models

We realized atomistic single-lamellar interface models of γ/α_2 and three γ/γ variants (i.e., γ/γ_{PT} , γ/γ_{RB} , and γ/γ_{TT}), realized by a rotation around the $\langle 111 \rangle_\gamma$ axis by 60° , 120° and 180° , resulting in a pseudo twin (γ_{PT}), rotational boundary (γ_{RB}) and true twin (γ_{TT}) with box dimension of $22.6 \text{ nm} \times 22.2 \text{ nm} \times 22.4 \text{ nm}$, containing 675,840 atoms. Here, $\gamma\text{-TiAl}$ was modeled with the crystallographic orientations of X-direction [1–1 0], Y-direction [1 1–2], and Z-direction [1 1 1]. Similarly, we modeled $\alpha_2\text{-Ti}_3\text{Al}$ with X-direction [1–1 0], Y-direction [1 1 0], and Z-direction [0 0 1]. The single-lamellar interface models comprised two phases, namely A and B in Fig. 1e–h. Phase A denotes $\gamma\text{-TiAl}$, phase B denotes either $\alpha_2\text{-Ti}_3\text{Al}$ or one of the three variants (γ_{PT} , γ_{RB} , γ_{TT}). Phase A and phase B represent one of the possible constituent intermetallic phases of the target microstructure, respectively. Furthermore, we alloyed the above atomistic models with different Nb concentrations (1.0 at.%, 2.0 at.%, 5.0 at.%, 6.5 at.%, and 7.5 at.%) to understand the relation between Nb, microstructure features (e.g., interfaces/boundaries) and thermo-mechanical properties at high temperatures (e.g., 700 K, 900 K).

2.1.4. Microstructure-informed atomistic models

We considered microstructure-informed atomistic models (MIAMs) as introduced previously in [40]. They consisted of 12 nano-sized colonies in a periodic domain of the size 12 nm (X) $\times$ 160 nm (Y) $\times$ 120 nm (Z), containing 14,612,466 atoms. Each colony comprises several of one of the following four interfaces: γ/α_2 , γ/γ_{PT} , γ/γ_{RB} , and γ/γ_{TT} . The MIAMs used the experimentally informed microstructure parameters like lamella thickness (λ' , λ^{a_2}), α_2 volume fraction (φ^{a_2}), and distance between subsequent α_2 lamellae (d^{a_2}) reported already in our previous work [40]. The orientation of each colony (designated with

a colony ID) around the X-axis is provided in Fig. 1 (i, j), along with the centrosymmetry parameter (CSP) values. The CSP measures the local lattice distortion of an atom, thus, aiding in characterizing defects, interfaces, and colony boundaries in the MIAM. We considered the Nb concentrations 0.0 at.%, 0.5 at.%, and 1.0 at.% at temperatures 300 K, 500 K, 700 K and 900 K.

2.1.5. Alloying

Many experimental studies [21,22] along with several other works [24–26,59,63] show Nb atoms prefer Ti sites in $\gamma(\text{TiAl})$ -based alloys. Our DFT calculations yielded similar site preferences of Nb at Ti sites (see section 2.2). Therefore, we realized the ternary atomistic models by substituting Nb atoms at Ti sites to achieve different target Nb concentrations. e.g., 0.0–5.0 at.% with 1.0 at.% increments in the single-phase models. Similarly, in the single-lamellar interface models, Nb was introduced at random Ti sites either in Phase A or Phase B regions (Nb-rich phase), interface regions (Nb-rich interface) or homogeneously everywhere (Fig. 1 e–h) to reach target concentrations of 1.0 at.%, 2.0 at.%, 5.0 at.%, 6.5 at.%, and 7.5 at.%, respectively. Based on our preliminary investigation of the Farkas potential for different microstructures, we studied the transferability limits of this potential to microstructure models with complex interfaces. The transferability limit of an interatomic potential refers to the extent to which the potential can reasonably describe the material system's behavior beyond those for which it was originally parameterized. Thus, in our MIAM models, we considered only low Nb concentrations (0.5 at.% Nb and 1.0 at.% Nb), wherein Nb atoms were introduced at a distance of at least 4 nm from the grain boundaries. We also reached the transferability limit of the Farkas' potential for Nb concentrations above 2 at.% near the single lamellar interface models for Nb-rich interfaces, see Fig. 1). Note the observed transferability limits are valid only for the assumption that Nb atoms occupy Ti sites in our atomistic models.

2.1.6. Simulated tension tests

To simulate tension tests, we applied in our MD models uniaxial tension loading along the Z-direction at a strain rate of $10^9/\text{s}$, with periodic boundary conditions in all directions. To this end, we used single phase models of $\gamma\text{-TiAl}$ and $\alpha_2\text{-Ti}_3\text{Al}$, single-lamellar interface models, and MIAM models both with and without Nb to understand Nb's influence on the high-temperature deformation behavior of TiAl -based alloys.

First, we performed a free energy minimization of the atomistic models using a combination of conjugate gradient (CG) and FIRE algorithms in LAMMPS. Then, we equilibrated the models for temperature (1 K) and pressure (1 bar) using NVT and NPT for 100 ps each. Later, the simulation models were heated to the target temperature of 300 K for single phase models, 700 K and 900 K for interface models, and 300 K, 500 K, 700 K and 900 K for MIAMs. Note that the defect-free single-phase models were considered only for the ground state (to enable comparisons with DFT) and room temperature where possible. The heating rate for all the considered models was 3 K/ps followed by thermal equilibration for 100 ps. The timestep size was 1 fs for models without Nb using the Zope, Farkas, and Kim potentials) and 0.1 fs for models with Nb using the Farkas' potential.

2.1.7. Implementation

The MD simulations were realized using the classical MD code Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) [64]. We used some in-house tools, along with AtomsK [65] and VESTA, for constructing the atomistic models used in this work (Fig. 1). We used OVITO [66] for visualization.

2.2. Density functional theory (DFT)

We performed some Density Functional Theory (DFT) calculations using Quantum Espresso [67] to assess the chemical accuracy of the

Table 1

TiAl material parameters computed with the interatomic potentials of Farkas [31], Zope [29], and Kim [32]. C_{11} - C_{66} denote the entries of the material stiffness matrix (Voigt convention) for characterizing elastic constants, $\Delta E_f V_{Ti}$ and $\Delta E_f V_{Al}$ denote raw formation energy of Ti and Al vacancies, $\Delta E_f Al_{Ti}$ and $\Delta E_f Ti_{Al}$ denote raw formation energy of Al and Ti antisites, and E_{coh} denotes the cohesive energy, respectively. Elastic constants (C_{11} - C_{66}) are given in GPa, whereas raw formation energies and cohesive energy are given in eV.

Quantities	γ -TiAl			α_2 -Ti ₃ Al		
	Farkas	Zope	Kim	Farkas	Zope	Kim
C_{11}	245.50	196.67	181.11	233.60	199.96	200.58
C_{33}	352.02	212.79	234.39	285.27	224.51	238.92
C_{12}	116.24	107.49	76.27	109.27	88.51	107.78
C_{13}	190.85	113.53	133.75	87.58	74.43	91.17
C_{44}	146.02	92.06	86.00	57.77	51.42	45.56
C_{66}	71.58	85.16	61.56	62.21	56.04	46.44
$\Delta E_f V_{Ti}$	-6.50	-6.81	-6.60	-6.28	-6.73	-6.60
$\Delta E_f V_{Al}$	-5.21	-5.37	-5.32	-5.86	-6.07	-5.81
$\Delta E_f Al_{Ti}$	-1.97	-1.84	-1.67	-1.23	-1.03	-1.10
$\Delta E_f Ti_{Al}$	0.97	1.08	0.90	0.56	0.37	-0.056
E_{coh}	-4.40	-4.51	-4.50	-4.68	-4.77	-4.78

Farkas' potential [31] for the target intermetallic phases at the ground state. The electron-ion interactions were described using projected augmented wave (PAW) [68] potentials. Exchange and correlation effects were treated within the generalized gradient approximation (GGA) using the Perdew–Burke–Ernzerhof (PBE) [69] functional. We considered a plane wave cut-off energy of 400 eV with the electronic energy and atomic force components converged within 10^{-6} eV and 0.01 eV/Å, respectively. The simulation supercells represent the single intermetallic phases, i.e., γ -TiAl (ordered face-centered tetragonal L1₀) and α_2 -Ti₃Al (ordered hexagonal D0₁₉) consisting of 32 atoms, whose unit cell representation is provided in Fig. 1 a, c. The Brillouin zone of the supercell was sampled using a 7×7×7 Monkhorst-Pack [70] k-point mesh. We computed formation energy, electron localization function (ELF), and partial density of states (PDOS) by substituting an Nb atom at either Ti or Al lattice sites.

Several previous works [24–26,59,63,71] claimed a Nb preference of Ti sites. To verify this claim, we computed Nb's site preference, by substituting the Nb atom either at Ti or Al sites, following [24] and using

$$E_f^{Ti} = \frac{E_{Ti_{m-1}NbAl_n}^\eta - [(m-1)E_{Ti} + E_{Al} + E_{Nb} + nE_{Al}]}{m+n} \quad (1)$$

and

$$E_f^{Al} = \frac{E_{Ti_mNbAl_{n-1}}^\eta - [mE_{Ti} + E_{Al} + E_{Nb} + (n-1)E_{Al}]}{m+n} \quad (2)$$

Equations (1) and (2) refer to the formation energy of a TiAl atomic configuration with Nb atoms substituted either at Ti or Al sites, respectively. $E_{Ti_{m-1}NbAl_n}^\eta$ and $E_{Ti_mNbAl_{n-1}}^\eta$ denote the total energy of the supercell configuration describing the intermetallic phase η (i.e., γ -TiAl and α_2 -Ti₃Al), where one Nb atom replaces a single Ti or Al atom. E_{Al} , E_{Ti} , and E_{Nb} are the total energies of Al in FCC (face-centered cubic), Ti in HCP (hexagonal close-packed) and Nb in SC (simple cubic) structures, respectively.

Using DFT calculations for TiAl systems, we investigated Nb's site preference and Nb's chemical interactions with host atoms in γ -TiAl and α_2 -Ti₃Al. To this end, we computed the electron localization function (ELF), which measures the probability of finding an electron in the neighborhood of a reference electron with the same spin [72].

3. Results and discussion

3.1. Assessment of Farkas' interatomic potential

3.1.1. Binary TiAl systems

Herein, we evaluate the predictive ability of the Farkas [31] potential against the one of the Zope [29] and Kim [32] potential for

the target intermetallic phases, i.e., γ -TiAl and α_2 -Ti₃Al in TiAl system. Table 1 provides computed values of elastic constants, formation energy (ΔE_f), and cohesive energy (E_{coh}) of point defects for the different interatomic potentials considered. We also calculated the phase transition temperature of the intermetallic phases of interest. To evaluate the mechanical property prediction, we compared the Ultimate Tensile Strength (UTS) of intermetallic phases, i.e., γ -TiAl and α_2 -Ti₃Al.

Farkas potential [31] predicts slightly higher elastic constants compared to the Zope [29] and Kim potential [32]. Such discrepancy could stem from two aspects: first, the three interatomic potentials relied on different DFT and experimental data; second, Farkas' potential was targeted for ternary intermetallic regions, e.g., orthorhombic Ti₂AlNb with high Nb content.

Although the high elastic constants for the γ phase (at $T = 0$ K, refer Table 1) from Farkas potential can influence the reversible deformation behavior, those effects could be less pronounced in atomistic models (e.g., MIAMs) with high microstructure complexity encompassing phase fractions, lamellar thicknesses, lamellar interfaces, and colony boundaries. Considering the temperature and directional dependence of elastic constants, the interplay of temperature, strain rate, and microstructure complexity (i.e., phase volume fractions and their crystallographic directions) determine the thermo-mechanical behavior of TiAl alloys. Furthermore, we calculated the formation energy (ΔE_f) of four point defects, namely Ti vacancy (V_{Ti}), Al vacancy (V_{Al}), antisite at Ti sublattice (Al_{Ti}), and antisite at Al sublattice (Ti_{Al}). Our computations suggest that Ti vacancy (V_{Ti}) and antisite at Ti sublattice (Al_{Ti}) are preferred energetically over Al vacancy (V_{Al}) and antisite at Al sublattice (Ti_{Al}) in both γ -TiAl and α_2 -Ti₃Al. To this end, vacancy and antisite formation mechanisms might act as precursors for the diffusion of alloying elements in γ -TiAl and α_2 -Ti₃Al, preferably at Ti sites. Thus, a critical point defect concentration in the system is foreseen to favor diffusion. In general, the values of the cohesive energy E_{coh} and the energy of ΔE_f for point defects agreed reasonably well between the three potentials of Farkas, Zope and Kim. This suggests a generally good transferability of Farkas potential also for binary intermetallic phases, i.e., γ -TiAl and α_2 -Ti₃Al.

To evaluate the ability of Farkas' [31] potential to predict melting points and phase transitions, the single intermetallic phase models (i.e., α_2 -Ti₃Al and γ -TiAl) were heated to 2400 K at a heating rate of 2 K/ps. The models showed phase transitions around 1774 K (α_2 -Ti₃Al) and 2055 K (γ -TiAl), respectively (Fig. 2). Compared to Farkas' potential, the other two potentials predicted higher transition temperatures for both phases (Zope potential: 2181 K (α_2 -Ti₃Al) and 2053 K (γ -TiAl), Kim potential: 2712 K (α_2 -Ti₃Al), 2475 K (γ -TiAl)). Note that the experimentally [1,73] reported melting points of 1473 K for α_2 -Ti₃Al and 1723 K for γ -TiAl are much closer to the prediction based on Farkas' potential compared to the other two potentials. These experimental ob-

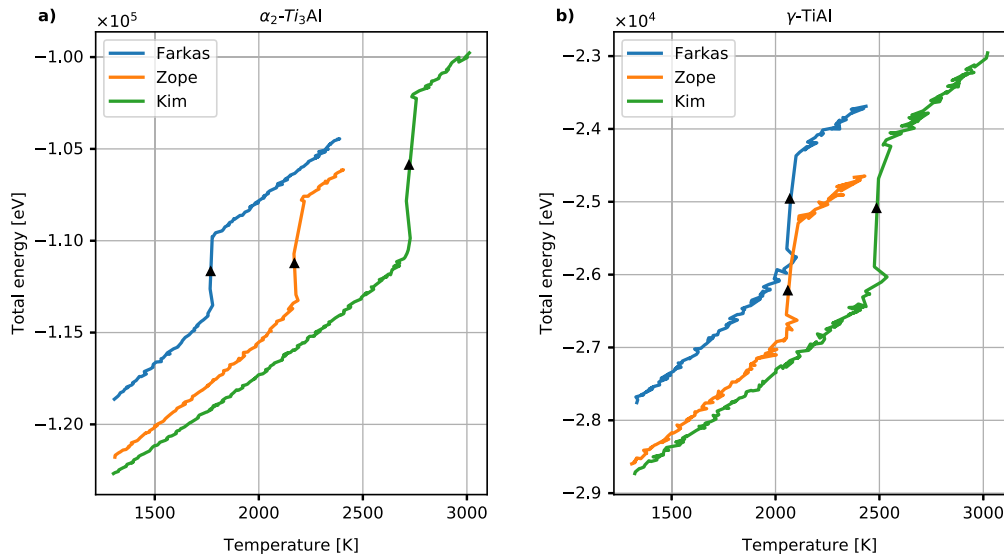


Fig. 2. Comparison of heating curves for single phases of (a) α_2 -Ti₃Al and (b) γ -TiAl using the interatomic potentials of Farkas [31], Zope [29] and Kim [32]. The black markers denote the phase transition region for each intermetallic phase.

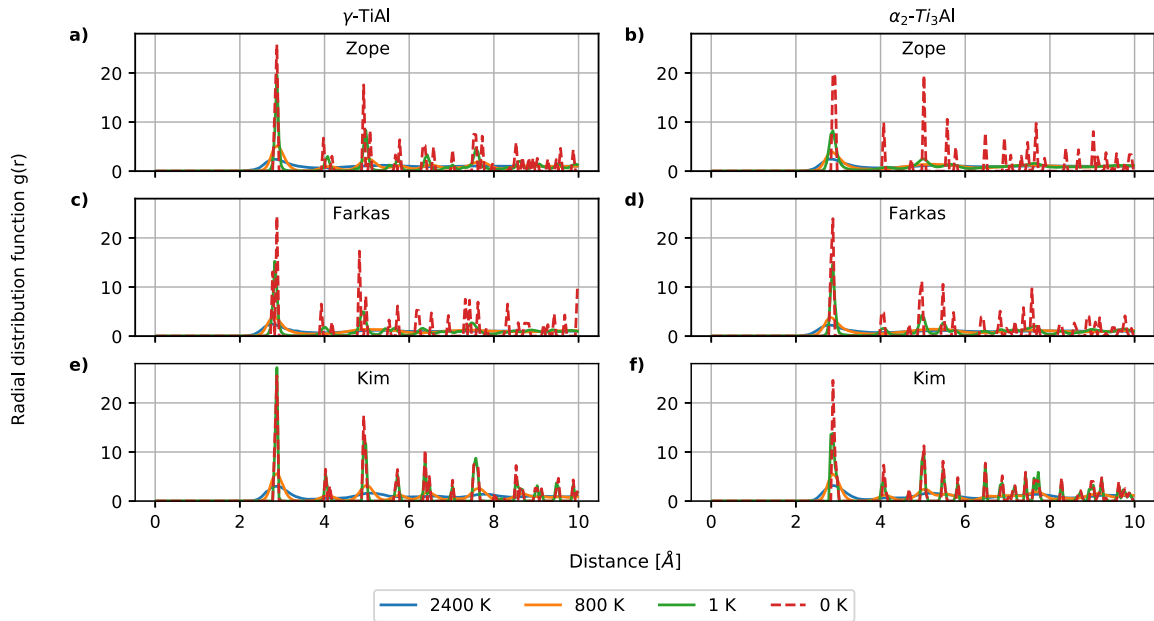


Fig. 3. Radial distribution function $g(r)$ for γ -TiAl (left) and α_2 -Ti₃Al (right) and the potentials of Zope [29] (top), Farkas [31] (middle), and Kim [32] (bottom) at temperatures 2400 K, 800 K, 1 K, and 0 K (ground state for comparison) to demonstrate the ability of interatomic potentials to sample the phase transition.

servations agree with [74], reporting that γ -TiAl precipitated or formed either from α_2 -Ti₃Al or α -Ti, underlining that the α_2 -Ti₃Al phase has a lower melting point than the γ -TiAl phase. While this is in agreement with the computations based on Farkas' potential, the potentials of Kim and Zope predict an inverse order of the melting points.

After simulating heating, we simulated also cooling to evaluate the capability of Farkas' potential to capture phase transformation. We cooled the configurations from 2400 K to 1 K at a general cooling rate of 2 K/ps and a cooling rate of 0.01 K/ps specifically for γ -TiAl because in agreement with [75] we observed difficulties in generating an ordered intermetallic phase at higher cooling rates. Fig. 3 shows the radial distribution function (RDF) for γ -TiAl and α_2 -Ti₃Al for selected temperatures (0 K (ground state), 1 K, 800 K, 2400 K). The agreement between the peaks at low temperatures (0 K, 1 K) for the three different potentials of Farkas, Zope and Kim underlines the ability of Farkas' po-

tential to capture the same effects as Zope's potential though at a much higher cooling rate, i.e., 2 K/ps.

We simulated tension tests to evaluate how different potentials predict the mechanical behavior of the TiAl system. To this end, we applied a strain rate of $10^9/\text{s}$ on single phase models of γ -TiAl and α_2 -Ti₃Al equilibrated at 300 K. The corresponding stress-strain curves of the tension tests are plotted in Fig. 4. For γ -TiAl, the Farkas and Zope potentials yield similar UTS, while the Kim potential showed a slightly higher value. For α_2 -Ti₃Al, the Kim and Zope potential yield a similar UTS, the Farkas potential a significantly lower one. Note that Farkas' potential yields a lower strength for α_2 -Ti₃Al than for γ -TiAl (although along the [0 0 1] direction), which will be crucial in interpreting the results of the complex models below. Fig. 5 shows the dislocation density evolution of single phase γ -TiAl and α_2 -Ti₃Al during simulated tensile tests, using DXA analysis [66]. For γ -TiAl, both the Farkas and Zope

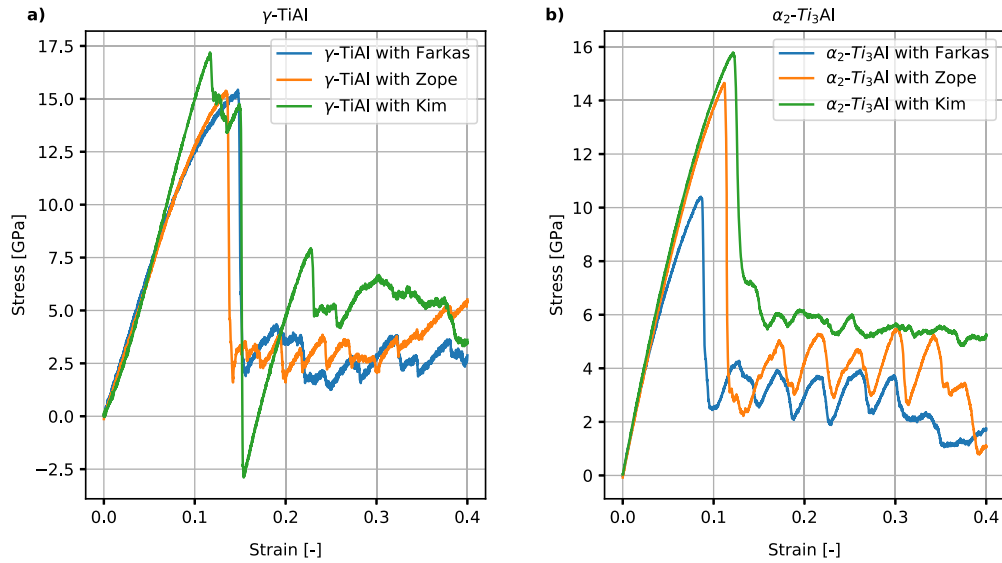


Fig. 4. Stress-strain curves in simulated tensile tests at 300 K with a strain rate of $10^9/s$ for a) γ -TiAl and b) α_2 -Ti₃Al using the three different interatomic potentials of Farkas [31], Zope [29], and Kim [32].

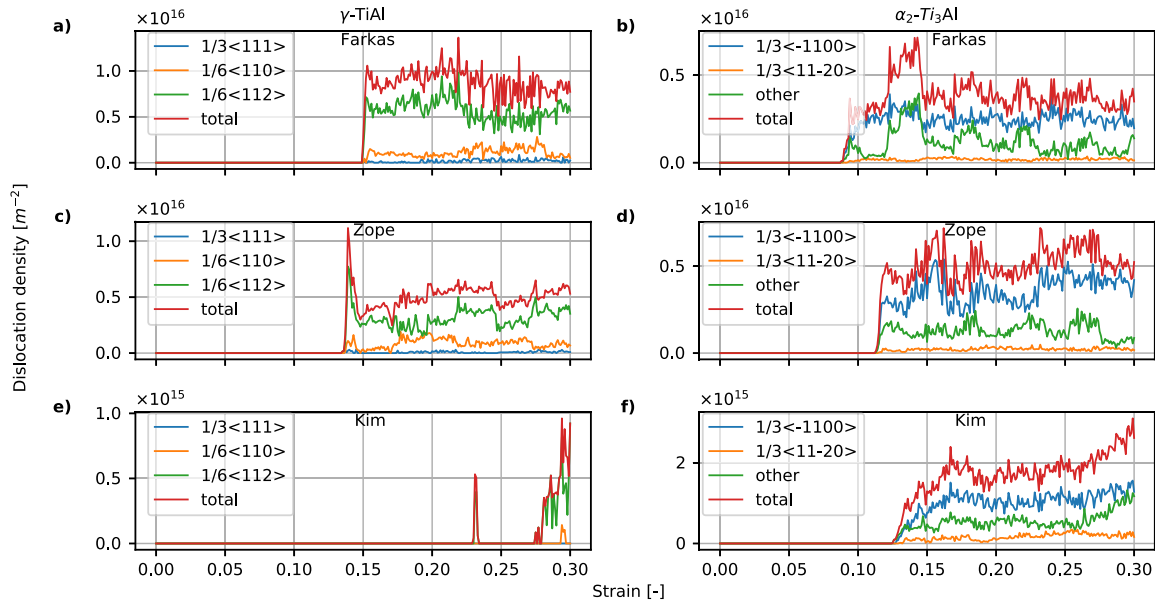


Fig. 5. Dislocation density in simulations of tension tests using MD for the three different interatomic potentials of Farkas [31], Zope [29], and Kim [32].

potential showed qualitatively similar dislocation densities (Fig. 5 a, c), whereas the Kim potential delayed the onset of dislocations (Fig. 5 e). For α_2 -Ti₃Al, again only minor differences in the dislocation evolution are observed between the Farkas and Zope potential, whereas the Kim potential leads to a remarkably late onset of dislocation formation but at the same time considerably higher overall densities for higher strains. These observations agree with [76], where dislocation densities between simulations with the Zope and Kim potential were compared. We attribute the deviations of the results obtained with the Kim potential compared to the other two potentials to its underlying MEAM (Modified Embedded Atom Method) formulation, which systematically accounts for the angular contributions.

Herein, we evaluated the transferability of the Farkas potential in predicting the behavior of single intermetallic phases, i.e., γ -TiAl and α_2 -Ti₃Al, by comparing it with the Zope and Kim potential.

In summary, our computations provided elastic constants, point defect formation energies, cohesive energies, transition temperature, and predictions of the mechanical behavior. The critical analysis of our

results indicates that the ternary Farkas potential is applicable (i.e., transferable) also to binary intermetallic phases (i.e., γ -TiAl and α_2 -Ti₃Al) that are of engineering interest. That is, it can also be used in Nb-free atomistic models in the forthcoming subsections.

3.1.2. Effect of Nb in intermetallic phases

We performed DFT calculations to investigate how Nb influences lattice constants, site preference (energetics), and electron localization functions of neighboring Ti and Al atoms in single intermetallic γ -TiAl and α_2 -Ti₃Al. These DFT calculations served as a reference point to evaluate the ability of Farkas' potential [31] to capture the effects of Nb in γ -TiAl and α_2 -Ti₃Al phases. The existing claim of Nb preference of Ti sites [24–26,59,63,71] was verified by our DFT calculations, which helped us to construct microstructure-informed atomistic models. Analyzing the electron localization function (ELF) and the partial density of states (PDOS) sheds light on the atomic interactions and chemistry of the bonding in the neighborhood of Nb atoms.

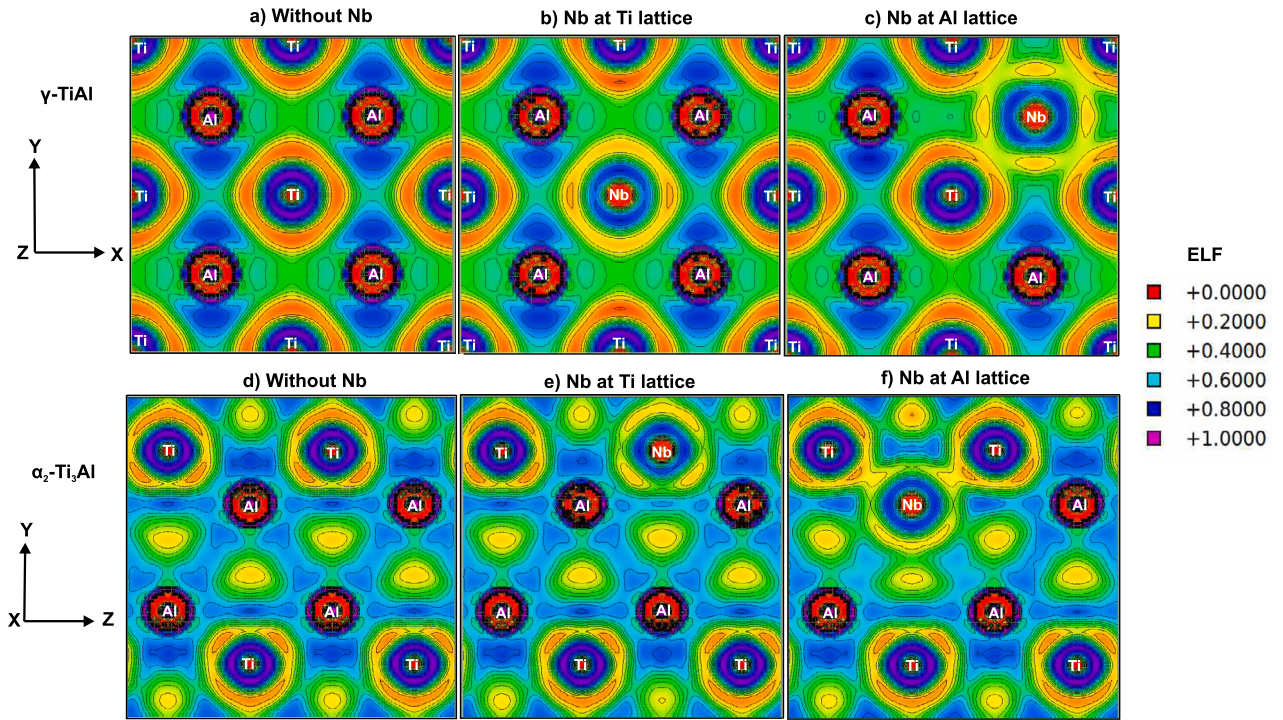


Fig. 6. Comparison of electron localization function (ELF) using DFT: γ -TiAl (top) and α_2 -Ti₃Al (bottom) before and after introducing an Nb atom either at Ti or Al site.

For the γ -TiAl phase, the formation energies (E_f^{Ti}) were computed as -1.5697 eV (Nb placed at Ti site) and -0.7908 eV (Nb placed at Al site), respectively. Similarly, for α_2 -Ti₃Al the computed formation energies were -0.7919 eV (Nb placed at Ti site) and -0.5915 eV (Nb atom placed at Al site). Herein, for both intermetallic phases, Nb at the Ti site had a lower formation energy, i.e., was energetically preferred. Hence, we placed Nb atoms at the Ti lattice sites in our atomistic models.

Fig. 6 shows the ELF for γ -TiAl and α_2 -Ti₃Al when a Nb atom is placed at a Ti and Al sites, respectively. The ELF measures the probability of finding an electron in the neighborhood of a reference electron with the same spin [72]. The bonding interaction is considered metallic if $\text{ELF} \leq 0.5$ and covalent if $\text{ELF} > 0.5$. Fig. 6a shows that in the region between Ti and Al atoms $\text{ELF} > 0.5$, suggesting a Ti(3d)-Al(3p) covalent bond. Similarly, the region between Ti and Ti atoms exhibits covalent Ti(3d)-Ti(3d) bonds. In γ (TiAl)-based alloys, Ti(3d)-Al(3p) covalent bonds with strong direction sensitivity are shown to increase the shear strength [77,78]. The ductility of γ (TiAl)-based alloys can be improved by adding alloying elements (Nb, Be, Si, V, Cr, Mn, Fe, Co, Ni, Zr, Mo) which in turn reduce the Ti(3d)-Al(3p) bonding or increases the Ti(3d)-Ti(3d) bonding [77,79]. In contrast to Fig. 6a, no significant changes are observed in Fig. 6b for electron localization in the vicinity of Nb atoms. However, Fig. 6c shows strong electron localization between Nb and neighboring Ti and Al atoms due to enhanced Nb(d)-Ti(d) and Nb(d)-Al(p) bonds. Similarly, for α_2 -Ti₃Al (Fig. 6d-f), Nb at Al sites shows strong electron localization (Fig. 6f) with the neighboring Al and Ti atoms due to the strengthening of Nb(d)-Ti(d) and Nb(d)-Al(p) bonds. Our observation agrees very well with [77], which reported that the addition of 4d transition elements (X) in TiAl systems strengthens both the X(d)-Ti(d) and X(d)-Al(p) bonds.

Figs. 7 and 8 show the PDOS (partial density of states) of γ -TiAl and α_2 -Ti₃Al, when a Nb atom is placed at a Ti and Al lattice site, respectively. Both show the existence of bond interactions between Ti(3d)-Al(3p) as their PDOS peaks at similar energy values. For γ -TiAl, Nb atoms reduce the electron density of Al(3p) electrons, whereas for α_2 -Ti₃Al the reduction is slightly more pronounced. Such reduction weakens Ti(3d)-Al(3p) interactions due to Nb explaining possibly the

increased ductility in γ -TiAl and α_2 -Ti₃Al. Fig. 7 shows a higher peak of the local density of 4d electrons of Nb atoms when placed at the Ti sites rather than Al sites in γ -TiAl, whereas the density is similar for α_2 -Ti₃Al. Our observation agrees with [26], which reported more 4d electrons of Nb to participate in the bond formation in γ -TiAl, thus, showing the site preference of Nb at Ti sites over Al sites.

Besides DFT calculations, we performed also MD simulations to study the effect of Nb atoms on the thermo-mechanical behavior of the target intermetallic phases. To this end, we investigated the effects of varying Nb concentrations (i.e., 0.0 - 5.0 at.%) on the lattice constants of γ -TiAl and α_2 -Ti₃Al. For γ -TiAl, Nb atoms at Ti sites increase the lattice constant 'a' but decrease 'c' (Fig. 18a in the appendix). This observation agrees with other previous works [8,59,71]. By contrast, for α_2 -Ti₃Al, our simulations showed negligible changes of the lattice constant 'a' but decreased 'c' due to Nb alloying (Fig. 18b in the appendix).

Fig. 9b (γ -TiAl) and 9d (α_2 -Ti₃Al) show computed displacements for selected Ti and Al atoms in the neighborhood of an Nb atom. For γ -TiAl, Ti and Al atoms move away from and towards Nb atoms, respectively, along the X and Y directions (Fig. 9). Along the Z direction, they move towards the Nb atoms. Note that the computed atomic displacements of neighboring Ti and Al atoms are in agreement with the previously discussed increase in the lattice constant 'a' and decrease in the lattice constant 'c' as shown in Fig. 18a. For α_2 -Ti₃Al, such direction-specific displacements could not be clearly distinguished since the neighboring Al and Ti atoms showed collective displacements (Fig. 9d), which explains the unchanged lattice constant 'a' observed in Fig. 18b.

Next, we simulated tension tests of single intermetallic phase models (models b, d in Fig. 1) using MD simulations at finite temperatures with varying Nb concentrations (0.0 - 5.0 at.%) to understand the effect of Nb on thermo-mechanical deformation behavior. The resulting stress-strain curves are plotted in Fig. 10 and Table 2 shows the computed UTS values. The enhanced fluctuations in the stress-strain curves for single phase γ -TiAl and α_2 -Ti₃Al may be attributed to pressure fluctuations resulting from lattice distortions due to high Nb alloying. Furthermore, it is worth mentioning that Farkas' ternary interatomic potential was originally developed for Nb-rich phases as host systems, whereas the

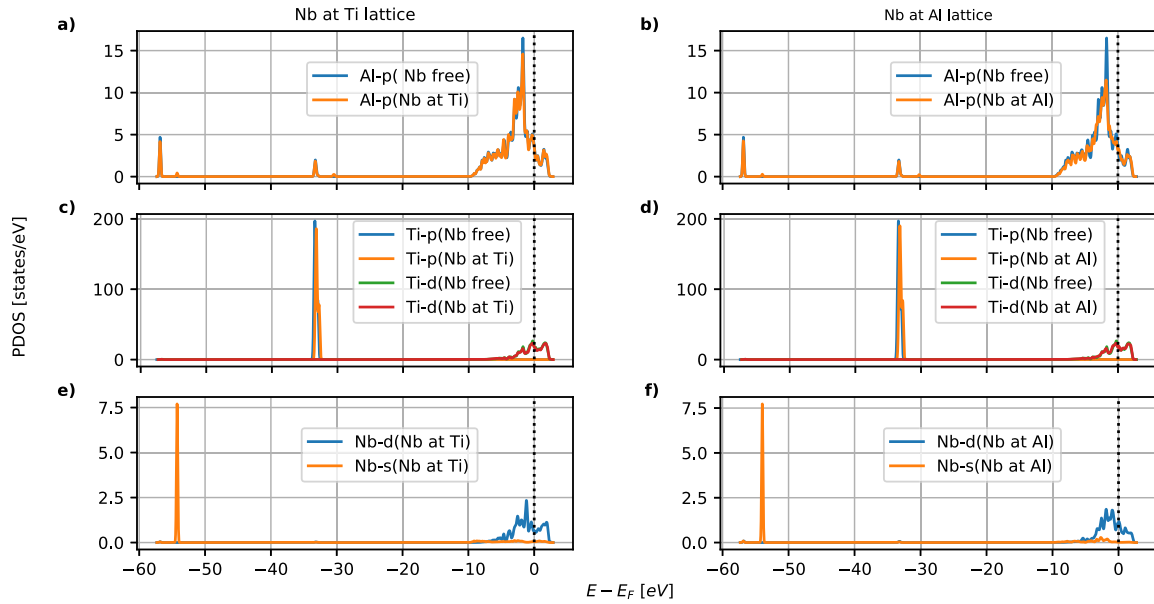


Fig. 7. Partial density of states (PDOS) of γ -TiAl with Nb at Ti site (left) and Nb at Al site (right) and PDOS of Al-p orbitals (top), Ti-p and Ti-d orbitals (middle) and Nb-d and Nb-s orbitals (bottom). The black dotted lines depicts the Fermi energy level.

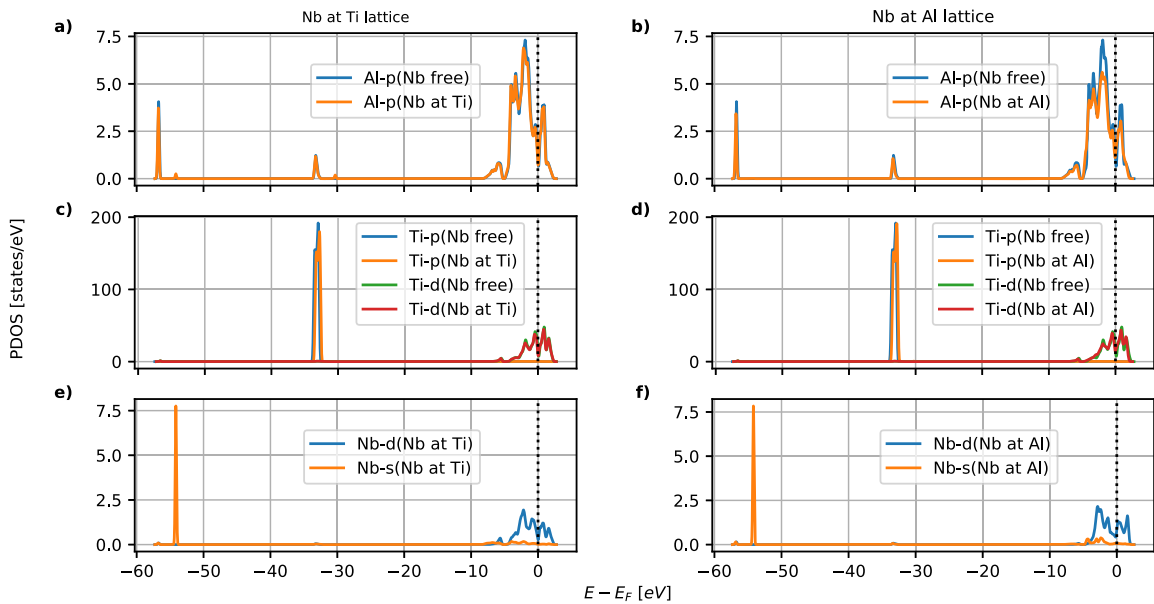


Fig. 8. Partial density of states (PDOS) of α_2 -Ti₃Al with Nb at Ti site (left) and Nb at Al site (right) and PDOS of Al-p orbitals (top), Ti-p and Ti-d orbitals (middle) and Nb-d and Nb-s orbitals (bottom). The black dotted lines depict the Fermi energy level.

Table 2

Computed ultimate tensile strength (UTS) of single phase γ -TiAl and α_2 -Ti₃Al with increasing Nb concentration.

Nb conc [at.%]	UTS of γ -TiAl [GPa]	UTS of α_2 -Ti ₃ Al [GPa]
0.0	15.33	10.39
1.0	11.76	8.19
2.0	10.69	7.04
3.0	10.26	6.14
4.0	10.29	5.30
5.0	9.69	4.42

current configurations are different from the intended target scenarios based on our preliminary studies. We observed that the UTS values of single phase γ -TiAl and α_2 -Ti₃Al decrease with Nb content (Table 2 and Fig. 10). Note that this observation requires careful interpreta-

tion because the simple single intermetallic phase models considered here were oriented in primitive orientations and were devoid of other complex features such as grain boundaries and secondary alloying elements.

In this subsection, our investigation yielded several interesting aspects of Nb in both γ -TiAl and α_2 -Ti₃Al. We evaluated the site preference using DFT, showing that Nb atoms prefer Ti lattice sites over Al sites. The ELF revealed the bonding of Nb to neighboring atoms in both γ -TiAl and α_2 -Ti₃Al. The PDOS plots (Fig. 7 and 8) provided a possible explanation for the enhanced ductility of γ -TiAl and α_2 -Ti₃Al resulting from Nb addition the weakening of Ti(3d)-Al(3p) interactions. Our MD simulations of single phase γ -TiAl and α_2 -Ti₃Al also aid in understanding the effect of Nb alloying (0.0 – 5.0 at.%) on lattice constants. Accordingly, in γ -TiAl, the lattice constants ‘a’ increased and ‘c’ decreased with Nb, whereas for α_2 -Ti₃Al, ‘a’ re-

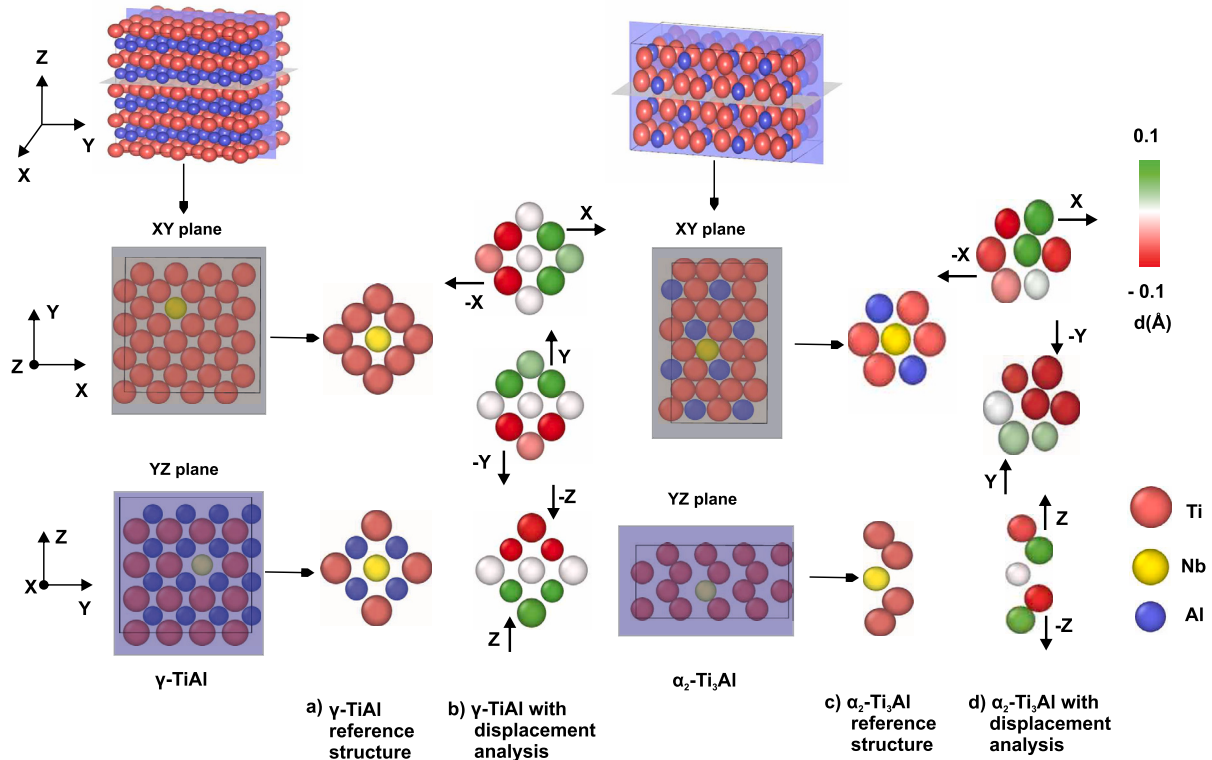


Fig. 9. Displacement profile of the atomic neighborhood in γ -TiAl and α_2 -Ti₃Al resulting from Nb introduction. Herein, a) and c) correspond to the reference structure of γ -TiAl and α_2 -Ti₃Al, whereas b) and d) denote the respective phases with displacement analysis. The atoms are color-coded as per their chemical types (Ti, Al, Nb), except for the displacement analysis (b, d), where the atoms are color-coded with their displacement (color bar).

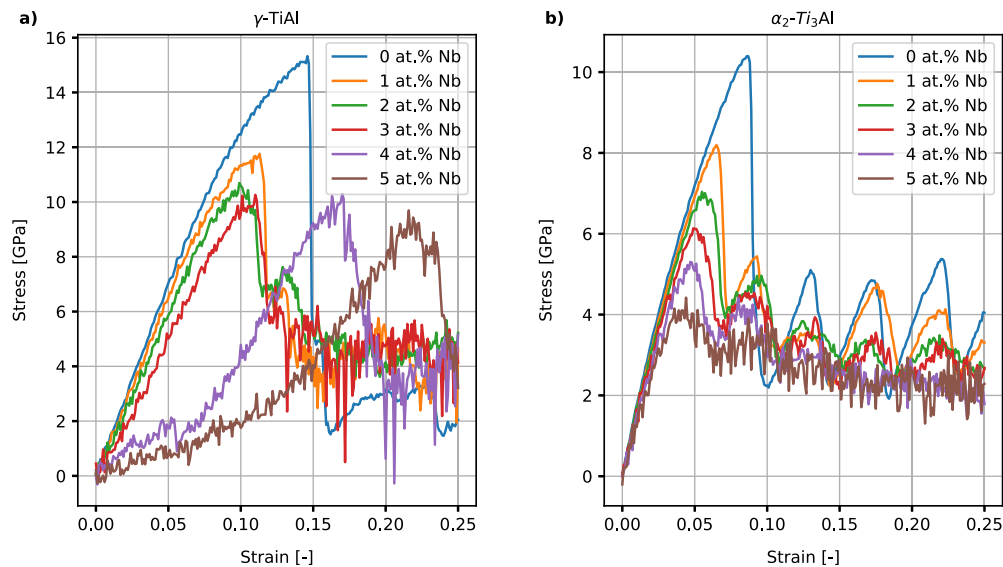


Fig. 10. Stress-strain curves for a) γ -TiAl and b) α_2 -Ti₃Al in simulated tension tests at room temperature with a strain rate of 10^9 /s using Farkas' [31] potential for different Nb concentrations (0.0 at.%, 1.0 at.%, 2.0 at.%, 3.0 at.%, 4.0 at.%, and 5.0 at.%).

mained nearly constant, but 'c' decreased. The displacement analysis of Nb's neighboring Ti and Al atoms in Fig. 9 underpins these observed changes. Understanding how the introduction of Nb affects lattice constants provides insights into the potential lattice distortions that may arise in simulations involving complicated models containing Nb. Our DFT calculations and atomistic MD simulations have contributed to a deeper comprehension of the site preference of Nb atoms, their influence on the lattice parameters, and their contribution to improved ductility in TiAl systems. These findings are

poised to play a crucial role in deciphering the intricate alterations induced by Nb addition within the upcoming complex atomistic models.

3.2. Thermo-mechanical behavior of TiAlNb

In this subsection, we aim to investigate the thermo-mechanical behavior of single-lamellar interfaces and more realistic MIAMs alloyed with Nb.

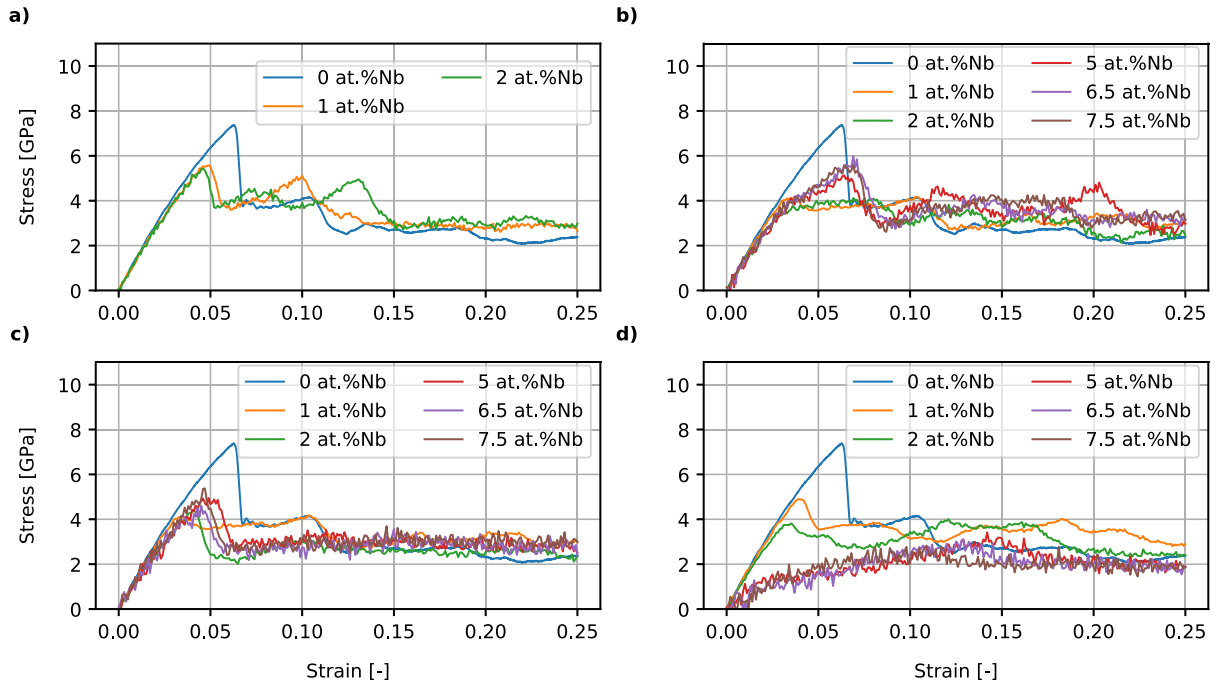


Fig. 11. The stress-strain curves for the γ/α_2 interface with Nb distributed a) near the Interface, b) in Phase B (α_2), c) in Phase A (γ), and d) homogeneously. Herein, the tension tests were performed at 700 K at a strain rate of 10^9 /s.

Table 3

Ultimate Tensile Strength [GPa] computed for the interface models with varying Nb concentrations from tension tests at 700 K and 900 K. For each interface model, we chose four sub-models with different spatial Nb distribution. Notations A, B, Interface, and Homogeneous denote Nb alloying in Phase A (γ -TiAl), Phase B (α_2 -Ti₃Al), γ_{PT} , γ_{TT} , γ_{RB}), at the interface and homogeneously distributed, respectively.

Model	Nb conc (at.%)	UTS at 700 K				UTS at 900 K			
		A	B	Interface	Homogeneous	A	B	Interface	Homogeneous
γ/α_2	0	7.36				6.30			
	1	5.54	4.17	5.57	4.89	4.92	3.80	4.94	4.25
	2	4.37	4.20	5.43	4.01	4.68	4.08	4.56	3.40
	5	4.95	5.13		3.41	4.65	4.60		2.63
	6.5	4.60	5.99		3.07	4.75	4.98		2.30
	7.5	5.38	5.58		2.83	4.88	4.36		2.06
γ/γ_{PT}	0	8.57				7.45			
	1	6.01	4.76	6.08	5.24	5.33	4.62	5.28	4.62
	2	5.52	5.72	5.95	4.88	4.90	4.87	5.17	4.13
	5	5.47	5.62		3.12	4.95	5.15		2.69
	6.5	4.82	5.65		2.73	4.77	5.28		2.41
	7.5	5.60	5.89		2.67	4.88	5.04		2.36
γ/γ_{TT}	0	8.78				8.16			
	1	5.71	5.98	6.84	5.36	5.28	5.46	6.72	4.60
	2	5.16	5.71	7.39	5.83	5.21	5.35	6.53	4.89
	5	5.79	6.08		3.93	5.65	5.93		3.05
	6.5	6.32	5.95		3.25	5.26	5.71		2.57
	7.5	6.48	6.19		3.10	5.40	6.38		2.27
γ/γ_{RB}	0	6.69				5.78			
	1	5.05	5.23	5.81	5.03	4.71	5.01	4.80	4.76
	2	4.43	4.99	5.38	4.78	3.92	4.51	5.00	4.26
	5	4.45	4.68		3.00	3.96	3.98		2.92
	6.5	4.64	4.82		2.92	4.43	4.59		2.42
	7.5	4.93	5.04		2.70	4.79	5.04		2.20

3.2.1. Effect of Nb in single-lamellar interface models

We alloyed four different single-lamellar interface models (one γ/α_2 and three γ/γ as shown e-h in Fig. 1) with varying Nb concentrations, i.e., 1.0, 2.0, 5.0, 6.5 and 7.5 (at.%). The atomistic models were initially equilibrated to target temperatures of 300 K, 500 K, 700 K, and 900 K. Then, we applied uniaxial loading conditions along the Z direction at

a strain rate of 10^9 /s. The resulting stress-strain curves for the γ/α_2 interface model are given in Fig. 11. Similar to our previous single-phase models, we observed fluctuations in stress-strain curves resulting from Nb alloying.

Table 3 shows the UTS values computed for the four interface models (Fig. 1) along with their subcategories a) Nb-rich phase/bulk, b)

Table 4

Computed total dislocation density (10^{17} m^{-2}) of single-lamellar interface model of γ/α_2 with increasing Nb concentration computed at 20 ps of the tensile simulations.

Nb conc (at.%)	A	B	Interface	Homogeneous
1 at. %	1.18	1.18	2.24	1.17
2 at. %	1.48	1.15	2.23	0.68
5 at. %	1.49	1.14		0.26
6.5 at. %	2.27	1.64		0.11
7.5 at. %	1.64	1.53		0.05

Nb-rich interface, c) Nb homogeneously distributed as discussed in subsection 2. For comparison, we included the strength of Nb-free models to evaluate the predictive ability of Farkas' potential. Among the Nb-free models, $\gamma/\gamma_{\text{TT}}$ showed superior strength followed by $\gamma/\gamma_{\text{PT}}, \gamma/\alpha_2$ and $\gamma/\gamma_{\text{RB}}$. In our previous work [39], the computed yield stress for single lamellar interface models showed the following trend $\gamma/\alpha_2 > \gamma/\gamma_{\text{PT}} > \gamma/\gamma_{\text{TT}} > \gamma/\gamma_{\text{RB}}$.

It is worth noting that our two works encompassing single-lamellar interface models contain considerable differences spanning atomistic model dimensions, lattice constants of the constituent intermetallic phases, i.e., γ, α_2 (different potentials), and of course, boundary conditions (uniform deformation of simulation box (this work) vs. boundary driven deformation (previous work)). Despite these differences in the simulation conditions, the observed relation showed a swap only between two interface models, i.e., γ/α_2 and $\gamma/\gamma_{\text{TT}}$. This swap in observed relation among the single interface model strength between our two works using Zope (previous [39]) and Farkas (this work) stems from the fact that Farkas potential showed a low UTS value for α_2 intermetallic phase. Herein, the UTS values obtained for α_2 -Ti₃Al differ between two interatomic potentials, whereas for γ -TiAl, the UTS values obtained were similar (Fig. 4). This difference in the UTS values from constituent lamellae contributes to the change in the hierarchy of the strength values of γ/α_2 .

Note that Farkas' potential combines two binary potentials (i.e., Nb-Ti and Ti-Al) and was mainly developed for the orthorhombic phase (Ti₂AlNb) so that some of the Nb-free models studied here may be challenging for Farkas' potential so that for such models Zope's potential may be more trustworthy.

Furthermore, Table 3 shows that each interface model followed the same hierarchy: Nb-rich interface (herein, for 1.0 at.% and 2.0 at.%) > Nb-rich phase/bulk > Nb homogeneously distributed. We observed that except for some of the homogeneously distributed Nb configurations and Nb-rich interfaces, the UTS values generally increased with Nb concentration (except when compared to 0.0 at.% Nb). Considering such irregularities, one has to keep in mind the limitations of our models. Our simulations of atomistic single interface models assume some degree of idealization by eliminating the contributions of grain boundaries, secondary alloying elements, processing-induced defects, and diffusion-dominated mechanisms. Also recall that we considered Nb substitution only at Ti sites based on site-preference energy obtained using DFT calculations. Here, we investigated atomistic models with Nb concentrations up to 7.5 at.% for both Nb-rich phase/bulk and homogeneously distributed Nb categories. However, only Nb concentrations until 2.0 at.% were feasible for Nb-rich interface models. Note that such transferability limit of the Farkas potential (until 2.0 at.% Nb) could have some implication on the physical fidelity; however, it is valid only for the special cases of atomistic models that contain high local Nb distribution in the vicinity of sharp boundaries, e.g., lamellar interfaces and colony boundaries. Due to this implication, simulating Nb-rich interface models above 2.0 at.% Nb were not feasible using Farkas potential, as shown in Table 3.

Fig. 12 sheds light on the nanomechanical events during the simulated uniaxial tension tests. We computed the atomic shear strain in the γ and α_2 phases of the single interface model without Nb (Fig. 12 a-d) and with Nb (Fig. 12 e-p). Some dislocations originated in the α_2 phase,

Table 5

Dislocation density of MIAMs for different Nb concentrations at 700 K.

Timesteps (ps)	Total dislocation density ($\times 10^{16} \text{ m}^{-2}$)		
	0 at.% Nb	0.5 at.% Nb	1 at.% Nb
0	4.5085	4.5974	5.8045
15	4.5657	4.6878	5.8630
30	4.6064	5.3535	8.2365
45	5.3364	7.2416	9.9968
60	7.2820	7.9126	10.5955
75	8.5360	8.6321	11.5630
90	9.4767	9.5895	12.1548

then slowly propagated to the interface and then to γ phase (yellow arrows). One possible explanation could be that the dislocation originating in the α_2 phase was due to the fact that Farkas' potential yields relatively low UTS values for the α_2 -Ti₃Al phase (Fig. 4). In Fig. 12 n-p, we could observe that the dislocations which originated in α_2 phase, while propagating to the γ phase, suffer obstructions at the interface and the remaining dislocations get dissipated/annihilated in the Nb-alloyed region of the α_2 phase. This indicates a strengthening effect, where the Nb atoms hinder the propagating dislocations. We could also observe regions of high strain (red regions in Fig. 12 l and p), resulting from dislocation gliding. From nanomechanical simulations as illustrated in Fig. 12, we extracted the total dislocation density of four types of a single interface γ/α_2 . As shown in Table 4, the dislocation density reflected (with just one exception) exactly the same increases and decreases observed for the UTS in Table 3. This underlines, that the differences in the UTS can be attributed to differences in dislocation density, and the previous discussion of Fig. 12 suggests that the reason for the different dislocation densities is a hindering effect of Nb atoms, for example, for dislocation propagation.

In this subsection, we investigated the influence of Nb alloying on the single-interface models and their variants. We found that a concentration of Nb atoms around the interface was particularly beneficial for the UTS compared to a wider distribution of the Nb atoms in one or both phases. The order of the UTS of the Nb-enriched interfaces was as follows: $\gamma/\gamma_{\text{TT}} > \gamma/\gamma_{\text{PT}} > \gamma/\alpha_2 > \gamma/\gamma_{\text{RB}}$. A nanomechanical analysis suggested that the hindering of dislocation propagation by Nb atoms around the interface may play a key role in the beneficial effect that Nb atoms around the interface have on the UTS. Generally, Nb alloying reduced the UTS compared to Nb-free models in the Nb concentration regime studied.

3.2.2. Effect of Nb alloying in MIAMs

We further investigated the effects of Nb alloying on microstructure-informed atomistic models (MIAMs). Here, we considered two global Nb concentrations (0.5 at.% and 1.0 at.%) by substituting Nb at random Ti sites. After equilibrating the simulation models to their target temperatures (300 K, 500 K, 700 K and 900 K), we simulated uniaxial loading along the Z direction at a strain rate of $10^9/\text{s}$.

Fig. 13 shows the stress-strain curves of MIAMs at selected temperatures and Nb concentrations. Similar to single intermetallic phases (section 3.1.2) and interface models (section 3.2.1), the UTS of MIAMs decreased with Nb alloying. Furthermore, Nb-free MIAMs at all temperatures showed UTS, followed by a considerable drop in stress at relatively low strains. In contrast, for MIAMs containing 1.0 at.% Nb, UTS was achieved with much higher strains and not followed by a similarly clear drop of stress, indicating improved ductility at the nanoscale. In other words, Nb alloying delayed the failure point in MIAMs, particularly at high temperatures and high Nb concentrations (1 at.%).

To unravel the nanomechanical origin of improved ductility (enhanced dislocation activities) with Nb alloying, we computed the dislocation density of MIAMs both qualitatively (Fig. 14) and quantitatively (Table 5) during the simulated tension tests. Table 5 shows the increasing dislocation densities with Nb computed at selected time steps during

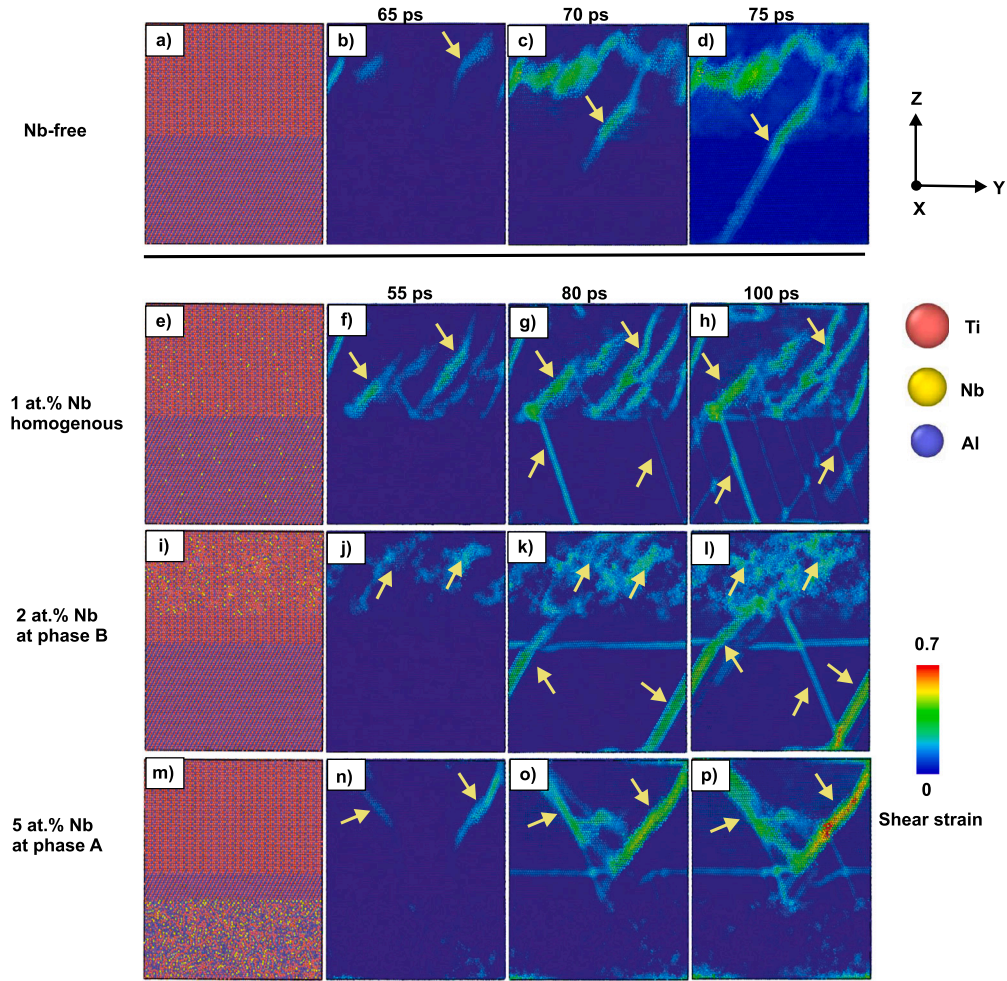


Fig. 12. Atomic shear strain distribution of γ/α_2 interface models from tension tests (strain rate: 10^9 /s, $T = 700$ K) at selected time steps for a Nb-free model (a-d), 1.0 at.% Nb homogeneously distributed (e-h), 2.0 at.% Nb distributed in phase A, i.e., α_2 -Ti₃Al (i-l), and 5.0 at.% Nb distributed in phase A, i.e., γ -TiAl (m-p). The yellow markers denote the dislocations.

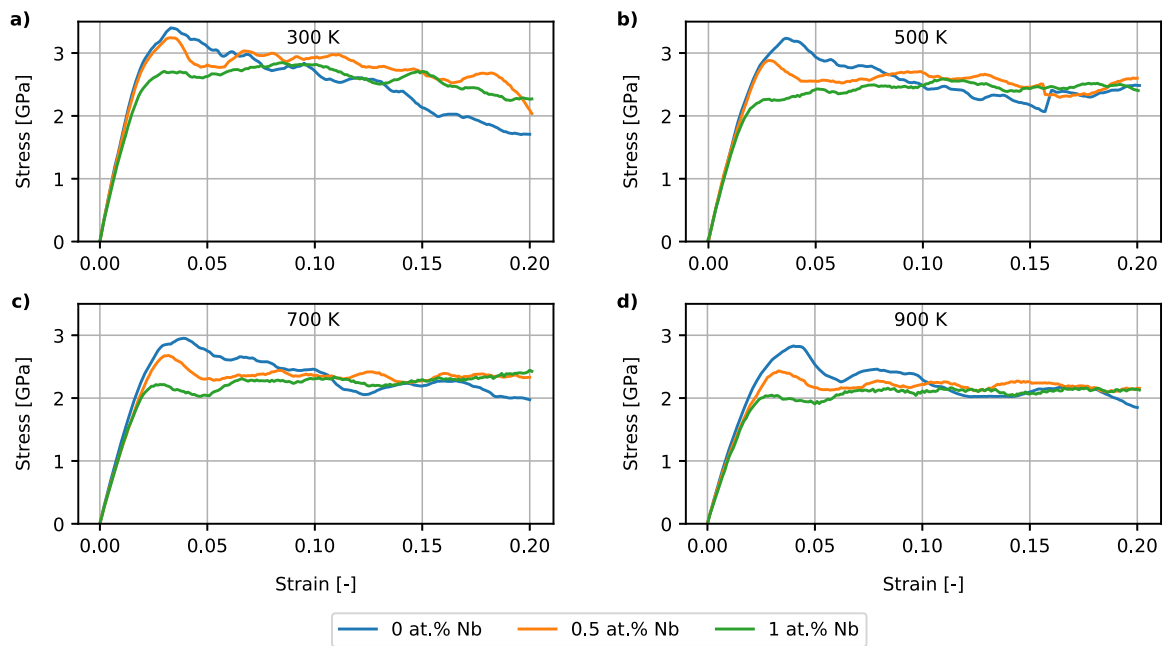


Fig. 13. The stress-strain curves for MIAMs with 0.0% Nb, 0.5% Nb and 1.0% Nb from tension tests with a strain rate of 10^9 /s at selected temperatures (300 K, 500 K, 700 K, and 900 K.)

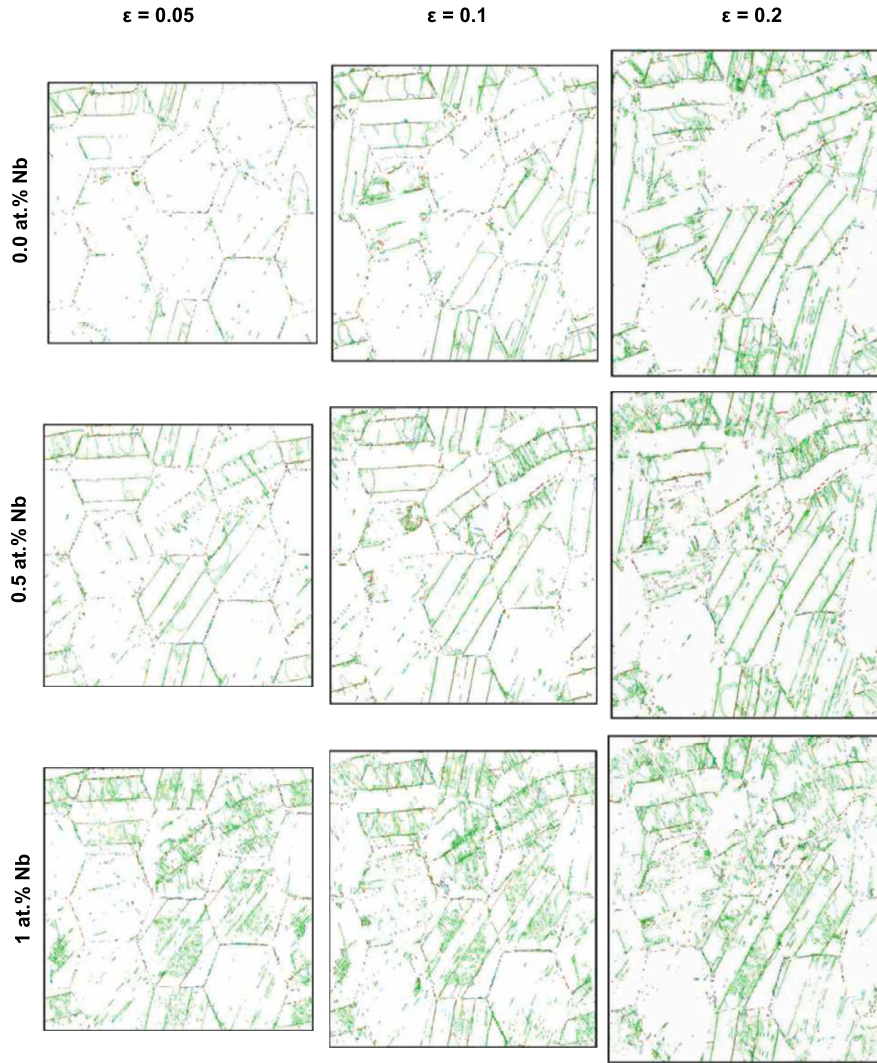


Fig. 14. Dislocation evolution in MIAMs with varying Nb concentrations of 0.0 at.% Nb (top), 0.5 at.% Nb (middle) and 1.0 at.% Nb (bottom) under thermo-mechanical loading at 900 K observed at strain values of 0.05 (left), 0.1 (center) and 0.2 (right).

increasing tension at 700 K. Note that the relative increase in dislocation density remains high with Nb alloying, explaining improved ductility with Nb addition. Fig. 14 shows that the Nb-free model exhibits at large strains mostly interface-dominated dislocations, whereas for increasing Nb content, more intra-lamellar dislocations are observed. Our MIAMs (1.0 at.%) showed that Nb facilitated the early onset of dislocations even at a much lower strain of $\epsilon = 0.05$ at 900 K. Nb alloying delayed the failure point in MIAMs, particularly at high temperatures and high Nb concentrations (1 at.%). Fig. 15 shows the interplay between Nb alloying and temperature in reducing the occurrence of intergranular fracture (the reason behind the stress drop after UTS) along the colony boundaries. Apparently, the fracture is delayed, and dislocation activities are increased both by Nb alloying and increasing temperature.

Stress-induced transformation of γ (face-centered tetragonal [FCT] atoms) to α_2 (hexagonal close packed [HCP] atoms) in TiAl-based alloys was studied in several works [80–83]. Note that the stress-induced α_2 phase has different lattice parameters compared to the ordered hexagonal α_2 (DO_{19}) phase. As we simulated tension with Nb-free MIAMs, stress-induced $\gamma \rightarrow \alpha_2$ transformation occurred in most γ phases and their variants γ_{PT} , γ_{TT} , γ_{RB} . We computed the volume fraction of HCP atoms to analyze this transformation (Table 6). As shown in Table 6, the HCP volume fraction decreased for γ/γ_{RB} and γ/α_2 with an increase in Nb concentration, whereas a reverse trend was observed for both γ/γ_{TT} and γ/γ_{PT} colonies. [80] observed a similar discrepancy in the trend re-

Table 6

HCP structure volume fraction of MIAMs during tension tests (strain rate = 10^9 /s, T = 700 K.) for different points in time with Nb concentrations.

MIAM colony type	Nb concentration [at. %]	Simulated time [ps]		
		30	60	90
γ/γ_{RB}	0	10.5	14.6	15.2
	0.5	10.5	14.4	15.0
	1	8.5	12.4	14.8
γ/α_2	0	43.6	41.5	39.4
	0.5	40.1	39.9	38.8
	1	38.5	39.3	36.3
γ/γ_{TT}	0	7.6	7.4	8.7
	0.5	9.1	10.9	13.6
	1	7.4	11.7	15.9
γ/γ_{PT}	0	10.1	14.2	14.4
	0.5	10.2	15.1	16.1
	1	11.2	17.2	19.7

porting that the stress-induced $\gamma \rightarrow \alpha_2$ transformation primarily occurred at twin intersections and in twinned regions.

To examine whether this phenomenon occurs also in our simulations, we focus specifically on the colonies representing each case γ/γ_{TT}

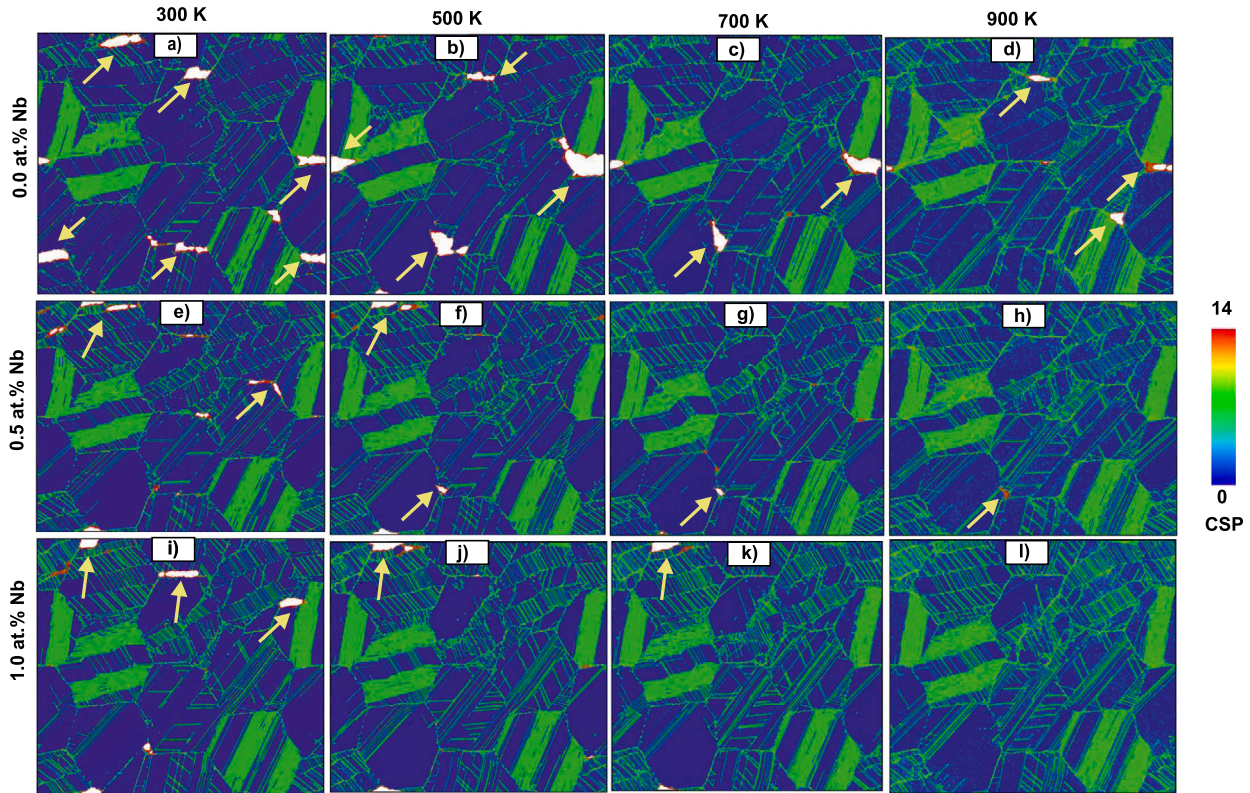


Fig. 15. MIAMs with Nb concentrations of 0.0 at.% Nb (top), 0.5 at.% Nb (middle) and 1.0 at.% Nb (bottom) deformed at temperatures of (from left to right) 300 K, 500 K, 700 K, and 900 K after 200 ps. The yellow markers indicate the intergranular fracture along the colony boundaries.

and γ/γ_{RB} lamellae. In Fig. 16, the microstructure evolution of the γ lamella and the adjacent γ_{TT} lamellae with varying Nb concentrations are shown. During simulated tension, MIAMs with Nb exhibited several dislocation gliding events and twinning (black arrows in Fig. 16), thus contributing to the enhanced HCP volume fraction in these colonies. Nb alloying facilitates the stacking fault formation, and the dislocations glided parallel and oblique to the lamellar interfaces as shown in Fig. 16 d, k, and l. MIAMs with Nb exhibited significantly more dislocations and twins at the same strain. An enlarged view of one of the twin boundaries (γ region of γ_{TT} grain) is shown in Fig. 16 k. The black arrows in the figure correspond to the twin boundary region, while all other HCP regions correspond to the stacking faults traced by dislocation gliding.

Fig. 17 shows the γ/γ_{RB} colony at various Nb concentrations. To obtain nanomechanical insights, we focused here on a single colony of a MIAM and it is observed that the γ_{RB} region of this colony has undergone a phase transformation to α_2 (several layers of HCP atoms). The same regions in the colony during the simulations were compared to study the slow phase transformation in a sequence like 1a, 1b, and 1c, etc. Herein, Nb alloying hampered $\gamma \rightarrow \alpha_2$ transformation, thus retaining more activation regions of γ for dislocation activities to enhance the plasticity at the nanoscale, as observed in Fig. 14. Note in this respect, in particular Fig. 17 3c - 6c underlining a delayed α_2 formation by Nb.

Studying the γ/γ_{RB} lamellae (Fig. 17), we noticed stacking-faults dominated deformation with some blade-shaped bundles perpendicular to lamellar interfaces. TEM investigations of the deformed samples [84] reported similar blade-shaped bundles and stacking faults along the lamellar boundaries governing the plastic deformation. For both these mechanisms, gliding of $1/6[1\ 1\ \bar{2}]$ partial dislocations on $\{1\ 1\ 1\}$ planes act as a precursor, as observed in our MD simulations. Fig. 16 shows increasing stacking fault bundles from lamellar interfaces with Nb to relieve stress concentration. Our MD simulations (Figs. 16 and 17) showed the glide of $1/6\langle 1\ 1\ 2 \rangle$ Shockley partial dislocations on the alternate $(1\ 1\ 1)$ plane induced the phase transformation of $\gamma \rightarrow \alpha_2$, re-

sulting in more α_2 laths. Wen et al. [85] reported similar stress-induced γ to α_2 phase transformation of TiAl-based alloys at room temperature.

The interplay between the thermal driving force (temperature effects) and Nb solutes plays a crucial role in the deformation behavior. For instance, the Nb concentrations at room temperature did not improve the ductility, as we observed intergranular fracture near colony boundaries (Fig. 15 a, e, and i), whereas the combination of high temperature and Nb concentration stimulated the dislocation slipping and twinning. These observations align with the previous arguments that several deformation mechanisms coexist in TiAl depending on the lamellae variants and their corresponding interfaces, colony orientation, Nb alloying, and temperature. In some regions (γ_{RB} , γ/α_2), Nb solutes retained the γ region to offer more dislocation sources – by retarding $\gamma \rightarrow \alpha_2$ transformation – supported by higher temperatures. In other twinned regions (γ_{PT} , γ_{TT}), Nb alloying promoted the early onset of dislocation activities at much lower strain, enhancing the ductility at the nanoscale and supported again by temperature.

Generally, in atomistic thermo-mechanical simulations of MIAMs we observed a poor room temperature ductility. In particular, Nb-free MIAMs exhibited intergranular fracture along the colony boundaries (Fig. 15). Nb alloying delayed the crack's nucleation and propagation by relieving the stress concentrations through dislocation nucleation, propagating further into adjacent colonies, thus improving the deformability. Nb alloying triggered the early onset of nanomechanical events like dislocation gliding and twinning even at moderate temperature by lattice distortions and reduced stacking fault energy, especially in the γ region and twinned lamellae variants. CNA analysis revealed that Nb alloying and temperature distorted lamellae inside the colonies, enhancing the deformability at high strains (Fig. 17). However, with increasing Nb, colonies with α_2 showed significant deformation, such as stepping behavior and colony restructuring resulting in serrated colony boundaries. Despite considering only low Nb concentrations in MIAMs, Nb enhanced the ductility of TiAl alloys, specifically at high temperatures. Herein, MD simulations of MIAMs revealed that several deformation

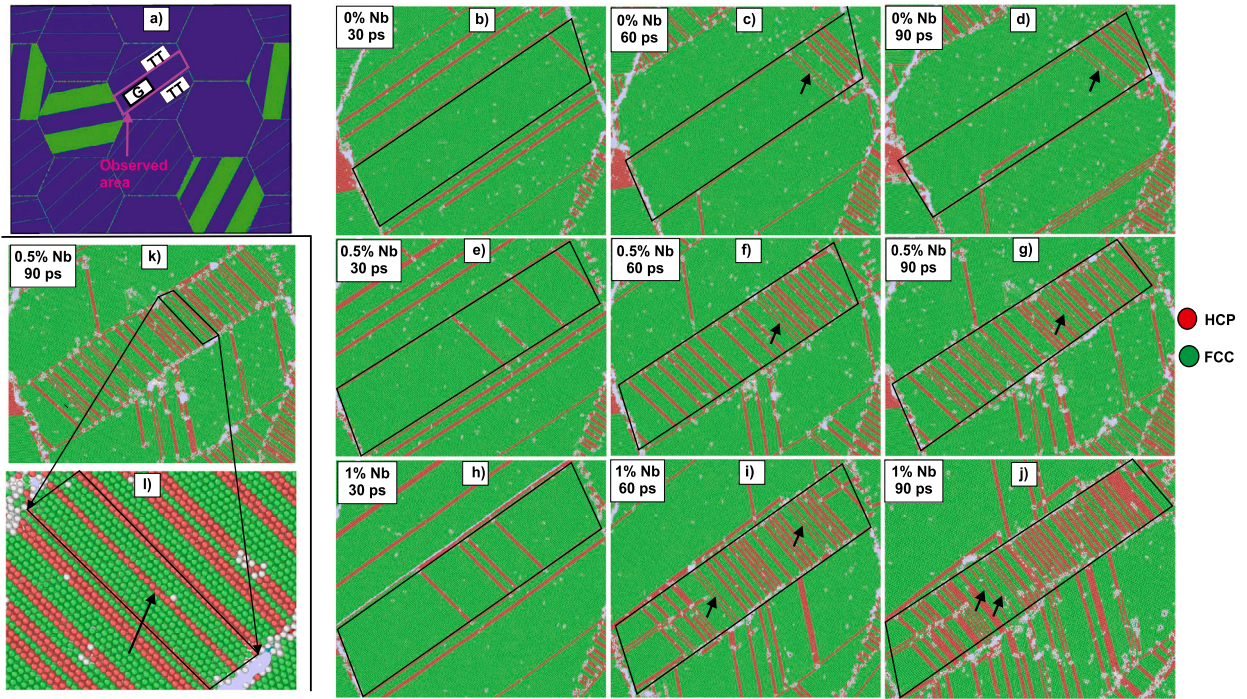


Fig. 16. The formation of mechanical twins in a γ/γ_{TT} colony of a MIAM (a) with 0.0% Nb, 0.5% Nb, and 1% Nb at (from left to right) 30 ps, 60 ps and 90 ps during tension tests at a rate of 10^9 /s. The situation for 0.5% Nb at 90 ps is depicted in detail in (k) and (l).

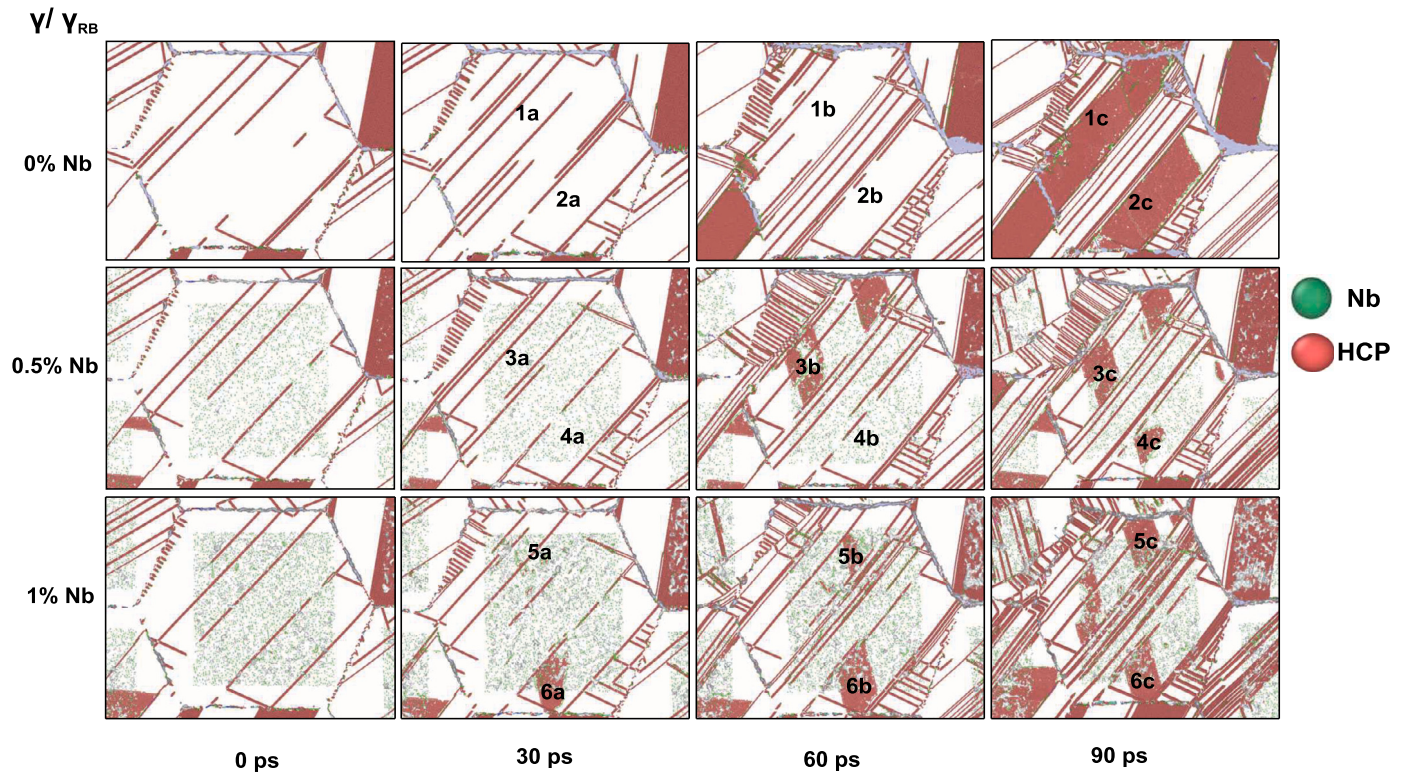


Fig. 17. Retardation of stress-induced $\gamma \rightarrow \alpha_2$ transformation observed in γ/γ_{RB} colony of MIAM with Nb concentrations of 0.0 at.% Nb (top), 0.5 at.% Nb (middle) and 1.0 at.% Nb (bottom) at (from left to right) 0 ps, 30 ps, 60 ps and 90 ps during tension test at a strain rate of 10^9 /s and at a temperature of 700 K.

mechanisms coexist depending on the volume fraction of lamellae variants and their corresponding interfaces, colony orientation, Nb alloying, and temperature. For instance, Nb hampered the stress-induced $\gamma \rightarrow \alpha_2$ transformation in γ_{RB} and γ/α_2 region to offer more dislocation sources, whereas, in other twinned regions (γ_{PT} , γ_{TT}), Nb facilitated

dislocation activities at low strains resulting in some finer lamellae increasing the interface volume fraction. Note that our study supports that Nb addition promotes dislocation gliding and twinning in TiAl-based alloys as it lowers the stacking fault energy of γ -TiAl [1]. Experimental estimates suggest a typical Nb concentration of 5 - 10 at.% to achieve

improved mechanical properties, exhibiting superior strength and ductility.

MIAMs with Nb showed microstructure refinement (fine lamellae creation) due to reduced stacking fault energy as a precursor for dislocation formation. Generally, the UTS values decreased with Nb concentration for all the atomistic models (i.e., single phase models, single lamellar interface models, MIAMs) studied herein with some exceptions, as discussed in 2.1.3. Furthermore, the computed dislocation densities were consistent with the observed trend in UTS, indicating a strong correlation between them. In single-lamellar interface models, the hindrance of dislocation motion by Nb atoms around the interface explains their effects on the UTS. To this end, the proximity of Nb atoms to microstructure boundaries (e.g., interfaces) appeared to play an important role, and similarly the ratios of twin boundaries and phase boundaries. Increasing dislocation activities with increasing Nb concentration suggest that improved mechanical properties stem from the microstructural changes rather than solid-solution strengthening [86]. Note that our MD simulations revealed that Nb alloying contributed to significant lattice distortions, further influencing the dislocation evolution under thermomechanical deformation. However, we strongly emphasize the need for future studies with mixed site models, high Nb concentrations, secondary disordered phases and other similar complex interfaces, and other interatomic potential to ensure the generality of the conclusions drawn. The atomistic models studied showed a decreasing UTS with Nb alloying, which may be attributed to the lattice distortions. However, the strain corresponding to the UTS increased with Nb alloying, particularly at high temperatures (900 K) for the MIAMs (1.0 at.% Nb) studied herein. Thus, combining our insights, Nb alloying improved ductility to some extent at the cost of strength [86].

4. Conclusions

This work investigated the effect of Nb alloying on the strength and ductility of TiAl-based alloys under thermo-mechanical conditions using atomistic simulations. Using single-phase models, we first confirmed the transferability of Farkas' ternary potential in our study by computing quantities such as elastic constants, point defect formation energies, and cohesive energies and by comparing them to two other binary interatomic potentials for TiAl-based alloys. It turned out that Farkas' ternary potential can capture also the behavior of binary intermetallic phases (i.e., γ -TiAl and α_2 -Ti₃Al). This ensured us that we could use it also in studies where in the same type of model, the Nb concentration was increased from zero (binary system) to a substantial fraction (ternary system).

Our DFT calculations reaffirmed a known site preference of Nb at Ti sites over Al sites. Analyzing the PDOS suggested a weakening of Ti(3d)-Al(3p) interactions by Nb addition resulting in increased ductility in γ -TiAl and α_2 -Ti₃Al.

Both in interface models and more complex microstructure-informed atomistic models (MIAMs), we observed that the ultimate tensile strength (UTS) was decreased by Nb. However, the maximal strain associated with UTS was increased by Nb due to the associated higher ductility. Our molecular dynamics (MD) simulations identified several nanomechanical mechanisms behind this effect, such as the delay of fracture onset by increased dislocation formation and propagation that relieved some stress without fracture. Also, Nb was found to hamper the stress-induced $\gamma \rightarrow \alpha_2$ transformation in γ_{RB} and γ/α_2 , hence favoring a situation with more dislocation sources. In other twinned regions (γ_{PT} , γ_{TT}), Nb facilitated dislocation activities at low strain, resulting in finer lamellae increasing the interface volume fraction.

Our atomistic simulations underlined the important role of microstructure boundaries (i.e., lamellar interfaces and colony boundaries) because many of the mechanisms how Nb altered mechanical properties appeared closely related to microstructural boundaries. They thus present an important means of optimizing the thermo-mechanical properties of TiAl-based alloys. At low Nb concentrations, we obtained

the novel insight that Nb assists in delaying the $\gamma \rightarrow \alpha_2$ phase transformation to improve ductility, besides the widely reported solid-solution strengthening (dislocations pinned/retarded by Nb) and microstructure changes.

For the low Nb concentrations we studied, we found that Nb helped to improve ductility, though to some extent at the cost of strength. The experimental investigations suggest that a high Nb concentration of 5 - 10 at.% leads to both high strength and ductility. Examining whether or how the thermo-mechanical properties change at higher Nb concentrations would be an important next step but may first require the development of enhanced interatomic potentials that can handle higher Nb fractions for atomistic models with complex microstructure.

CRediT authorship contribution statement

Anju Chandran: Conceptualization, Data curation, Investigation, Methodology, Software, Visualization, Writing – original draft. **Hari-prasath Ganesan:** Conceptualization, Funding acquisition, Investigation, Methodology, Project administration, Supervision, Visualization, Writing – review & editing. **Christian J. Cyron:** Funding acquisition, Supervision, Validation, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A

Additional information on [32]: the Kim potential is a second nearest neighbor Modified Embedded Atomic Model (MEAM). This potential reproduced the lattice parameters with errors within 0.5%. The Kim and Zope potential produced comparable cohesive energy values. The elastic constants C_{11} and C_{12} calculated using the Kim potential showed good agreement with the experimental values.

Additional information on [33]: Xiang and Guo [33] proposed a ternary potential of NbAlTi systems based on a MEAM. The authors have studied TiAlNb systems with Nb concentrations up to 10.0 at.%. Here the site of substitution of Nb was a mixture of both Ti and Al lattice sites. They studied the melting point and high-temperature deformation properties of γ and α_2 systems containing Nb and computed the elastic moduli and generalized stacking faults (GSFs) of Nb-alloyed γ and α_2 . The high-temperature deformed structure contained stacking faults (SFs) and twins, and they compared the stress-strain curve for various Nb concentrations.

Our observation on the lattice constant (Fig. 18) 'a' differs from [61], where they observed an increase in 'a' with Nb (8 at.%) for α_2 -Ti₃Al. We believe that the discrepancy may stem from the difference in Nb concentration and Nb substitution sites assumed for alloying (i.e., our work considered only Ti sites and [61] might have considered both Ti and Al sites but not explicitly stated).

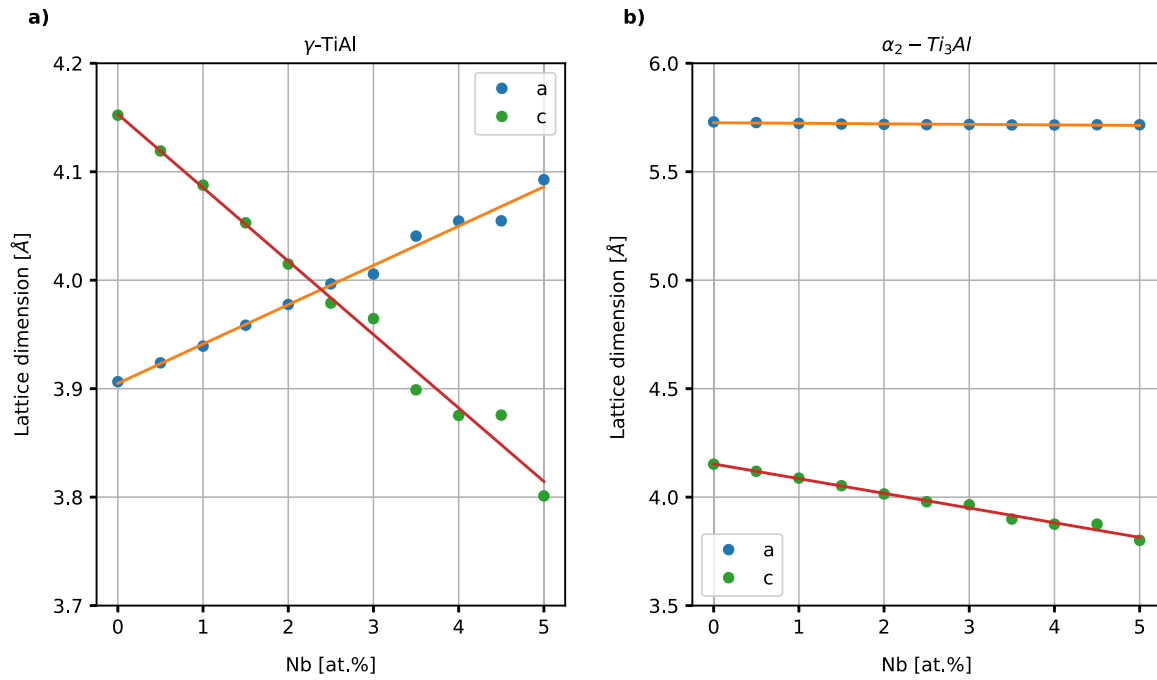


Fig. 18. The change in lattice parameters 'a' and 'c' with increasing Nb concentration in a) γ -TiAl and b) α_2 -Ti₃Al computed using Farkas' ternary potential [57].

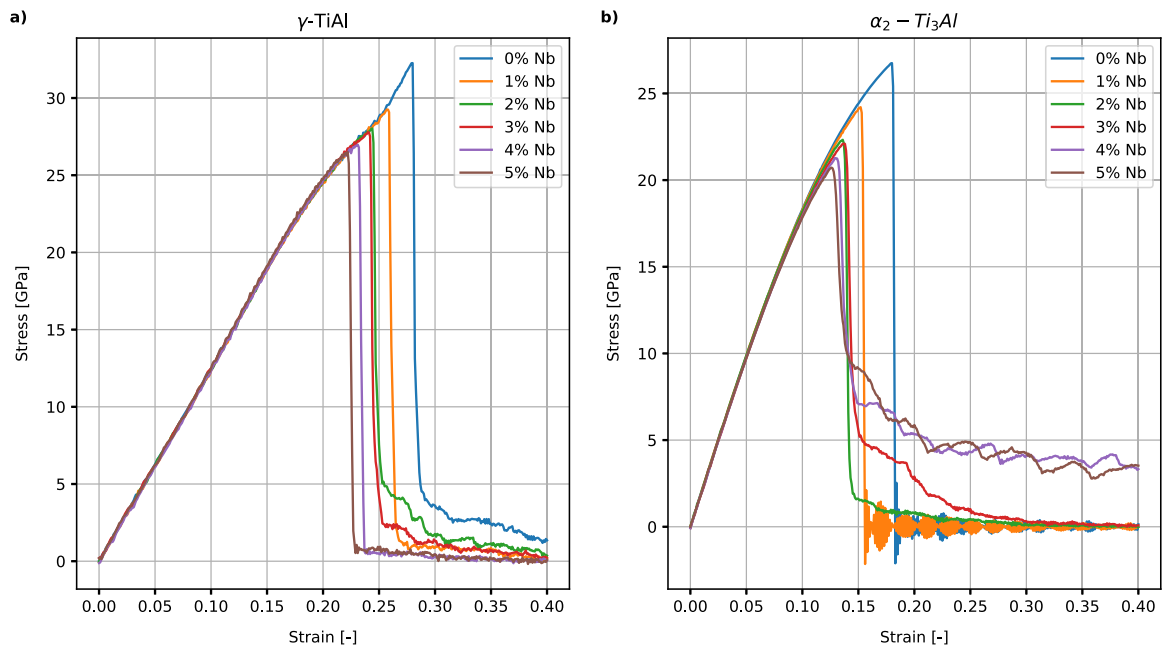


Fig. 19. Stress-strain curves for single phase systems with varying Nb concentrations using Xiang's interatomic potential [33].

We simulated tension tests to evaluate the mechanical behavior of single phase models of γ -TiAl and α_2 -Ti₃Al using Xiang potential [33] (refer Fig. 19). We applied a strain rate of 10^9 /s and the models were equilibrated at 300 K. The stress-strain curves present a contradiction to our previous observation (Fig. 10) using Farkas potential [31]. In our current study, we noted a decrease in UTS with increasing Nb concentration. Xiang potential [33] showed a decreasing trend in both UTS and ductility upon Nb introduction. However, this observation requires further investigation using Xiang potential, which is beyond the scope of this article.

We simulated tension tests on the interface models with varying Nb concentrations (1.0, 2.0, 5.0, 6.5, and 7.5 (at.%) using Xiang potential (Table 7). We applied a strain rate of 10^9 /s, and the models were equi-

librated at 700 K. The UTS showed a comparable trend to that observed in interface models using Farkas potential (refer 3.2.1).

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Table 7

Ultimate Tensile Strength [GPa] computed for the interface models with varying Nb concentrations from tension tests at 700 K using Xiang's potential [33]. For each interface model, we chose four sub-models with different spatial Nb distributions. Notations A, B, Interface, and Homogeneous denote Phase A (γ -TiAl), Phase B (α_2 -Ti₃Al, γ_{PT} , γ_{TT} , γ_{RB}), interface and homogeneous, respectively.

Model	Nb conc (at.%)	700 K			
		A	B	Int	Total
	0	18.20			
γ/α_2	1	18.17	15.19	11.43	16.53
	2	16.06	12.51	10.05	15.13
	5	13.09	10.86		14.67
	6.5	11.61	9.49		14.33
	7.5	11.19	8.68		13.91
	0	20.07			
γ/γ_{PT}	1	19.35	15.10	15.74	21.33
	2	17.08	12.85	12.38	15.69
	5	12.37	9.72		18.19
	6.5	12.61	7.92		16.17
	7.5	10.64	9.26		14.68
	0	21.56			
γ/γ_{TT}	1	18.77	17.47	14.61	18.44
	2	16.67	14.48	11.20	17.93
	5	12.37	9.72		18.19
	6.5	11.32	8.96		15.27
	7.5	11.08	8.35		14.65
	0	21.66			
γ/γ_{RB}	1	20.72	14.14	17.65	15.78
	2	18.59	14.01	13.45	15.10
	5	14.21	12.06		12.35
	6.5	12.96	12.32		11.39
	7.5	12.31	11.17		11.84

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A.2 Publication 2

Comparative analysis of ternary TiAlNb interatomic potentials: moment tensor vs. deep learning approaches

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Comparative analysis of ternary TiAlNb interatomic potentials: moment tensor vs. deep learning approaches

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Intermetallic titanium aluminides, leveraging the ordered γ -TiAl phase, attract increasing attention in aerospace and automotive engineering due to their favorable mechanical properties at high temperatures. Of particular interest are γ -TiAl-based alloys with a Niobium (Nb) concentration of 5–10 at.%. It is a key question how to model such ternary alloys at the atomic scale with molecular dynamics (MD) simulations to better understand (and subsequently optimize) the alloys. Here, we present a comparative analysis of ternary TiAlNb interatomic potentials developed by the moment tensor potential (MTP) and deep potential molecular dynamics (DeePMD) methods specifically for the above mentioned critical Nb concentration range. We introduce a novel dataset (TiAlNb dataset) for potential training that establishes a benchmark for the assessment of TiAlNb potentials. The potentials were evaluated through rigorous error analysis and performance metrics, alongside calculations of material properties such as elastic constants, equilibrium volume, and lattice constant. Additionally, we explore finite temperature properties including specific heat and thermal expansion with both potentials. Mechanical behaviors, such as uniaxial tension and the calculation of generalized stacking fault energy, are analyzed to determine the impact of Nb alloying in TiAl-based alloys. Our results indicate that Nb alloying generally enhances the ductility of TiAl-based alloys at the expense of reduced strength, with the notable exception of simulations using DeePMD for the γ -TiAl phase, where this trend does not apply.

KEYWORDS

TiAlNb alloy, machine-learning interatomic potentials, deep learning, moment tensor, molecular dynamics, density functional theory

1 Introduction

Gamma titanium aluminide (γ -TiAl) intermetallic alloys attract increasing attention in aerospace and automotive engineering as high-performance lightweight structural materials. This growing interest is primarily due to their unique combination of low density, remarkable oxidation resistance, and superior strength and creep resistance at elevated temperatures Appel et al. (2011). Key to these alloys are the primary intermetallic phases: γ -TiAl, with its ordered face-centered tetragonal structure ($I1_0$, $P4/mmm$) and α_2 -Ti₃Al, noted for its ordered hexagonal structure ($D0_{19}$, $P6_3/mmc$). The advancement of

γ -TiAl based alloys, particularly those tailored for higher service temperatures, such as TNM (TiAl-Nb-Mo) and TNB (TiAl-Nb-B) alloys, has been significant (Appel et al., 2011; Clemens and Mayer, 2012; Li et al., 2014; Klein et al., 2016; Zhang et al., 2016). These alloys typically contain 5–10 at.% Nb, along with small amounts of other elements like Mo, B, C, Si, W, Cr, Ta. Extensive research highlights Nb's crucial role in boosting mechanical properties, notably increasing the strength and ductility of TiAl alloys (Appel et al., 2011; Clemens and Mayer, 2012; Liu et al., 2002; Song et al., 2020; Cheng et al., 2016; Liu et al., 2022; Zhang et al., 2023). Despite these improvements, the specific contribution of Nb to the enhancement of strength and ductility in TNB and TNM alloys has remained somewhat unclear. In our previous work (Chandran et al., 2024), we addressed this gap by examining the effects of Nb on the thermo-mechanical properties of TiAl-based alloys through atomistic simulations. Utilizing Farkas' ternary interatomic potential (Farkas and Jones, 1996), we scrutinized various TiAl-based models, ranging from single-phase structures to single lamellar interfaces, and progressing to more complex microstructure-informed atomistic models (MIAMs) with nanoprecipitates. However, it became evident that it is a limitation of Farkas' potential that it could not adequately handle Nb concentrations above 1 at.% in MIAMs and 2 at.% in certain types of single lamellar interfaces. This limitation is significant, as higher Nb concentrations (5–10 at.%) are known to be most interesting for improving strength and ductility. To overcome this challenge, we pursue in this article the direction of developing machine learning (ML)-based interatomic potentials for molecular dynamics (MD) simulations.

Generally, MD simulations offer profound insights into the behavior of atomic systems, ranging in scale from around 10^3 – 10^9 atoms. These simulations are instrumental in capturing a wide array of interactions, including thermal, mechanical, chemical, and microstructural dynamics. However, the accuracy of MD simulations is contingent upon the selection of appropriate interatomic potentials or force fields, as well as the boundary conditions implemented. In recent years, the integration of machine learning with MD simulations has emerged as a rapidly evolving field. This integration primarily focuses on the modeling of interatomic potential energy surfaces (PES) using reference data derived from *ab initio* simulations. Various ML methodologies have made significant contributions to the study of condensed matter systems and can be broadly categorized into linear regression [e.g., moment tensor potentials (Shapeev, 2016; Podryabinkin and Shapeev, 2017; Gubaev et al., 2019)], kernel methods [e.g., gaussian approximation potential (Bartók et al., 2013; Szlachta et al., 2014; Dragoni et al., 2018), spectral neighbor analysis (Thompson et al., 2015; Chen et al., 2017; Li et al., 2018; Deng et al., 2019)], and deep neural network-based techniques (Behler and Parrinello, 2007; Behler, 2011; Zhang et al., 2018a), respectively.

Recent comparative studies have evaluated the effectiveness of these diverse techniques, as seen in the works of Zuo et al. (2020); Deringer et al. (2019); Unke et al. (2020, 2021). Among these, deep neural network-based potentials, particularly the deep potential (DP) and neural network potential (NNP), stand out due to their successes in modeling both ordered and disordered systems. The flexibility of the descriptor proposed by Zhang et al.

(2018a) for DP potentials is particularly noteworthy. The DP method has demonstrated its efficacy in various systems, such as LiF and FLiBe (Rodriguez et al., 2021), MgCl_2 -NaCl and MgCl_2 -KCl (Xu et al., 2023), AlN (Li et al., 2024), Cu (Du et al., 2022) and several others (Niu et al., 2020; Nguyen et al., 2022). In comparison, the moment tensor potential (MTP) is notable for its efficiency, derived from a polynomial basis of interatomic distances and angles. MTP not only outpaces the other methods in terms of speed but has also demonstrated equivalent accuracy in modeling various material systems, as shown in studies by Novikov et al. (2018); Podryabinkin et al. (2019); Novoselov et al. (2019). MTP has been recognized for its optimal balance between accuracy and computational efficiency, a comparison elucidated in Zuo et al. (2020) performance analysis. Tasnádi et al. (2021) developed an MTP potential for efficiently predicting the elastic properties of $\text{Ti}_{0.5}\text{Al}_{0.5}\text{N}$. Furthermore, the work by Lu et al. (2023) deserves attention, where the authors generated a DeepPMD potential for TiAlNb. They assert that this potential successfully validates the bulk material properties of TiAl-based alloys and provides accurate evaluations of the stacking fault energy and tensile properties of γ -TiAl. It is also worth mentioning that the neuroevolution potential (NEP) method, as used in the study by Zhao et al. (2024), was applied to train a general-purpose Ti-Al-Nb potential. The authors claim that this trained potential not only explains the high-temperature mechanical properties of TiAl-based alloys but also accurately reproduces the fundamental material properties.

In this paper, we initially create essential datasets for training TiAlNb-based interatomic potentials using *ab initio* molecular dynamics (AIMD) simulations. These datasets (TiAlNb datasets) are intended to serve as a benchmark for evaluating TiAlNb-based interatomic potentials. Utilizing them, we train the TiAlNb interatomic potentials using both MTP and DP methods. Subsequently, we conduct a comparative evaluation of these potentials through error analysis and performance metrics, in addition to calculating material properties such as elastic constants, equilibrium volume, and lattice constants. We further compute finite temperature properties, including specific heat capacity and thermal expansion. Furthermore, we assess the mechanical properties by performing simulated uniaxial tension tests and calculations of generalized stacking fault energy.

2 Materials and methods

2.1 Dataset generation

We performed a large number of AIMD simulations with the Vienna *ab initio* simulation package (VASP) Kresse and Furthmüller (1996a); Kresse and Furthmüller (1996b). These simulations employed projector augmented wave (PAW) Blöchl, (1994) method to intricately model the interactions between electrons and ions. We incorporated the generalized gradient approximation (GGA) (Perdew et al., 1996; Perdew et al., 1997) for addressing exchange and correlation effects, specifically using the Perdew–Burke–Ernzerhof (PBE) functional. Our computational framework was rigorously set up with a significant cut-off energy of

510 eV, which was crucial for ensuring the accuracy and precision of our calculations. Furthermore, a Γ -centered k-point mesh of dimensions $1 \times 1 \times 1$ was employed to efficiently sample the Brillouin zone, striking a balance between computational efficiency and temporal resolution with a time step of 0.5 femtoseconds in all AIMD simulations.

For the development of the interatomic potential, we curated a comprehensive dataset encompassing a wide spectrum of Nb concentrations in γ -TiAl and α_2 -Ti₃Al alloys, ranging from 1 at.% to 14 at.%. This compilation resulted in an extensive array of 28 unique structural configurations, featuring 108 atoms in γ -TiAl and 128 atoms in α_2 -Ti₃Al. Our primary focus was on the Nb concentration range of 1–10 at.%, although we extended our dataset to include concentrations up to 14 at.% Nb to enhance the training process. Among these 28 configurations, a subset was reserved and excluded from training to assess the potential's predictive accuracy. The introduction of Nb atoms into titanium lattice sites in both phases was carefully executed, taking into account the preferential site occupancy of Nb in these alloys. For a more detailed exposition of this methodology, readers are referred to our previous work (Chandran et al., 2024) and other pertinent literature (Holec et al., 2016; Ouadah et al., 2020; Ouadah et al., 2021; Song et al., 2000; Wei et al., 2012). The AtomsK tool (Hirel, 2015) was utilized for the precise generation of these datasets, ensuring an accurate representation of the structural configurations. It should be noted that defect configurations were not included in the training process.

Subsequent AIMD simulations were conducted with meticulous care across all these structures, using the NVT and NPT ensemble over a period of nearly 10 ps. To comprehensively analyze thermal behaviors, these simulations spanned a range of temperatures including 1 K, 300 K, 500 K, 700 K, and 900 K. In the NVT ensemble, particular attention was given to selecting frames post-equilibration for analysis. The snapshots derived from these AIMD simulations provided a rich and diverse data source for training both MTP and DeepMD potentials. The snapshots were partitioned in an 80:20 ratio between the training and test sets to optimize the learning process. More details of the training database are provided in the [Supplementary Material](#), in [Section 1](#).

The TiAlNb dataset developed for this research is now available for public access. It includes the initial structural files in VASP format, essential for generating AIMD frames, alongside the training and validation datasets for DeePMD, and the training and testing datasets for MTP. Additionally, input files necessary for model training are provided, ensuring that users can replicate the results. Access to the dataset is facilitated through the link: TiAlNb dataset offering comprehensive resources for further exploration and validation.

For training the DeePMD potential, we utilized the DeePMD kit (Wang et al., 2018) along with its corresponding library designed for integration with the LAMMPS package (Thompson et al., 2022) for executing MD simulations. Similarly, the training of the MTP potential was conducted using the MLIP package (Novikov et al., 2021), employing its library specifically developed for compatibility with the LAMMPS package (Thompson et al., 2022) to facilitate MD simulations.

2.2 MTP

MTP is a type of machine learning-developed interatomic potential, implemented using the MLIP package (Novikov et al., 2021). In MTP, the energy of each atomic configuration (denoted as c_{fg}) is expressed as the sum of contributions from the local atomic neighborhoods n_i for N atoms. This is mathematically represented as,

$$E^{mtp}(c_{fg}) = \sum_{i=1}^N V(n_i), \quad (1)$$

where each contribution V is linearly expanded via a set of basis functions B_α ,

$$V(n_i) = \sum_{\alpha=1}^{N_{lin}} \xi_\alpha B_\alpha(n_i). \quad (2)$$

In the above equation, the set of parameters $\xi = \{\xi_\alpha\}$ are derived during the training process and N_{lin} is the number of these parameters. The atomic environments are characterized using moment tensor descriptors or moments, which include both radial and angular components. The moment tensor descriptor for the i -th atom is defined as,

$$M_{\mu,v}(n_i) = \sum_{j=1}^{N_{nbh}} f_\mu(|r_{ij}|, z_i, z_j) \underbrace{r_{ij} \otimes \dots \otimes r_{ij}}_{v \text{ times}}, \quad (3)$$

where r_{ij} is the position of j -th atom relative to i -th atom and $|r_{ij}|$ is the corresponding interatomic distance. N_{nbh} is the number of atoms in neighborhood. The descriptor consists of the angular part, $r_{ij} \otimes \dots \otimes r_{ij}$ (the symbol ' $\otimes$ ' represents the outer product of vectors and, hence, the angular part is the tensor of order v) and the radial part, $f_\mu(|r_{ij}|, z_i, z_j)$ having the following form,

$$f_\mu(|r_{ij}|, z_i, z_j) = \sum_{\beta=1}^{N_Q} c_{\mu,z_i,z_j}^{(\beta)} Q^{(\beta)}(|r_{ij}|), \quad (4)$$

where $\{c_{\mu,z_i,z_j}^{(\beta)}\}$ and $Q^{(\beta)}(|r_{ij}|)$ represents the set of radial parameters and radial basis functions, respectively.

The basis functions B_α are constructed using the level of moments:

$$lev M_{\mu,v} = 2 + 4\mu + v, \quad (5)$$

These coefficients have been found to be optimal for many datasets Gubaev et al. (2019). The basis functions B_α are invariant to rotations, reflections, and permutations. For defining a functional form of MTP, we choose the maximum level, lev_{max} , and include all the basis functions whose level is less or equal to lev_{max} , i.e., $lev B_\alpha \leq lev_{max}$.

The MTP parameters θ comprise of the radial parameters c and ξ , which are obtained during the fitting procedures. The fitting and learning procedure of MTP consists of finding parameters θ by minimizing the optimization problem,

$$\begin{aligned} & \sum_{k=1}^K [w_e (E^{mtp}(c_{fg_k}; \theta) - E^{qm}(c_{fg_k}))^2 \\ & + w_f \sum_{i=1}^{N_k} |f_i^{mtp}(c_{fg_k}; \theta) - f_i^{qm}(c_{fg_k})|^2 \\ & + w_s |\sigma^{mtp}(c_{fg_k}; \theta) - \sigma^{qm}(c_{fg_k})|^2] \rightarrow \min_\theta \end{aligned} \quad (6)$$

where the training set contains configurations cfg_k , $k = 1, \dots, K$ and $E^m(cfg_k)$, $f_i^m(cfg_k)$ and $\sigma^m(cfg_k)$ are the quantum mechanical energy, forces and stress tensors, respectively. N_k is the number of atoms in configuration k and w_e , w_f and w_s are the non-negative weights of energies, forces and stresses during the optimization. Equations 1–6 pertain to MTP formulation.

2.3 DeePMD

The DP method used in this study was developed using DeePMD kit (Wang et al., 2018). In DeePMD, the total energy of the system is represented as sum of energies of all the atoms. Suppose a system contains N atoms, then the total energy of the system according to DeePMD model can be represented as,

$$E = \sum_{i=1}^N E_i. \quad (7)$$

Each atomic energy E_i is obtained by the position of i -th atom and its neighbours,

$$E_i = E_{s(i)}(R_i, \{R_j | j \in N_{R_c}(i)\}), \quad (8)$$

where s_i denotes the chemical specie of atom i and $N_{R_c}(i)$ denotes the set of neighbour atoms that are at a cut-off radius of R_c from atom i . The cut-off radius of the neighbouring atoms should be such that $R_{ij} = |R_i - R_j| \leq R_c$. In order to map the atomic positions, descriptors that guarantee translational, rotational and permutational symmetries are used. We have used the deep potential smooth edition (DeepPot-SE) Zhang et al. (2018b) suggested in DeePMD Kit that includes the radial and angular information of atomic configurations. These symmetry preserving descriptors are set up using an embedding net and are later passed to the fitting net to obtain the energy of each atom. Parameter optimization is performed by minimizing the loss function L where,

$$L(p_e, p_f, p_s) = \frac{p_e}{N} |\Delta E|^2 + \frac{p_f}{3N} \sum |\Delta F|^2 + \frac{p_s}{9N} \|\Delta \Xi\|^2, \quad (9)$$

where ΔE , ΔF and $\Delta \Xi$ correspond to the root mean square (RMS) error of energy, force and stress, respectively. During the optimization process, the prefactors of energy (p_e), force (p_f) and stress (p_s) are changed. The prefactors are formulated as

$$p(t) = p^{limit} \left[1 - \frac{r_l(t)}{r_l^0} \right] + p^{start} \left[\frac{r_l(t)}{r_l^0} \right], \quad (10)$$

where r_l^0 and $r_l(t)$ denotes the learning rate at the beginning and at the training step t , respectively. p^{start} shows the starting prefactor, which goes to p^{limit} at the end of the learning. Here, an exponential learning rate is considered as,

$$r_l(t) = r_l^0 \times d_r^{t/d_s}, \quad (11)$$

where d_r and d_s denote the decay rate and decay steps, respectively. Equations 7–11 pertain to DeePMD formulation.

2.4 Potential training

2.4.1 MTP

Our investigation commenced with an analysis of the convergence of the MTP towards density functional theory (DFT)

energy and force metrics. A key aspect of this study involved conducting a grid search to optimize the MTP parameters. This search varied the potential levels from 6 to 24 and the r_{cut} values from 5 Å to 8 Å at 1 Å intervals. This strategic approach enabled us to explore a wide range of potential configurations to ascertain the most effective one. This analysis was conducted using a selection of 1800 datasets that were not included in the fitting of the potential, as illustrated in Figure 1. A pivotal aspect of the convergence test was the assignment of greater weight to energy, reflecting its critical role in the overall accuracy of the potential.

The convergence analysis revealed that the energy per atom began showing signs of convergence at level 16. This was a key observation, indicating that the MTP was effectively capturing the energy characteristics consistent with DFT calculations from this level. Similarly, the force values also demonstrated significantly low root mean square error (RMSE) commencing at the same level, further affirming the reliability of the potential from level 16 onwards.

Given these findings, and to ensure a conservative and robust approach, we ultimately selected level 18, for the final model. This level was deemed to provide an optimal balance between complexity and accuracy. Additionally, r_{cut} was set to 7 Å, for ensuring sufficient interaction range while maintaining computational efficiency. The minimum radius (r_{min}) was chosen to be 2 Å. This parameter is crucial for determining the minimum distance at which interactions are considered in the potential, thereby influencing the model's sensitivity to shorter-range forces.

For the MTP's training objective function, weights were thoughtfully assigned as follows: $w_e = 1$ for energy, $w_f = 0.01$ for force, and $w_s = 0.001$ for stress, with a priority on energy precision. This weighting strategy ensured a nuanced consideration of both force and stress, albeit with a lesser priority compared to energy, essential for crafting a nuanced and precise interatomic potential. The dataset leveraged for this purpose contained 40,000 frames, distributed in an 80:20 ratio between training and validation sets, to support effective model training and subsequent validation.

2.4.2 DeePMD

In our study, the DeepPOT-SE model, as implemented in the DeePMD-kit package, was employed. The cut-off radius for the model was set to 6 Å, with the smoothing function commencing at 4 Å, ensuring a smooth transition and reducing potential artifacts in the force calculations.

The model's architecture included radial and angular embedded-atom neural networks, each featuring three hidden layers. These layers were composed of 10, 20, and 40 nodes, respectively, providing a robust framework for capturing the complex interatomic interactions. Additionally, the fitting networks were designed with three hidden layers, each containing 100 nodes.

Regarding the training parameters, the initial learning rate was set at 0.001, gradually decreasing to a final rate of $3.51 \cdot 10^{-8}$. This gradual reduction in the learning rate allowed for finer adjustments as the training progressed, leading to more accurate model predictions. The weighting factors for energies and forces were also carefully calibrated. Initially, the weights for energies were set at 0.02, increasing to 1 in the final stages, whereas the weights for forces started at 1,000 and were reduced to 1. This

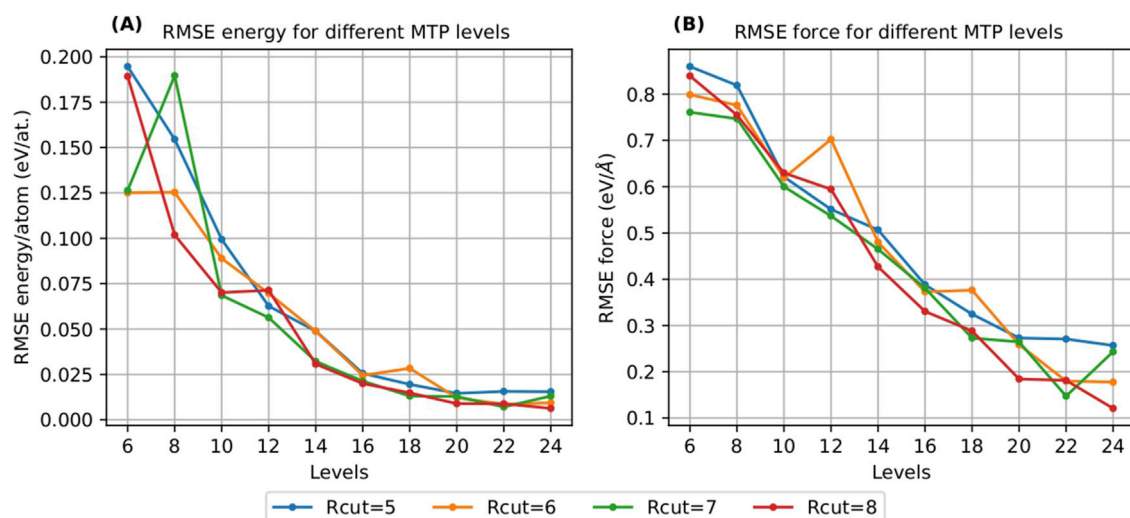


FIGURE 1
Convergence of MTP with respect to RMSE energy/atom (A) and RMSE force (B) with increasing MTP levels and cut-off radius (Rcut).

approach prioritized force accuracy in the initial stages of training and gradually shifted the focus towards energy accuracy.

It is noteworthy that virial data was not included in the training process. This decision was made to streamline the training and focus on the most critical aspects of the potential. The training was conducted over a substantial number of epochs, totaling 2,00,000, to ensure comprehensive learning and optimization of the model parameters. The utilized dataset comprised 3,33,340 frames, divided into a training and validation set following an 80:20 ratio, facilitating effective model training and validation.

3 Results and discussion

3.1 Error analysis

The correlation between predictions from ML models and DFT calculations is pivotal in ascertaining the models' accuracy, particularly in simulating potential energy surfaces and material dynamics. Our study methodically evaluates this correlation for Nb-alloyed γ -TiAl and α_2 -Ti₃Al phases using both MTP and DP models. Figures 2, 3 are instrumental in this analysis, showcasing the comparison of MTP and DP model predictions against DFT data, respectively.

The primary metrics for this comparison are energy per atom and the three components of force. For the MTP model, the recorded root mean square errors (RMSEs) are 0.0031 eV/atom for energy and 0.1285 eV/Å for the x -direction force (f_x), 0.1329 eV/Å for the y -direction force (f_y), and 0.1292 eV/Å for the z -direction force (f_z). Conversely, the DP model exhibits RMSEs of 0.0011 eV/atom for energy, 0.0783 eV/Å for f_x , 0.0795 eV/Å for f_y , and 0.0820 eV/Å for f_z . The DP model's RMSEs are notably lower than those of the MTP model, suggesting a marginally superior precision. Nevertheless, the energy and force RMSEs for both models are within acceptable ranges, confirming their effectiveness in reflecting DFT outcomes.

Additionally, the RMSEs for energy per atom, f_x , f_y , and f_z were computed for the training datasets and are presented in Table 1.

Both models' predictions exhibit a close alignment with the $y = x$ line, highlighting their capacity to accurately reproduce the test dataset energies and atomic forces. This congruence is a robust indicator of the models' exceptional accuracy, which extends to untrained test data, suggesting their effectiveness in generalizing beyond the configurations they were trained on. The considered test data encompasses structures over a complete temperature range (1 K, 300 K, 500 K, 700 K and 900 K) and varying Nb concentrations (1–14 at.%). The lower RMSEs for both energy and force underscore the DP model's superior ability to achieve DFT-level accuracy compared to the MTP model. This indicates the potential of the DP model as a more precise tool for simulating the behaviors of Nb-doped γ -TiAl and α_2 -Ti₃Al phases, backed by its comparative closeness to DFT calculations.

3.2 MD simulations

3.2.1 Energy volume curve

To augment the validity of the DeePMD and MTP potentials, this study meticulously examines the energy-volume relationships obtained using the Murnaghan fit in Nb-alloyed α_2 -Ti₃Al and γ -TiAl phases. Illustrations of these relationships are effectively depicted in Figure 4. Remarkably, the outcomes from MD simulations employing DeePMD and MTP exhibit exceptional congruence with those derived from DFT calculations. This alignment highlights the adeptness of the trained potentials in precisely capturing the energy-volume characteristics inherent to these materials.

Table 2 shows the equilibrium volume predicted by DFT, MTP and DeePMD for α_2 -Ti₃Al and γ -TiAl. For the α_2 -Ti₃Al, equilibrium volumes predicted by DFT, MTP, and DeePMD are respectively 15.88 Å³, 16.40 Å³ and 14.52 Å³. In a similar vein, for γ -TiAl

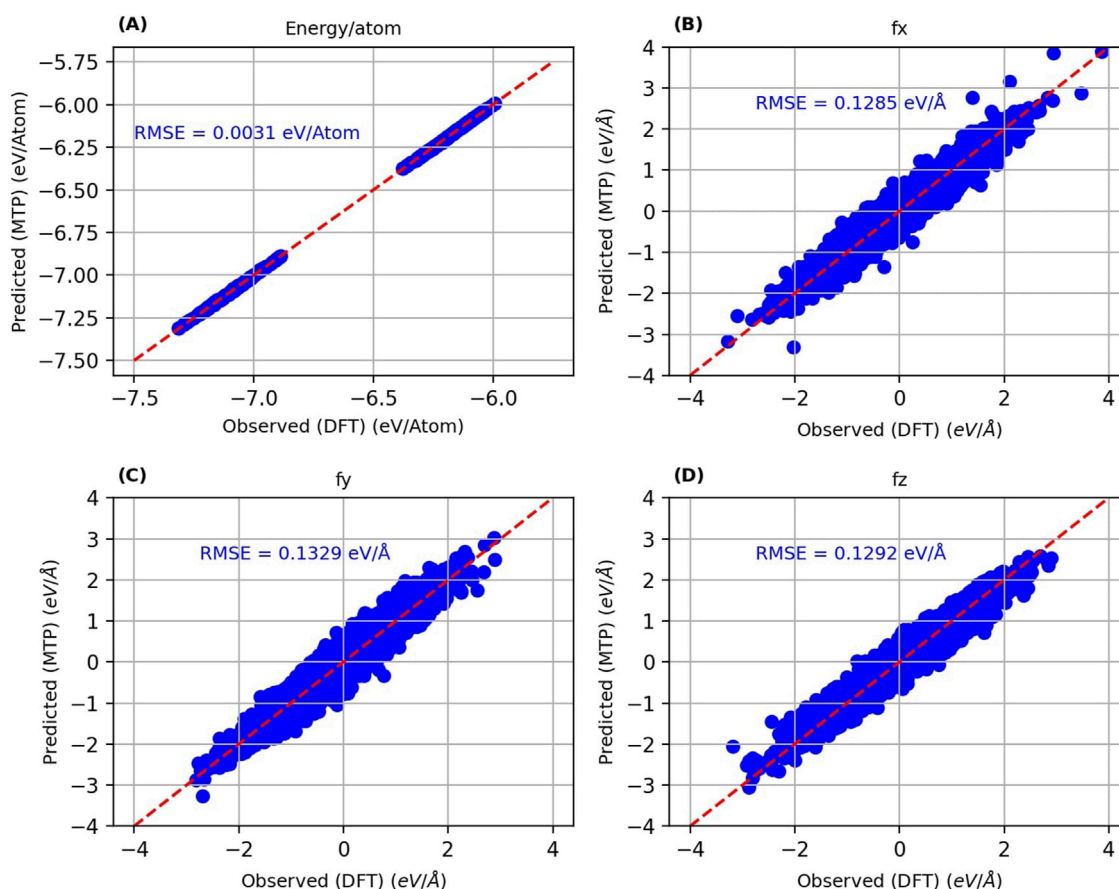


FIGURE 2

Parity plots of MTP predicted energy per atom (A), x-component of force (B), y-component of force (C) and z-component of force (D) against the corresponding DFT values.

phase, the corresponding values are $16.22 \text{ \AA}^3$, $15.76 \text{ \AA}^3$ and $15.46 \text{ \AA}^3$. Notably, the equilibrium volume predictions by MTP align more closely with those from DFT than do those by DeePMD for both phases.

The lattice constants predicted by DFT, MTP and DeePMD for α_2 -Ti₃Al and γ -TiAl are compared to corresponding experimental values (PEARSON, 1958; He et al., 1997) in Table 2. For the α_2 -Ti₃Al phase, MTP predicts lattice constants of $a = 5.683 \text{ \AA}$ and $c = 4.633 \text{ \AA}$. These closely match the DFT-calculated values of $a = 5.625 \text{ \AA}$ and $c = 4.586 \text{ \AA}$ and experimental values (PEARSON, 1958) of $a = 5.770 \text{ \AA}$ and $b = 4.620 \text{ \AA}$. Conversely, DeePMD predicts $a = 5.453 \text{ \AA}$ and $c = 4.446 \text{ \AA}$, slightly diverging from the DFT lattice parameters. The equilibrium lattice constants for γ -TiAl as forecasted by MTP— $a = 3.910 \text{ \AA}$ and $c = 4.099 \text{ \AA}$ —exhibit a striking resemblance to the DFT-determined values of $a = 3.950 \text{ \AA}$ and $c = 4.140 \text{ \AA}$ whereas the experimental values (He et al., 1997) are $a = 3.998 \text{ \AA}$ and $c = 4.067 \text{ \AA}$. The DeePMD predictions, $a = 3.910 \text{ \AA}$ and $c = 4.099 \text{ \AA}$, are identical to those of MTP, showcasing their close alignment with DFT values.

Accurate energy prediction in competitive phases is vital to avert unphysical phase segregation during MD simulations. Therefore, the integration of machine learning potentials like DeePMD and MTP notably enhances the structural analysis and prediction capabilities in Nb-alloyed α_2 -Ti₃Al and γ -TiAl phases, underscoring

their significance in the realm of advanced materials research. In terms of equilibrium volume and lattice constant prediction, MTP demonstrates superior performance compared to DeePMD.

3.2.2 Elastic constants

Next we assessed the elastic constants of α_2 -Ti₃Al and γ -TiAl alloys utilizing the different computational methodologies DFT, MTP, and DeePMD. The outcomes, quantified in gigapascals (GPa), provide a detailed comparison of the mechanical attributes predicted by these models against established experimental values (Tanaka and Koiwa, 1996; Tanaka, 1996) in Table 2.

For the α_2 -Ti₃Al alloy, notable discrepancies emerge in the elastic constants across the computational approaches compared to experimental standards (Tanaka and Koiwa, 1996). Specifically, the DeePMD's C_{11} prediction of 339 GPa significantly overshoots the figures from DFT (157 GPa), MTP (147 GPa), and experimental data (183 GPa), indicating an overestimation of longitudinal stiffness by DeePMD. The variations in C_{12} and C_{33} , which reflect differences in predicted interatomic bond strengths and compressibility along distinct crystallographic directions, are particularly striking, with DFT (103 GPa and 219 GPa) and experimental (89 GPa and 225 GPa) benchmarks. The DeePMD's divergence in C_{13} , which measures axial-longitudinal strain

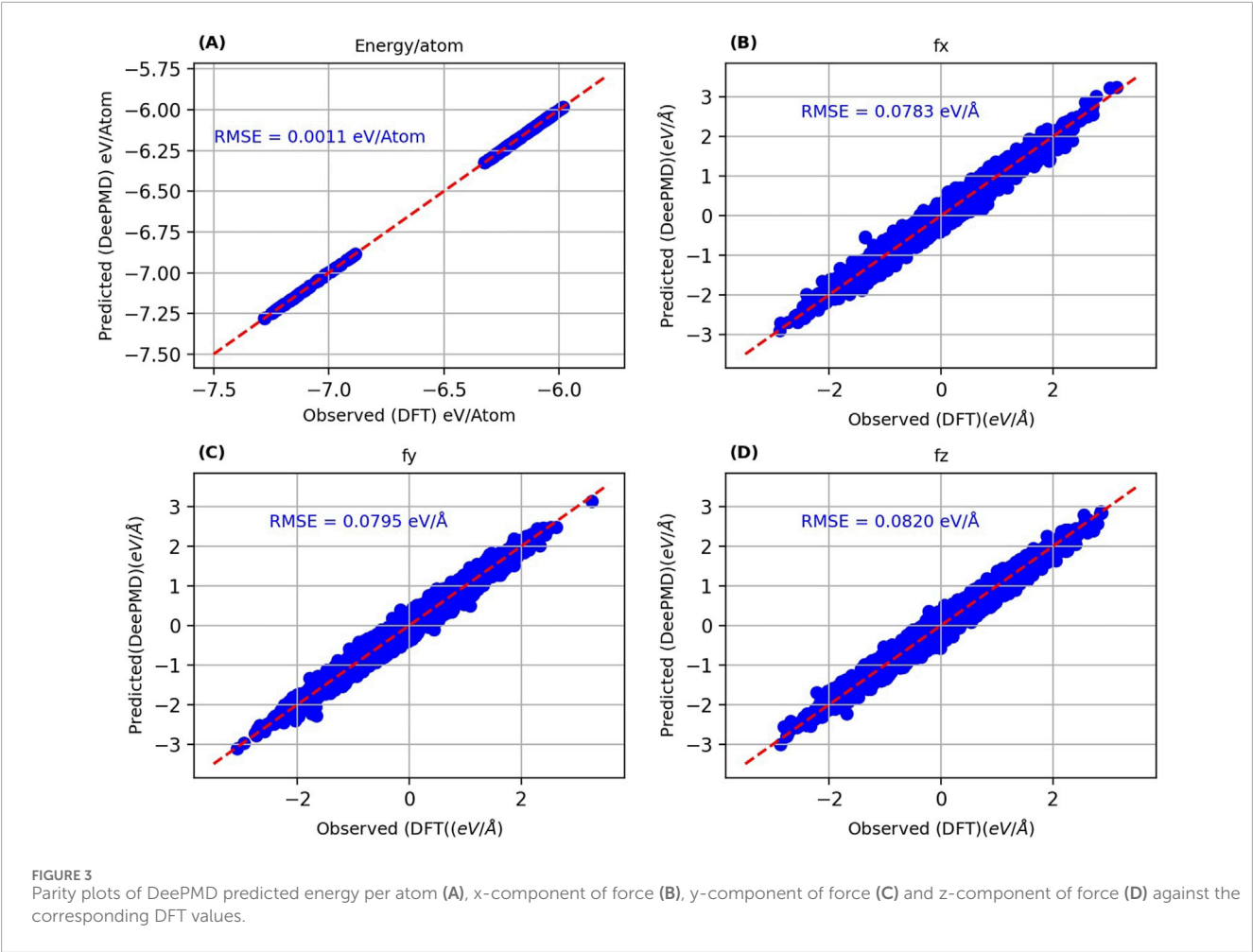


TABLE 1 Comparison of RMSE values of energy/atom, fx, fy, and fz for MTP and DeePMD using training data.

Potential	RMSE			
	Energy/atom (eV/atom)	fx (eV/Å)	fy (eV/Å)	fz (eV/Å)
MTP	0.00303	0.13365	0.13705	0.13188
DeePMD	0.00107	0.06463	0.06947	0.06886

interactions, sharply contrasts with DFT’s 74 GPa and the experimental value of 63 GPa. Additionally, the C_{44} constant, crucial for evaluating resistance to shear deformation, shows variation, with MTP predicting 34 GPa against DFT’s 46 GPa and experimental findings of 64 GPa. It is noteworthy that the C_{66} constant is not applicable for the α_2 -Ti₃Al phase in this analysis.

In the analysis of the γ -TiAl alloy, the elastic constants exhibit significant variances across the computational techniques, juxtaposed with experimental findings (Tanaka, 1996). The C_{11} constant, indicative of the material’s longitudinal rigidity, shows a broad range of values, with DFT’s prediction of 179 GPa closely mirroring the experimental value of 187 GPa, in contrast to the lower estimations of 146 GPa by both MTP and DeePMD. Conversely, the shear-related C_{44} constant demonstrates minimal

variation among the models, aligning closely with the experimental measure of 109 GPa and DFT value of 102 GPa.

Table 3 presents a comparison of the elastic constants for α_2 -Ti₃Al and γ -TiAl at 300 K, 500 K, 700 K, and 900 K. As expected, the elastic constants for both α_2 -Ti₃Al and γ -TiAl decrease with increasing temperature, consistent with findings reported in Qi et al. (2023). Our calculated values have been compared to available experimental data (He et al., 1997; Tanaka, 1996) for γ -TiAl; (Tanaka et al., 1996) for α_2 -Ti₃Al, though it is important to note that experimental values are not available for γ -TiAl at 900 K and for α_2 -Ti₃Al at 500 K, 700 K, and 900 K. Moreover, the C_{66} constant does not apply to the α_2 -Ti₃Al phase in this analysis. The discrepancies observed in the elastic constants in Table 2 are also present here; however, the overall trend of decreasing elastic constants with

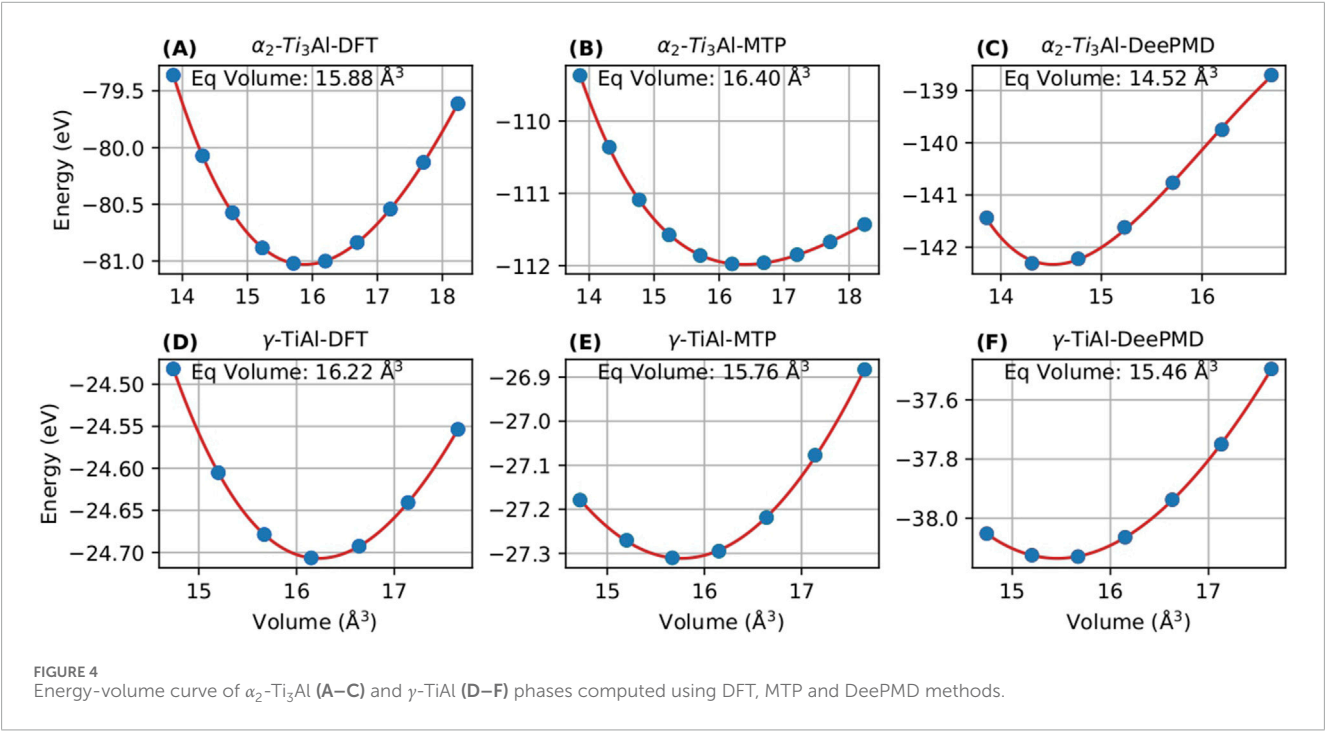


TABLE 2 Comparison of lattice constants (a, c), equilibrium volume (V_0) and elastic constants for γ -TiAl and α_2 -Ti₃Al using DFT, MTP, and DeePMD methods along with the experimental values. Lattice constants are in Å, equilibrium volume in Å³, and elastic constants are in GPa.

Quantities	α_2 -Ti ₃ Al				γ -TiAl			
	DFT	MTP	DeePMD	Exp ^{a,b}	DFT	MTP	DeePMD	Exp ^{c,d}
a	5.625	5.683	5.453	5.770	3.950	3.910	3.910	3.998
c	4.586	4.633	4.446	4.620	4.140	4.099	4.099	4.067
V_0	15.88	16.40	14.52	N/A	16.22	15.76	15.46	N/A
C_{11}	157	147	339	183	179	146	146	187
C_{12}	103	63	204	89	121	48	111	75
C_{33}	219	165	261	225	177	279	65	182
C_{13}	74	10	155	63	94	111	124	75
C_{44}	46	34	50	64	102	88	96	109
C_{66}	N/A	N/A	N/A	N/A	75	81	109	81

^aTanaka and Koiwa, 1996.

^bPEARSON, 1958.

^cTanaka, 1996.

^dHe et al., 1997.

increasing temperature for both phases remains consistent with the previous studies mentioned.

The observed disparities in elastic constants among the DFT, MTP, and DeePMD methodologies, relative to experimental data (Tanaka and Koiwa, 1996; Tanaka, 1996), highlight the intricate challenges of accurately simulating material behaviors. These differences can be attributed to the unique features of each

approach, including DFT's detailed electron correlation handling, MTP's potential function structure, and DeePMD's reliance on extensive training datasets and neural network designs. Our analysis underscores the critical importance of selecting appropriate computational strategies tailored to specific material characteristics and the need for cautious interpretation of computational findings in materials science. This study reinforces the indispensable role of

TABLE 3 Comparison of elastic constants of α_2 -Ti₃Al and γ -TiAl at 300 K, 500 K, 700 K and 900 K using MTP, DeePMD and experimental methods. Elastic constants are measured in GPa.

Elastic constants	Temperature (K)	α_2 -Ti ₃ Al			γ -TiAl		
		MTP	DeePMD	Exp ^a	MTP	DeePMD	Exp ^{b,c}
C_{11}	300	135.3	314.5	156.9	145.5	134.8	181.8
	500	119.4	287.0	N/A	143.3	130.1	177.2
	700	109.6	251.0	N/A	140.6	126.0	170.5
	900	106.4	246.6	N/A	139.4	119.5	N/A
C_{12}	300	57.3	203.2	87.4	46.8	107.9	73.5
	500	54.2	190.7	N/A	44.3	91.5	74.9
	700	48.6	189.3	N/A	43.3	88.9	73.5
	900	39.4	182.7	N/A	42.3	80.1	N/A
C_{13}	300	9.3	140.4	61.5	110.9	118.8	73.5
	500	8.0	120.4	N/A	110.4	117.6	72.5
	700	6.5	100.2	N/A	110.3	116.6	71.7
	900	6.2	103.0	N/A	108.3	112.7	N/A
C_{33}	300	164.2	218.7	216.3	46.8	64.7	174.0
	500	163.4	211.5	N/A	46.5	63.8	170.0
	700	161.7	202.4	N/A	44.3	62.0	163.1
	900	161.3	193.9	N/A	43.3	60.6	N/A
C_{44}	300	33.5	46.8	61.5	87.7	95.9	103.1
	500	32.4	45.9	N/A	86.6	95.9	98.0
	700	32.3	42.1	N/A	85.6	94.1	94.2
	900	31.3	38.6	N/A	83.3	92.2	N/A
C_{66}	300	N/A	N/A	N/A	80.6	109.5	71.6
	500	N/A	N/A	N/A	80.6	107.0	66.1
	700	N/A	N/A	N/A	78.3	106.4	62.9
	900	N/A	N/A	N/A	75.0	104.9	N/A

^aTanaka et al., 1996
^bHe et al., 1997
^cTanaka, 1996.

experimental validation in confirming the veracity of computational predictions.

3.2.3 Radial distribution function

The radial distribution function (RDF) serves as an instrumental tool for analyzing the structural properties of materials. In our study, illustrated in Figures 5, 6, we present a comparative analysis of RDFs predicted by MTP and DeePMD against

those obtained from AIMD for Nb in α_2 -Ti₃Al. Additionally, Figures 7, 8 show a similar comparison for Nb in γ -TiAl. It is important to note that the structural instances used for RDF analysis with DFT were not part of the training sets for either the MTP or DeePMD models. The agreement between the outcomes underscores the dependability of DeePMD and MTP models in probing the structure of Nb-alloyed α_2 -Ti₃Al and γ -TiAl phases.

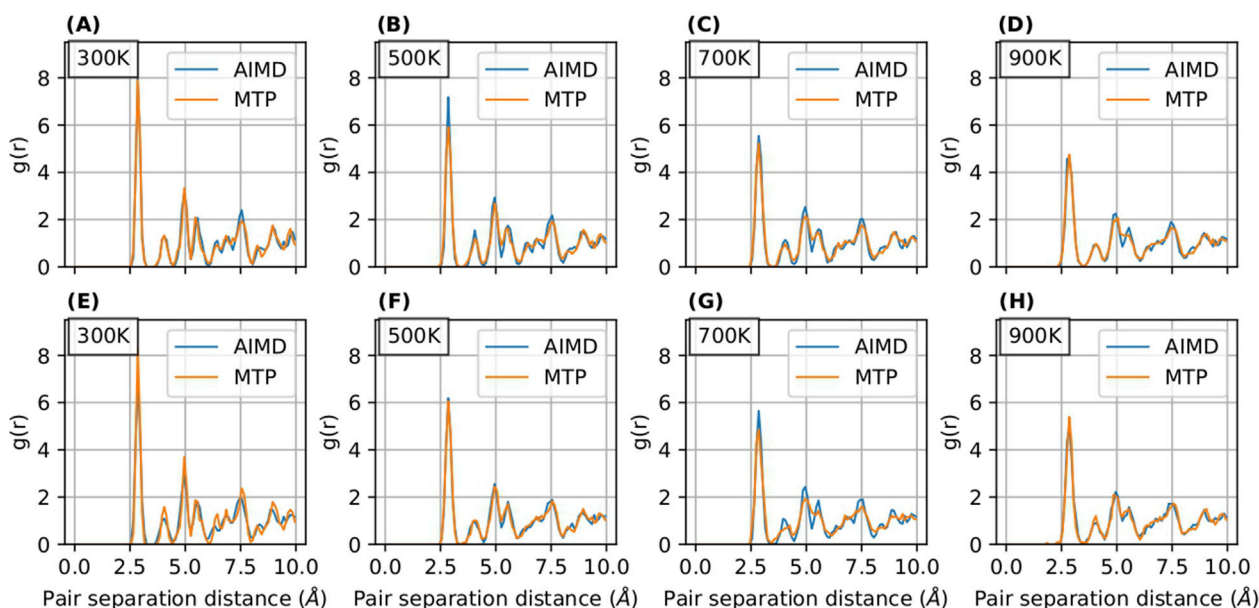


FIGURE 5
RDF curves for α_2 -Ti₃Al obtained using MTP potential for 2.3 at.% Nb (A–D) and 6.3 at.% Nb (E–H) models.

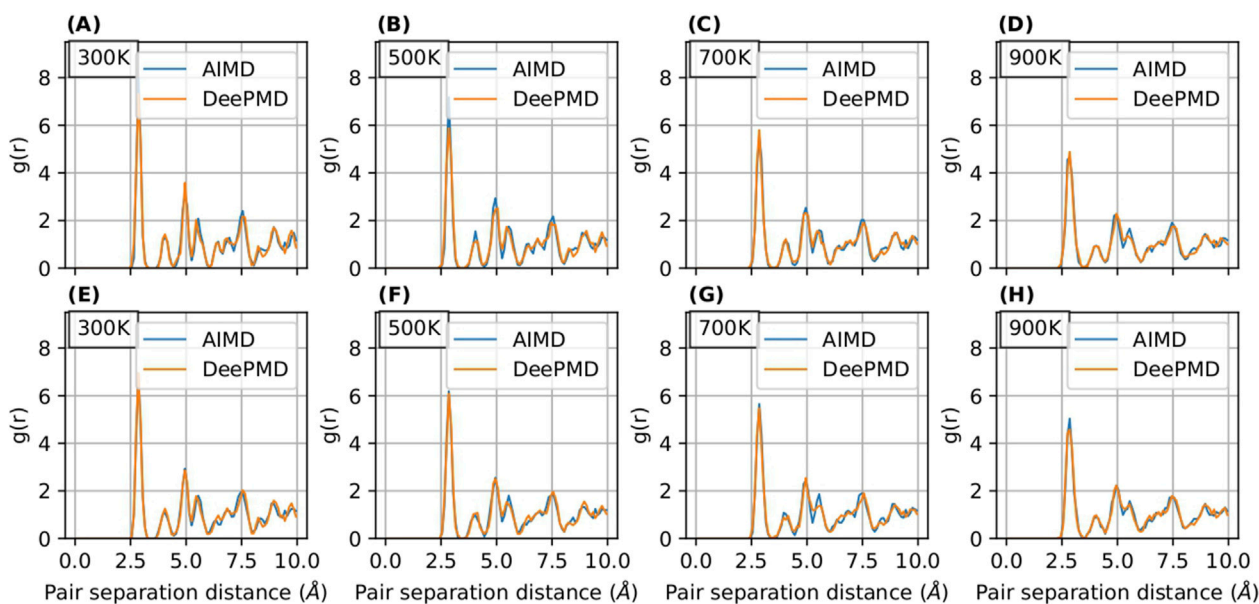


FIGURE 6
RDF curves for α_2 -Ti₃Al obtained using DeePMD potential for 2.3 at.% Nb (A–D) and 6.3 at.% Nb (E–H) models.

Notably, the RDFs for γ -TiAl from AIMD exhibited a closer match with the ML potentials compared to those for α_2 -Ti₃Al. This could be attributed to the ML models encountering challenges in accurately replicating structures when two different phases are present. In the case of α_2 -Ti₃Al, we observed a diminishing correlation between the RDFs from AIMD and ML potentials with increasing temperature, as demonstrated in Figure 5G (6.3 at.% Nb) and 6G (6.3 at.% Nb). Furthermore,

at 300 K, the RDF for 7.4 at.% Nb in γ -TiAl (Figure 8E) showed a discrepancy between AIMD and DeePMD, while the MTP model maintained a good correlation for the same scenario. These observations highlight the nuanced performance of ML potentials in different phase contexts and temperature conditions.

Additionally, we examined the scalability of the developed potentials with system sizes, as detailed in

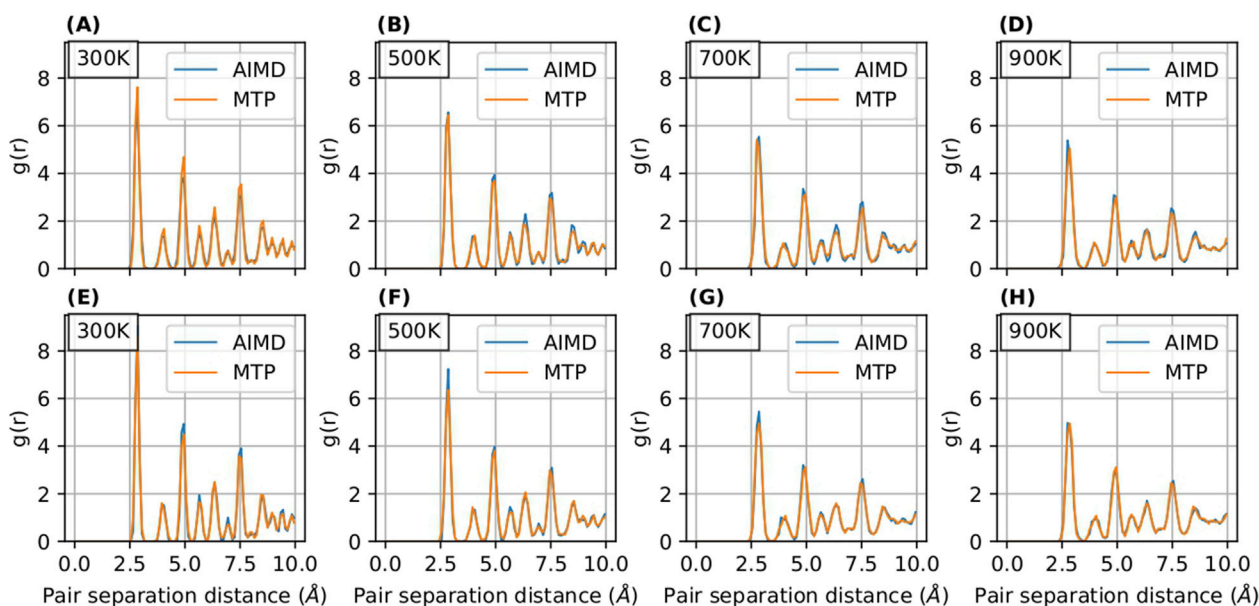


FIGURE 7
RDF curves for γ -TiAl obtained using MTP potential for 1.9 at.% Nb (A–D) and 7.4 at.% Nb (E–H) models.

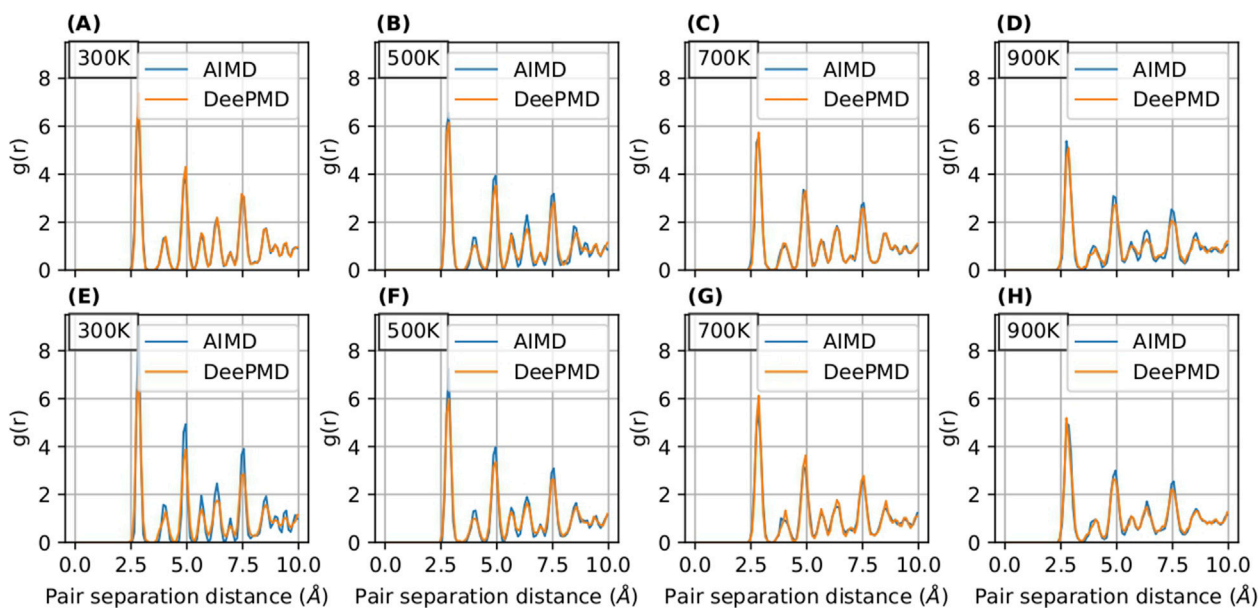


FIGURE 8
RDF curves for γ -TiAl obtained using DeePMD potential for 1.9 at.% Nb (A–D) and 7.4 at.% Nb (E–H) models.

Supplementary Figures S1–S4 in the [Supplementary Material](#) in Section 2. For further insights, please refer to this section in the [Supplementary Material](#). Our study also extended to comparing RDFs calculated by DFT, MTP, and DeePMD for a specific case of 14.8 at.% Nb concentration, which exceeds the training data's Nb concentration range. These results are documented in the [Supplementary Material](#) in Section 3, providing valuable perspectives on the models' performance beyond their initial training scope.

3.2.4 Specific heat capacity and thermal expansion

To assess the predictive capability of trained MTP and DeePMD potentials at finite temperatures, this section reports the computation of specific heat capacity (C_V) for 3.1 at.% Nb structures of α_2 -Ti₃Al and γ -TiAl and thermal expansion coefficients (α_a and α_c) for α_2 -Ti₃Al and γ -TiAl as the temperature increases. For the computation of specific heat capacity we have used phonopy (Togo et al., 2023; Togo, 2023).

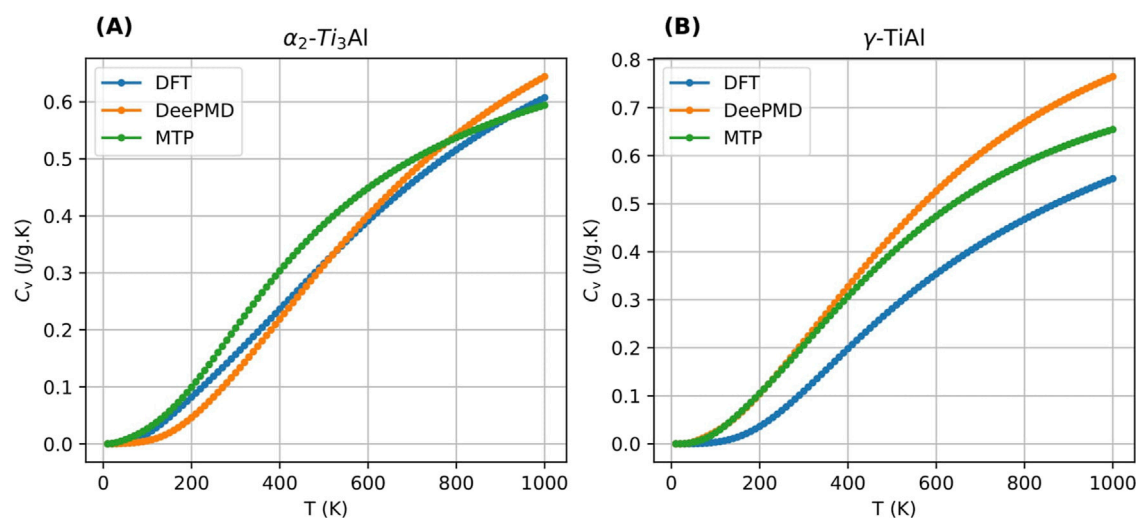


FIGURE 9 Comparison of specific heat capacity computed using MTP, DeePMD and MTP for 3.1 at.% Nb cases for α_2 -Ti₃Al (A) and γ -TiAl (B).

Figures 9A, B illustrate the comparisons of C_V values calculated using DeePMD and MTP against those derived from DFT for 3.1 at.% Nb of α_2 -Ti₃Al and γ -TiAl, respectively. It is observed that C_V progressively increases with temperature across all methods, reflecting the system's access to more degrees of freedom at higher temperatures. For α_2 -Ti₃Al, the alignment of the curves from the three methods is notably good. Specifically, the DeePMD results closely mirror the DFT outcomes, whereas the MTP results show minor deviations between 177 K and 777 K, aligning better at temperatures beyond this range. For quantitative clarity, at 200 K, C_V values are 0.045 J/g.K, 0.099 J/g.K, and 0.082 J/g.K for DeePMD, MTP, and DFT, respectively; at 600 K, they are 0.401 J/g.K, 0.449 J/g.K, and 0.392 J/g.K; and at 1000 K, they are 0.647 J/g.K, 0.595 J/g.K, and 0.608 J/g.K. It should be noted that the training set for these potentials only included structures up to 900 K. For γ -TiAl, although the MTP and DeePMD predictions initially closely match, they begin to diverge slightly beyond 288 K. Quantitative values at 200 K are 0.104 J/g.K for both DeePMD and MTP, and 0.355 J/g.K for DFT; at 600 K, the values are 0.525 J/g.K, 0.473 J/g.K, and 0.352 J/g.K; and at 1000 K, they are 0.765 J/g.K, 0.655 J/g.K, and 0.554 J/g.K.

In terms of thermal expansion, we computed the coefficients α_a and α_c corresponding to lattice parameters a and c at temperatures up to 900 K (Figure 10) for α_2 -Ti₃Al and γ -TiAl. These values were compared with experimental results for γ -TiAl (He et al., 1997) and DFT calculations for α_2 -Ti₃Al (Holec et al., 2019). It is important to note that low-temperature behaviors, often influenced by quantum effects, are not typically well captured by classical interatomic potentials. Both α_a and α_c typically increase with temperature, aiming to stabilize at higher temperatures. Experimental and DFT reference values are only available up to 750 K. At higher temperatures, α_a and α_c calculated using DeePMD are closer to these reference values, whereas MTP underpredicts these coefficients for both phases. Thus, DeePMD proves to be more accurate in predicting thermal expansion coefficients than MTP.

3.2.5 Tension test

Here, we aimed to assess the capabilities of the MTP and DeePMD potentials in capturing the thermo-mechanical characteristics of specific materials. To achieve this, we simulated uniaxial tension tests on chosen samples, applying a strain rate of 10^9 s^{-1} . The stress-strain curves obtained from these tension tests, utilizing both MTP and DeePMD potentials for the α_2 -Ti₃Al and γ -TiAl phases, are illustrated in Figure 11. Table 4 presents a comparative analysis of ultimate tensile strength (UTS) values for these materials.

We focused on comparing the UTS of the intermetallic phases to gauge the accuracy of mechanical property predictions. For the α_2 -Ti₃Al phase, MTP demonstrated consistency in UTS with increase in Nb concentration. Conversely, with the DeePMD potential, the UTS decreased as the Nb concentration increased. In the γ -TiAl phase, an increase in Nb concentration typically led to a rise in UTS with DeePMD, whereas with MTP, a decrease in UTS was observed with increasing Nb, except for the 7.4 at.% Nb scenario. The results of tension curves for γ -TiAl with Nb introduction using DeePMD shows a discrepancy with that presented in Lu et al. (2023).

Further insights into the nanomechanical behavior during uniaxial tension tests in the α_2 -Ti₃Al and γ -TiAl phases are provided in Figure 12. These figures employ color coding to depict the centrosymmetry parameter (CSP), which helps identify local lattice distortions and hence, defects. For the α_2 -Ti₃Al phase, the strain levels at which defects appeared remained relatively constant across different Nb concentrations with MTP, but showed a decrease with increasing Nb concentration in DeePMD, except in the case of 7 at.% Nb. These findings are in agreement with the stress-strain curves of depicted in Figures 11A, B. In the γ -TiAl phase, the strain threshold for defect formation decreased with increasing Nb concentration using MTP, while it increased with increasing Nb concentration in DeePMD. This observation aligns with the stress-strain data shown in Figures 11C, D.

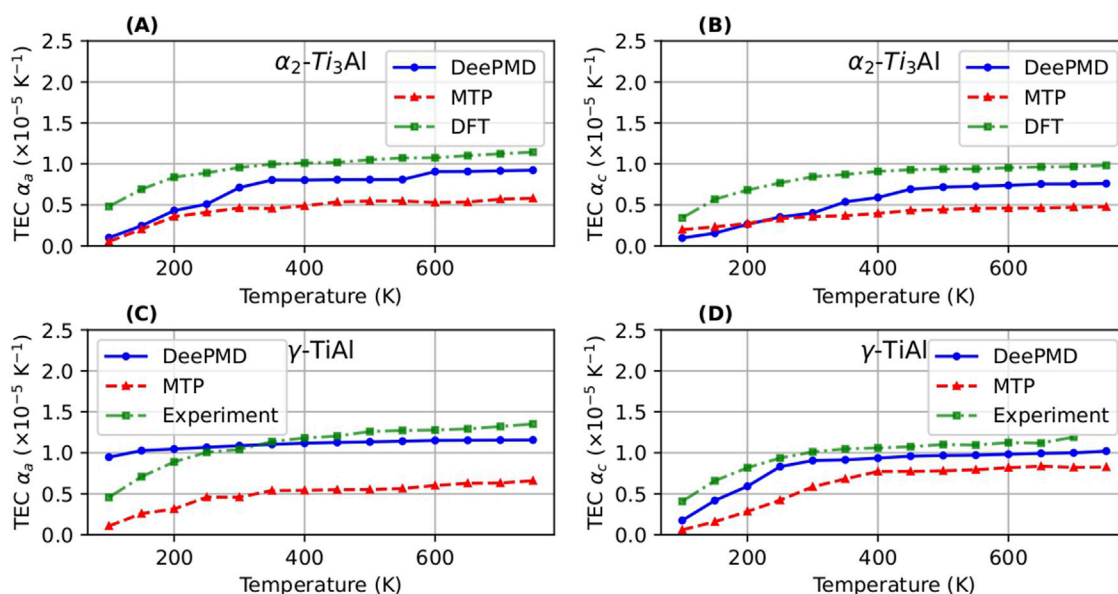


FIGURE 10 Thermal expansion coefficient (TEC) computed using DeePMD and MTP compared with the DFT values (Holec et al., 2019) and experimental values (He et al., 1997) for α_2 -Ti₃Al (A, B) and γ -TiAl (C, D) respectively.

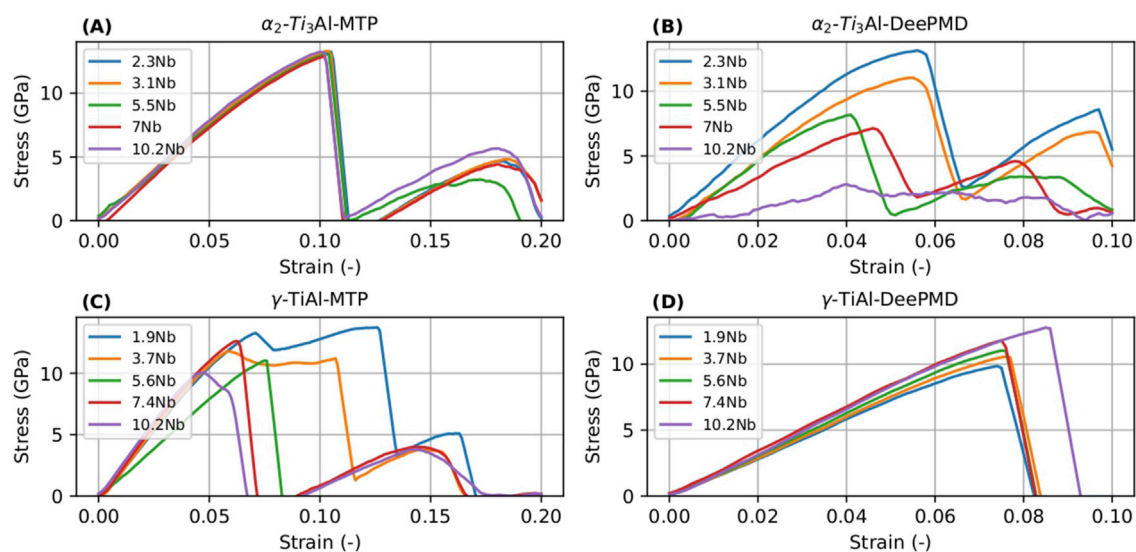


FIGURE 11 Stress-strain curves for α_2 -Ti₃Al (A, B) and γ -TiAl (C, D) with varying Nb concentration computed using MTP and DeePMD potentials.

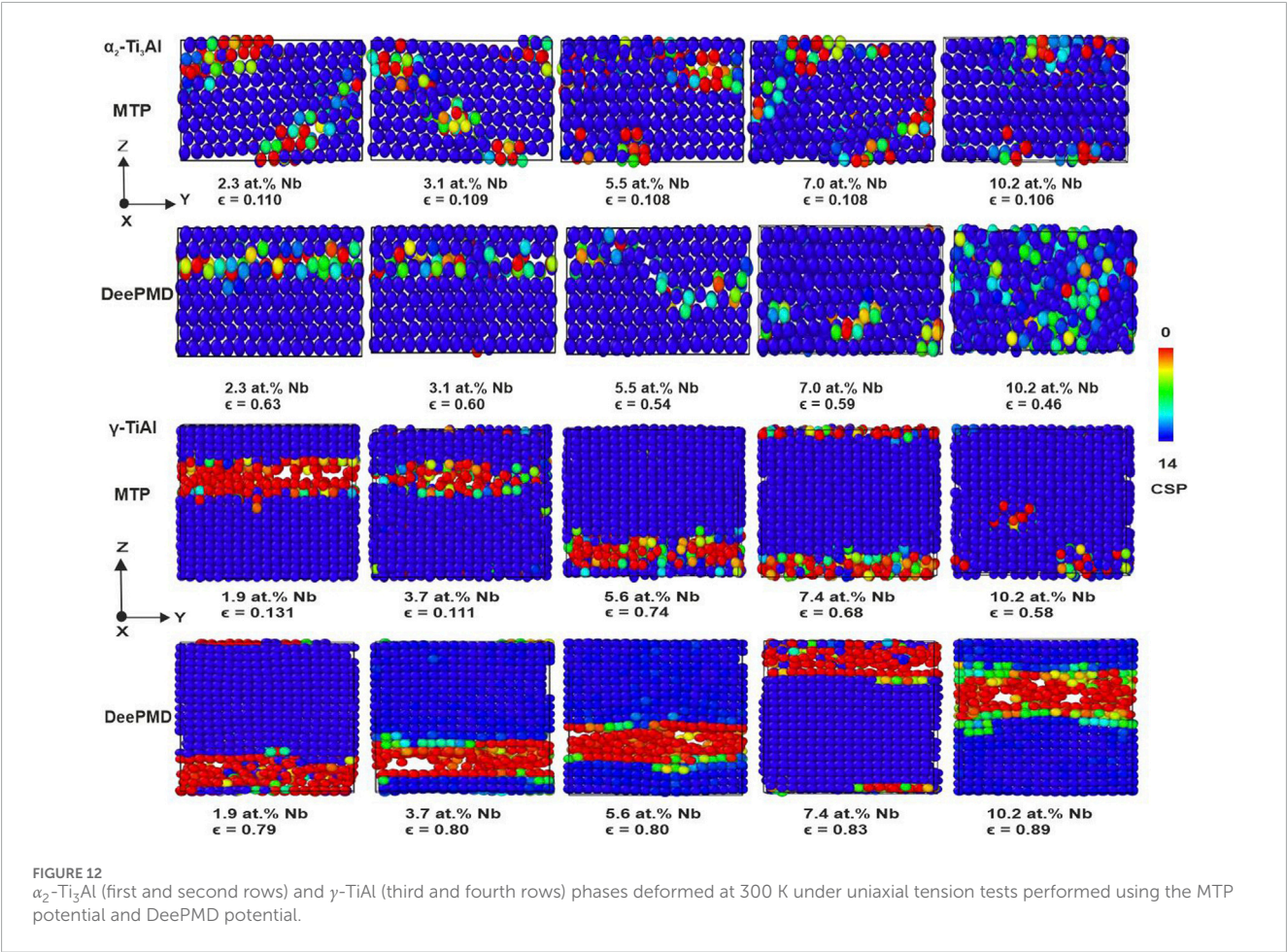
3.2.6 Generalized stacking fault energy

This section evaluates the generalized stacking fault energy (GSFE) of γ -TiAl and α_2 -Ti₃Al using both DeePMD and MTP potentials. Our analysis compares these findings with results derived from DFT and other referenced studies. Specifically, we examine studies such as Qi et al. (2023), which utilized the MTP potential, and (Lu et al., 2023), which employed the DeePMD method, where the values are available only for γ -TiAl. We used atomman (atomistic manipulation toolkit) (Becker et al., 2013; Hale et al., 2018) for the computation of stacking fault energy.

For γ -TiAl, dislocation glide occurs on the {111} close-packed planes, which are susceptible to three distinct types of stacking faults: intrinsic stacking fault (SISF), antiphase boundary (APB), and complex stacking fault (CSF). CSF is associated with ordinary $\langle 110 \rangle/2$ dislocations, while SISF and APB are linked to $\langle 110 \rangle$ and $\langle 11\bar{2} \rangle/2$ super-dislocations. Figures 13A–D shows the γ -lines along $[11\bar{2}]$ direction for γ -TiAl using both MTP and DeePMD. The energy values are documented in Table 5. MTP models generally align with the DFT and other referenced values for CSF, though MTP tends to overestimate SISF energy, in agreement with findings

TABLE 4 Comparison of UTS values for α_2 -Ti₃Al and γ -TiAl using MTP and DeePMD potentials.

α_2 -Ti ₃ Al			γ -TiAl		
Model (at.%)	MTP (GPa)	DeePMD (GPa)	Model (at.%)	MTP (GPa)	DeePMD (GPa)
2.3 Nb	13.48	13.47	1.9 Nb	13.86	10.06
3.1 Nb	13.50	11.36	3.7 Nb	12.05	10.78
5.5 Nb	13.25	8.41	5.6 Nb	11.09	11.23
7.0 Nb	13.07	7.47	7.4 Nb	12.82	12.11
10.2 Nb	13.42	3.14	10.2 Nb	10.21	13.13



from Qi et al. (2023). Conversely, DeePMD tends to align the SISF values more closely with DFT, while both models underpredict APB energy. Notably, the sequence SISF < CSF < APB remains consistent across both computational approaches as reported in Qi et al. (2023) and Lu et al. (2023). Generally, a higher GSFE indicates increased resistance and reduced mobility of dislocations, which directly contributes to the strengthening behavior in nanomaterials, whereas a lower GSFE suggests higher mobility of dislocations and improved ductility. With an increase in Nb concentration, there

is a noticeable decline in the energies for SISF, CSF, and APB for MTP, whereas DeePMD shows an increase (except for 4 at.% Nb case for CSF). Zhao et al. (2024) reported that an increase in Nb concentration reduced the stacking fault energies in the γ -TiAl phase. Similar findings were reported by Dumitraschkewitz et al. (2017), suggesting that MTP predicts this trend well. However, DeePMD presents a discrepancy where Nb alloying has increased the stacking fault energy for γ -TiAl, although this aligns with the tension curves in Figures 11C, D.

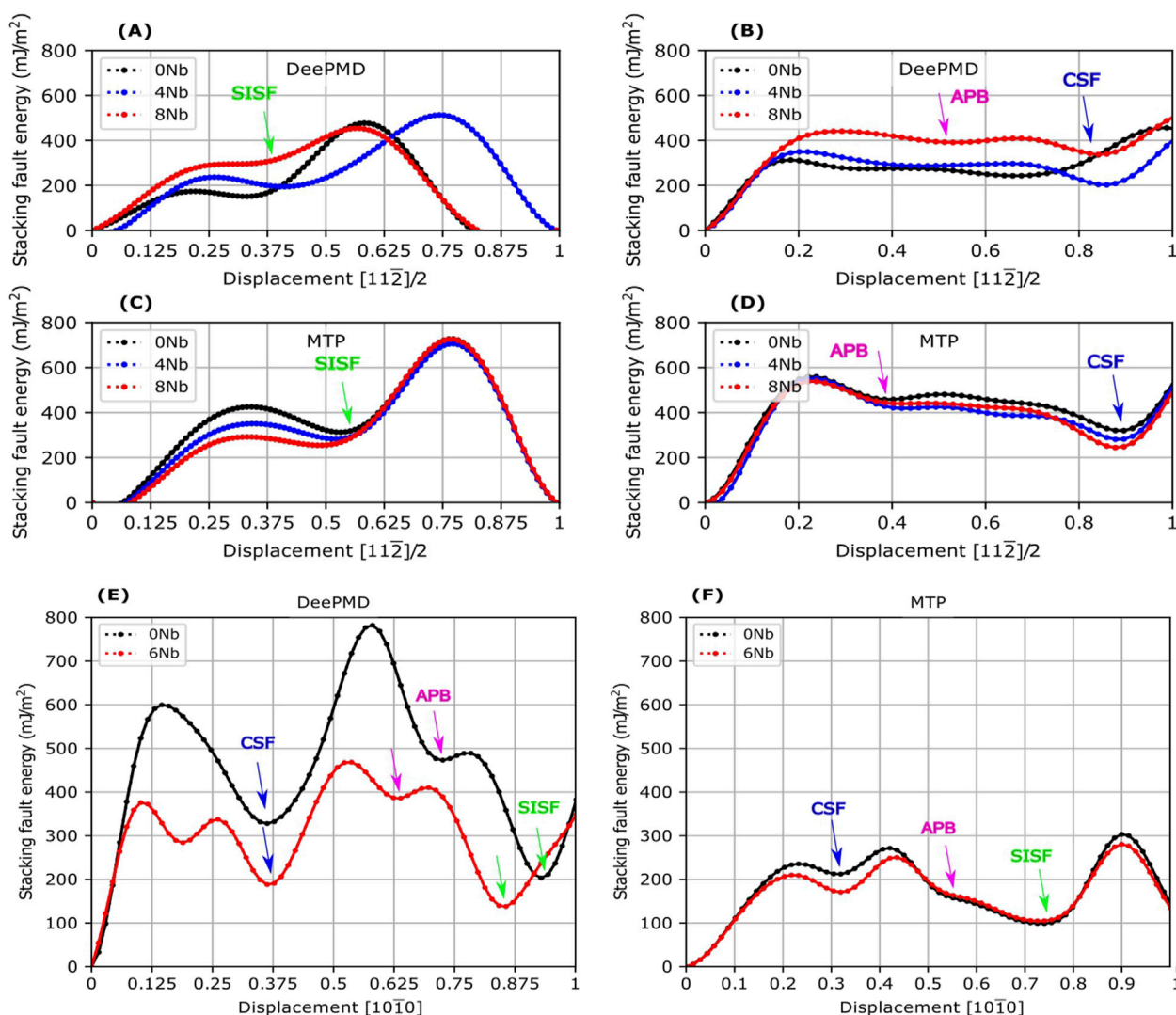


FIGURE 13

The generalized stacking fault energy (GSFE) on the {111} plane of γ -TiAl (A–D) and {0001} plane of α_2 -Ti₃Al (E–F) predicted by MTP and DeePMD. (A, C) The γ -line along the $\langle 11\bar{2} \rangle$ direction passing through the SISF. (B, D) The γ -line along the $\langle 11\bar{2} \rangle$ direction passing through the APB and CSF. (E, F) The γ -line along the $[10\bar{1}0]$ direction passing through SISF, CSF and APB.

Turning to the α_2 -Ti₃Al phase, characterized by hexagonal symmetry and various slip systems, our analysis primarily focuses on the fundamental {0001} plane. Figures 13E, F shows the γ -line along $[01\bar{1}0]$ direction obtained using MTP and DeePMD and the corresponding energy values are listed in Table 5. The results for α_2 -Ti₃Al show less consistency with DFT and referenced values (Qi et al., 2023) compared to γ -TiAl. MTP predictions for SISF align with DFT and referenced data (Qi et al., 2023), whereas DeePMD tends to slightly overestimate SISF and notably overpredict APB energy. However, DeePMD predictions for CSF energy are closer to DFT and reference values. With an increase in Nb concentration, SISF, APB, and CSF energies decrease with DeePMD, consistent with Figures 11A, B. However, MTP shows minimal change in APB and SISF values, with only a slight decrease in SISF upon Nb introduction, indicating a minimal impact of Nb alloying on the material's strength, as corroborated by the data in Table 4.

We have observed that, except for DeePMD for γ -TiAl, Nb alloying generally reduces the stacking fault energy in both phases, indicating improved ductility. Our previous work (Chandran et al., 2024) demonstrated that Nb alloying enhances dislocation density and thus overall ductility, albeit at the expense of reduced strength. The stacking fault energy computations presented here align with this finding except for DeePMD for γ -TiAl.

3.3 Comparison of performance of MTP and DeePMD

Here, we conducted a thorough assessment of the computational efficiency in MD simulations, focusing on DeePMD and MTP potentials. Figure 14 provides a graphical representation of simulation performance measured in hours per nanosecond

TABLE 5 Comparison of generalized stacking fault energies values for γ -TiAl and α_2 -Ti₃Al using MTP and DeePMD potentials.

Phases	Fault energy	Nb conc (at.%)	DFT	MTP	Ref ^a	DeePMD	Ref ^b
γ -TiAl	SISF (mJ/m^2)	0Nb	182 ^a , 194 ^c	312	322	149	129
		4Nb	—	285	—	193	—
		8Nb	—	251	—	294	—
	APB (mJ/m^2)	0Nb	560 ^d , 623 ^b	461	611	280	649
		4Nb	—	451	—	288	—
		8Nb	—	428	—	397	—
	CSF (mJ/m^2)	0Nb	356 ^a , 372 ^e	324	372	239	439
		4Nb	—	281	—	201	—
		8Nb	—	247	—	338	—
α_2 -Ti ₃ Al	SISF (mJ/m^2)	0Nb	93 ^a , 104 ^b	97	84	206	—
		6Nb	—	103	—	132	—
	APB (mJ/m^2)	0Nb	256 ^a , 257 ^f	160	213	471	—
		6Nb	—	166	—	388	—
	CSF (mJ/m^2)	0Nb	320 ^a	212	309	327	—
		6Nb	—	172	—	190	—

^aQi et al., 2023
^bLu et al., 2023
^cSeko, 2020
^dYoo and Fu, 1998
^eWoodward and Rao, 2004
^fKoizumi et al., 2006.

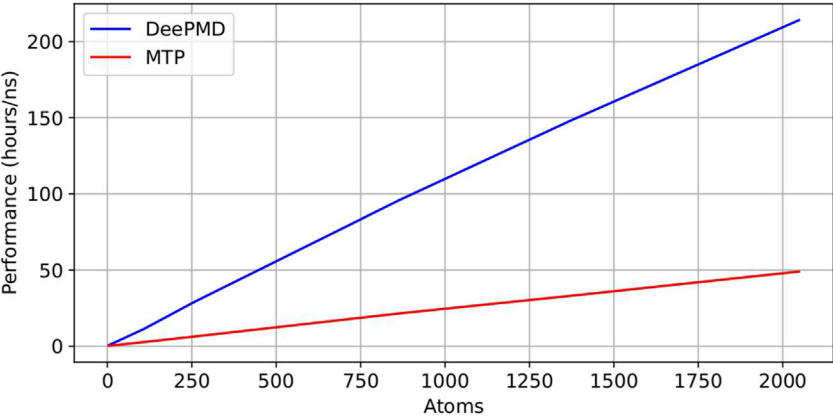


FIGURE 14 Comparison of performance (hours/ns) of DeePMD and MTP potentials in performing MD simulations.

(hours/ns) for varying numbers of atoms, specifically 4, 32, 108, 256, 864, 1,372, and 2048, in systems composed of γ -TiAl. These evaluations were conducted through short NPT simulations over a duration of 1 picosecond.

Our results demonstrate a linear correlation between computational time and atom count for both MTP and DeePMD potentials, highlighting the scalability of each potential with increasing system size. This aspect is crucial for simulations that

TABLE 6 Comparison of the timing breakdown for MPI tasks in MD simulations using DeePMD versus MTP potentials.

Tasks	MTP (seconds)	DeePMD (seconds)
Force computation	44.81	205.67
Inter-processor communication	1.15	4.33
Output	0.93	2.86
Modify	0.03	0.08
Other	1.41	2.73

involve large numbers of atoms. The “hours/ns” performance metric offers crucial insights into the computational demands, quantifying the time needed to complete 1 nanosecond of MD simulation. A significant efficiency disparity is evident from Figure 14, with MTP potential-based simulations exhibiting markedly superior speed compared to those employing DeePMD potential.

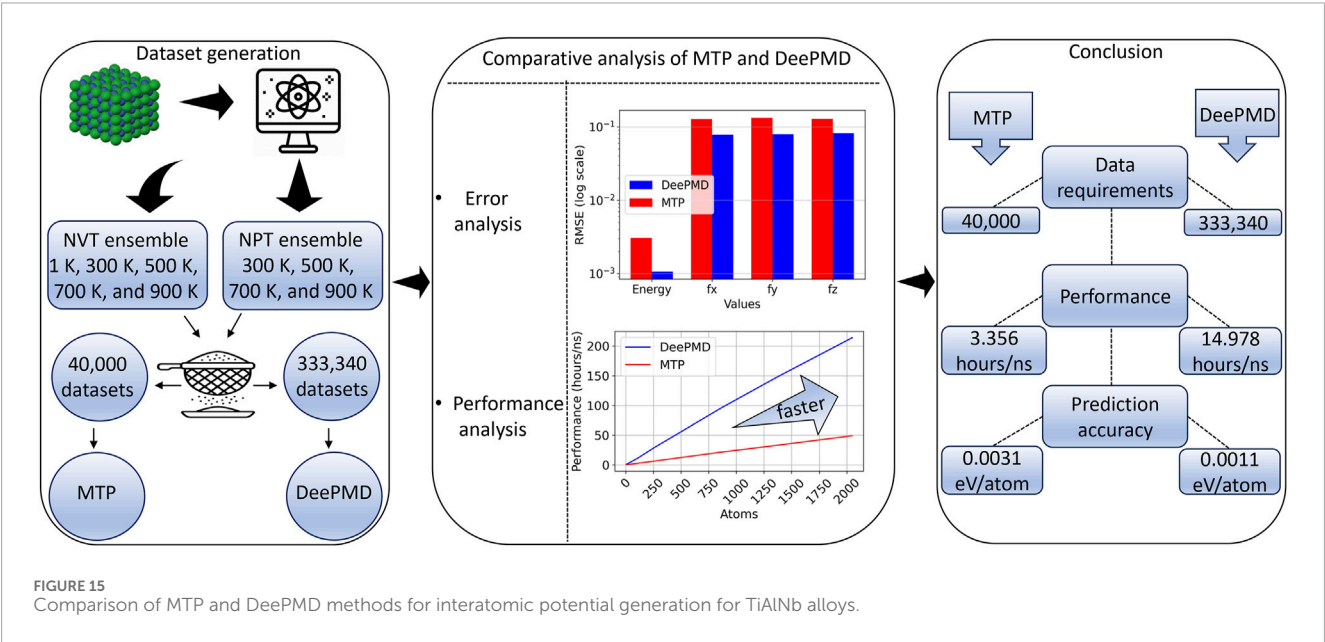
For a granular quantitative comparison, Table 6 details the message passing interface (MPI) task timings in an NVT simulation with 108 atoms over a span of 4 picoseconds (ps) for both potentials. Notably, MD simulations (NVT ensemble at 300 K for 4 ps on a system of 108 atoms) with DeePMD potential required 14.978 h/ns, whereas those with MTP potential required only 3.356 h/ns. This finding indicates that the MTP potential is approximately 4.5 times more efficient than the DeePMD potential. Furthermore, Table 6 delineates the time distribution across different simulation processes such as force computation, interprocessor communication, output generation, modifications via fixes, and other related tasks. These tests were carried out on a high-performance computing (HPC) cluster utilizing a single node equipped with 48 processors. This

detailed breakdown unequivocally shows that simulations with the DeePMD potential consume more computational time than those with the MTP potential for identical simulation conditions.

4 Conclusion

In our study, we conducted a comparative analysis of the interatomic potentials for TiAlNb alloys, developed through both MTP and deep learning approaches. We introduced a comprehensive dataset for TiAlNb alloys, aimed at serving as a benchmark for studying the Nb-alloyed phases of α_2 -Ti₃Al and γ -TiAl. Through our evaluation of the errors and performance metrics of these generated potentials, we concluded that MTP potentials offer a viable alternative in scenarios with limited computational resources. This is attributed to the lower computational cost of MD simulations relying on MTP, alongside its lower dataset requirements for effective training. However, it is important to note a slight compromise in accuracy when opting for MTP over DeePMD, despite the error margins remaining within acceptable bounds. MTP emerges as a preferable choice for applications where precise energy and force calculations are not paramount. Conversely, for endeavors requiring very high fidelity in energy and force predictions, and where it is possible to allocate more computational resources and training data, DeePMD stands out as the preferred choice. The entire workflow, encompassing dataset generation, method comparison, and the resulting conclusions, is comprehensively illustrated in Figure 15.

Our study analyzed material parameters like equilibrium volume, lattice constants, and elastic constants, revealing MTP’s equilibrium volume predictions were closer to DFT results than DeePMD’s for both α_2 -Ti₃Al and γ -TiAl phases. MTP also provided more accurate lattice parameters for the α_2 -Ti₃Al phase, while both methods agreed on the γ -TiAl phase. Elastic constant predictions of both deviated from DFT, but were more accurate for γ -TiAl. RDF curves from both models correlated well with DFT, especially



for γ -TiAl. Finite temperature properties such as specific heat and thermal expansion coefficients were in good agreement with their corresponding DFT and experimental data. Simulations of uniaxial tension tests indicated that Nb alloying decreases the strength of TiAl-based alloys, with the exception of the results from DeePMD for γ -TiAl. Additionally, computations of stacking fault energy revealed that Nb alloying enhances the ductility of TiAl-based alloys at the expense of strength, except in simulations using DeePMD for γ -TiAl. These findings align with the outcomes of our prior work (Chandran et al., 2024). Moreover, the results for generalized stacking faults—except for the DeePMD simulations of γ -TiAl—are consistent with those reported in Dumitraschkewitz et al. (2017) for Nb-alloyed systems and in Lu et al. (2023); Qi et al. (2023) for Nb-free systems, where the sequence SISF < CSF < APB has been accurately reproduced. This succinct analysis highlights the advantages and limitations of MTP and DeePMD in simulating TiAlNb alloys, guiding researchers towards the most suitable computational strategy in view of a trade-off between accuracy and resource needs.

During the model training phase, we encountered difficulties in accurately capturing the α_2 -Ti₃Al phase characteristics using both models. To address this and enhance model performance, our future work will incorporate advanced learning techniques, like active learning, into our methodology. This strategy aims to significantly improve model accuracy while reducing the reliance on extensive datasets.

Data availability statement

The datasets presented in this study can be found in online repositories. The names of the repository/repositories and accession number(s) can be found below: <https://zenodo.org/uploads/10639914>.

Author contributions

AC: Conceptualization, Data curation, Formal Analysis, Investigation, Methodology, Software, Visualization, Writing—original draft. AS: Validation, Writing—review and editing. CP: Validation, Writing—review and editing. PJ: Validation, Writing—review and editing. RA: Project administration, Supervision, Validation, Writing—review and editing. CC: Funding acquisition, Project

administration, Supervision, Writing—review and editing, Validation.

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During the preparation of this work the author(s) used ChatGpt (ChatGpt 4o-mini) in order to improve the language and readability of the text. After using this tool/service, the author(s) reviewed and edited the content as needed and take(s) full responsibility for the content of the publication.

Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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Supplementary material

The Supplementary Material for this article can be found online at: <https://www.frontiersin.org/articles/10.3389/fmats.2024.1466793/full#supplementary-material>

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Supplementary Material

In the supplementary materials, we have included detailed information regarding the training database in Section 1. We have provided a detailed analysis on the scalability of the MTP and DeePMD potentials as system sizes increase, which can be found in Section 2. Additionally, we have conducted evaluations to test the validity of the MTP and DeePMD potentials in systems with high concentrations of Nb, detailed in Section 3.

1 TRAINING DATABASE

We analyzed 28 initial structures for both γ -TiAl and α_2 -Ti₃Al, where some Ti atoms were substituted by Nb atoms to achieve the specified Nb concentration. Among these 28 configurations, a subset was set aside and not included in the training to evaluate the potential's predictive accuracy. Each of these structures (20 structures) underwent AIMD simulations in both the NVT and NPT ensembles at temperatures of 1 K, 300 K, 500 K, 700 K, and 900 K. Overall, we conducted 180 AIMD simulations: NVT simulations at 1 K, 300 K, 500 K, 700 K, and 900 K, and NPT simulations at 300 K, 500 K, 700 K, and 900 K. Snapshots from these AIMD simulations was treated as separate datasets, culminating in a total of 1,320,000 datasets. For the training and test sets, we carefully selected individual snapshots from the AIMD trajectories at specific intervals, thereby avoiding correlated data within our chosen datasets.

2 SCALABILITY OF POTENTIALS WITH SYSTEM SIZES

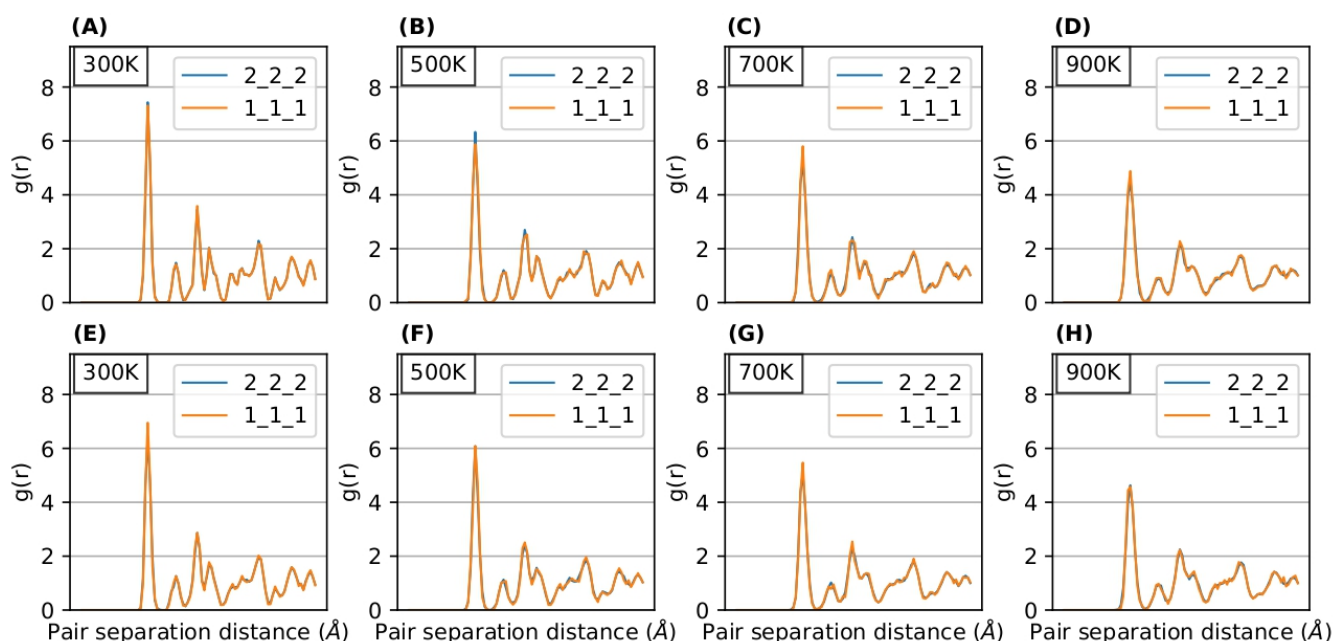


Figure S1. RDF curves for $2 \times 2 \times 2$ supercell and $1 \times 1 \times 1$ counterpart using DeePMD for α_2 -Ti₃Al. The top row (A-D) and the bottom row (E-H) correspond to the 2.3 at.% Nb and 6.3 at.% Nb models, respectively.

Here, we assess the scalability of the newly developed MTP and DeePMD potentials by varying the size of the system. Figures S3 and S1 display the RDF for the α_2 -Ti₃Al systems, comparing them with their

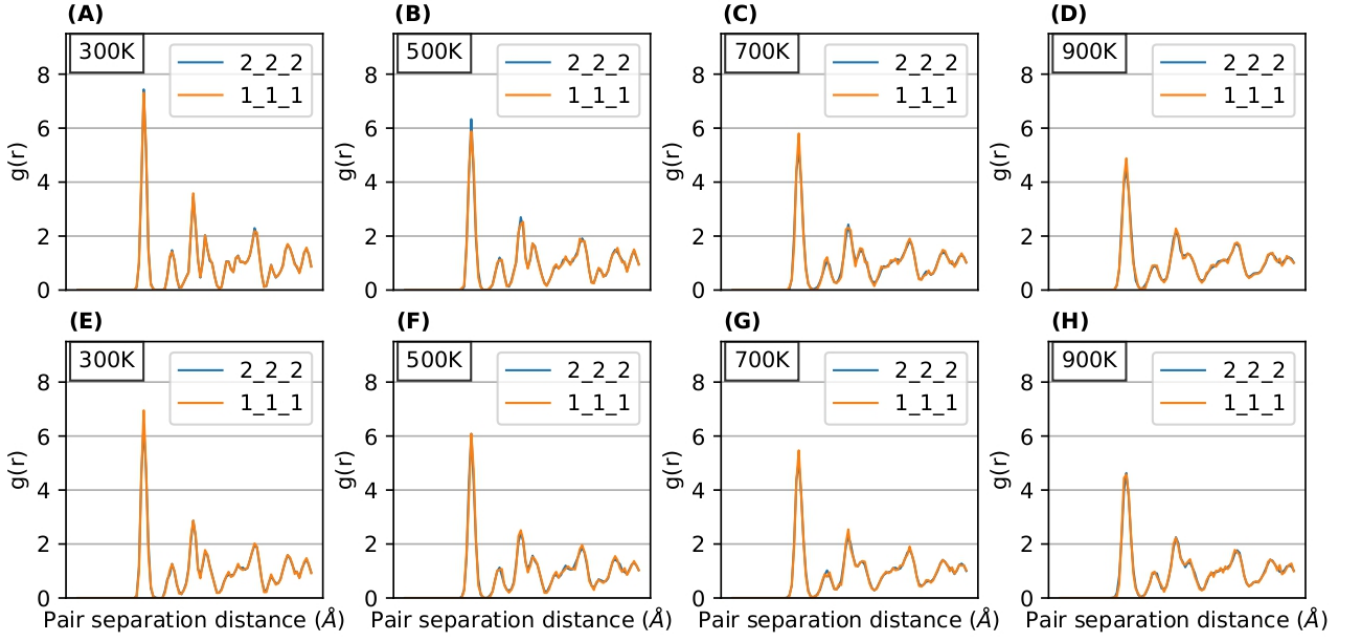


Figure S2. RDF curves for $2\times 2\times 2$ supercell and $1\times 1\times 1$ counterpart using DeePMD for γ -TiAl. The top row (A-D) and the bottom row (E-H) correspond to the 1.9 at.% Nb and 7.4 at.% Nb models, respectively.

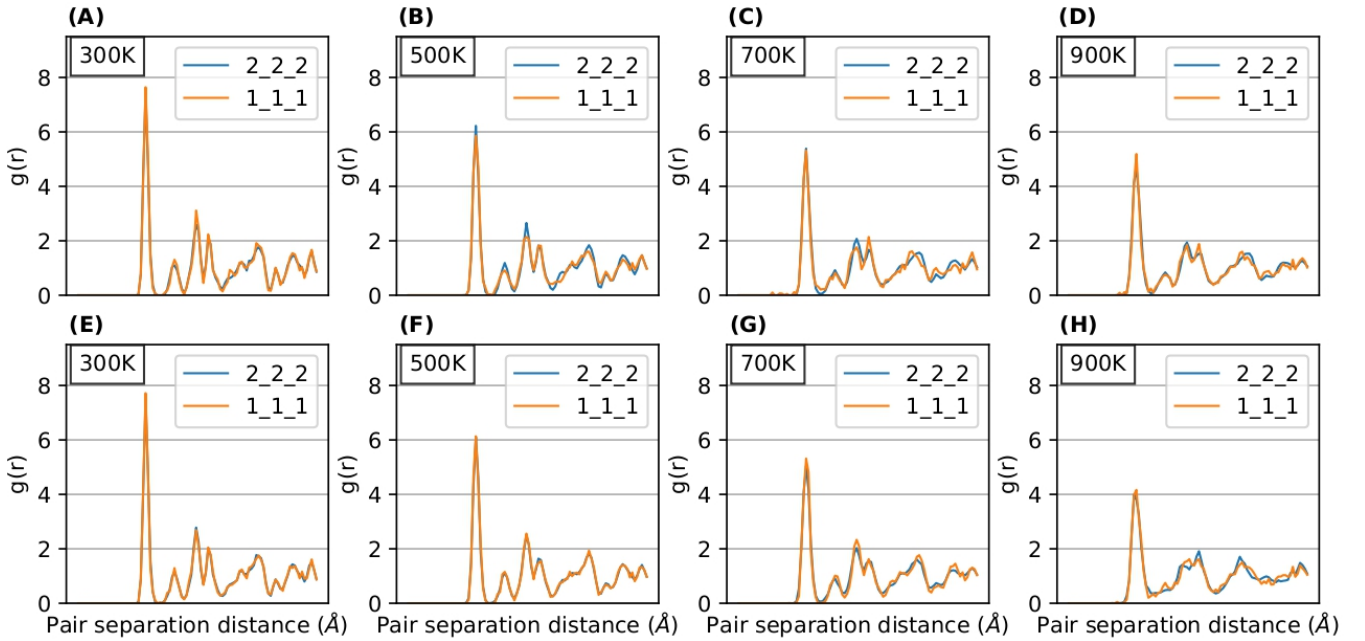


Figure S3. RDF curves for $2\times 2\times 2$ supercell and $1\times 1\times 1$ counterpart using MTP for α_2 -Ti₃Al. The top row (A-D) and the bottom row (E-H) correspond to the 2.3 at.% Nb and 6.3 at.% Nb models, respectively.

respective $2\times 2\times 2$ supercells containing 1024 atoms. Similarly, Figures S4 and S2 present the RDF for the γ -TiAl systems alongside their $2\times 2\times 2$ supercells containing 864 atoms. These figures demonstrate a good match between the RDF of the systems and that of their supercells, indicating that both MTP and DeePMD potentials exhibit effective scalability when the system size is increased.

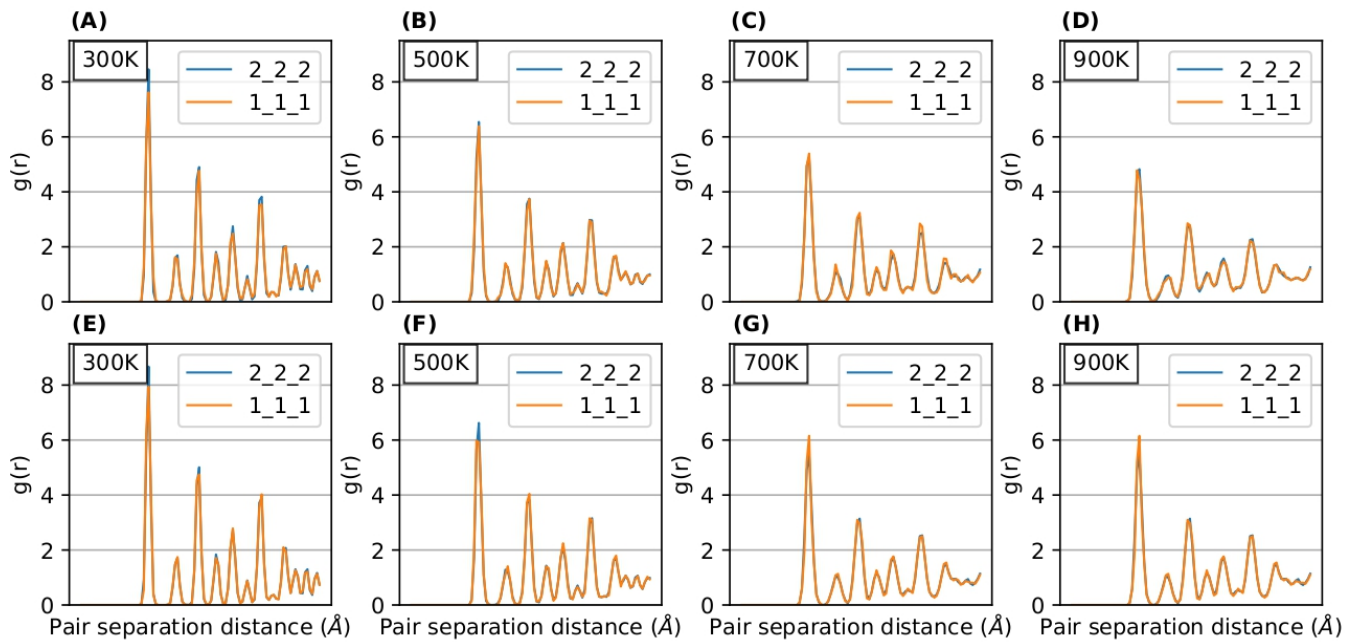


Figure S4. RDF curves for $2 \times 2 \times 2$ supercell and $1 \times 1 \times 1$ counterpart using MTP for γ -TiAl. The top row (A-D) and the bottom row (E-H) correspond to the 1.9 at.% Nb and 7.4 at.% Nb models, respectively.

3 TESTING THE VALIDITY OF POTENTIALS ON A SYSTEM WITH HIGH NB CONCENTRATION

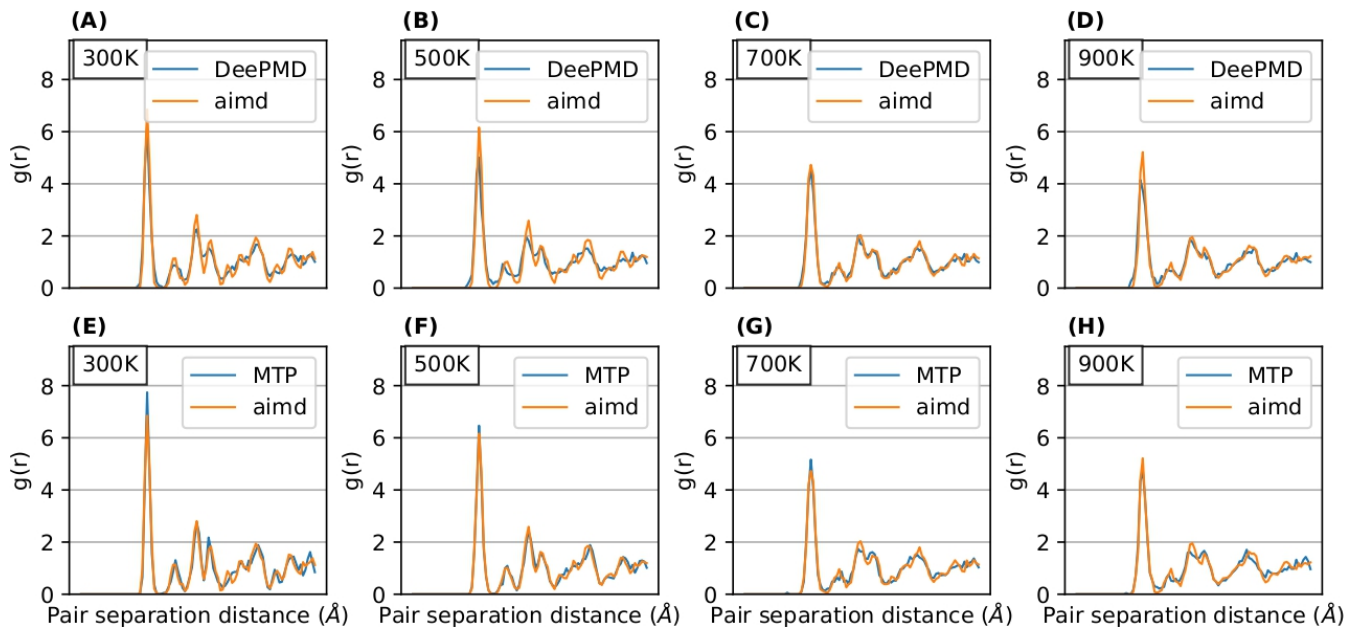


Figure S5. RDF curves of 14.8 at.% Nb concentration for α_2 -Ti₃Al using DeePMD (A-D) and MTP (E-H) potentials.

We also evaluated the validity of MTP and DeePMD potentials by testing them on a system with 14.8 at.% Nb concentration, which represents a higher Nb concentration than those included in the training

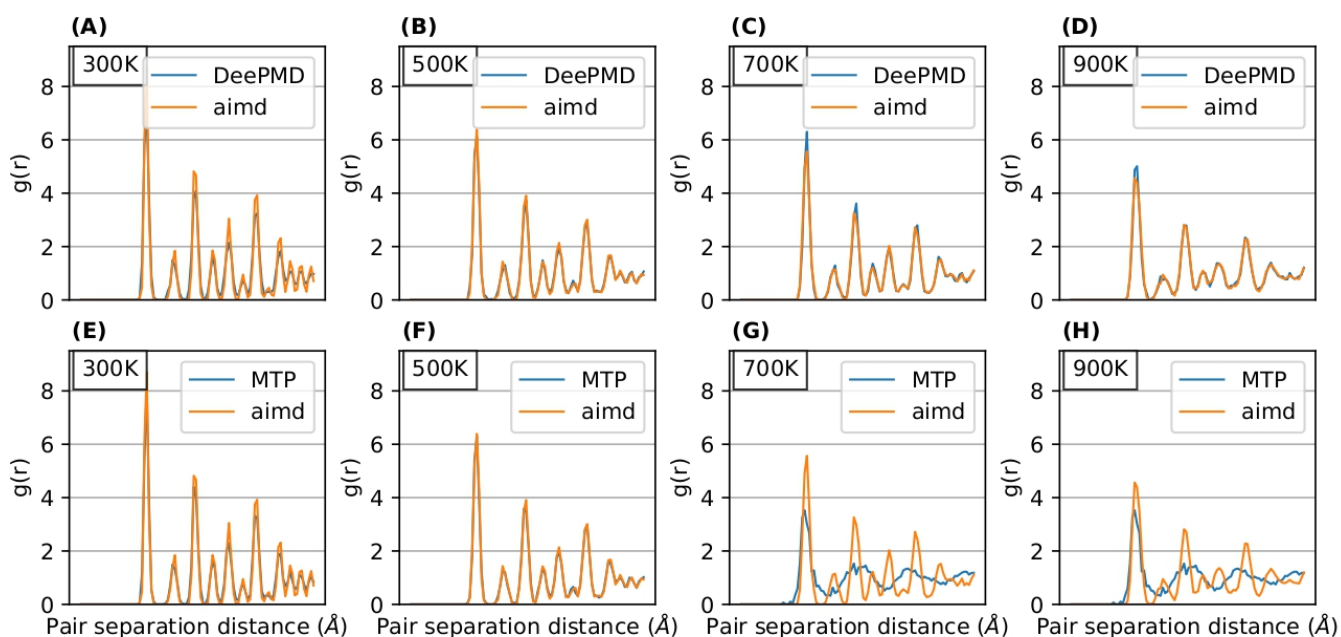


Figure S6. RDF curves of 14.8 at.% Nb concentration for γ -TiAl using DeePMD (A-D) and MTP (E-H) potentials.

datasets. Figures S5 and S6 compare the RDF of molecular dynamics (MD) simulations on models of α_2 -Ti₃Al and γ -TiAl with 14.8 at.% Nb concentration to those of AIMD simulations. The DeePMD potential demonstrated superior performance compared to MTP, showing a smaller deviation in RDF compared to that of AIMD. Specifically, in the case of MTP for γ -TiAl, the RDF exhibited a noticeable lack of agreement with that of AIMD, especially at higher temperatures (700 K and 900 K).

A.3 Publication 3

TiAlNb alloy interatomic potentials: comparing passive and active machine learning techniques with MTP and DeePMD

Anju Chandran, Archa Santhosh, Claudio Pistidda, Paul Jerabek, Roland C. Aydin and Christian J. Cyron

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TiAlNb alloy interatomic potentials: comparing passive and active machine learning techniques with MTP and DeePMD

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Intermetallic titanium aluminides are interesting for aerospace and automotive applications due to their superior high-temperature mechanical properties. In particular, γ -TiAl-based alloys containing 5–10 at.% Niobium (Nb) have attracted significant attention. Molecular dynamics (MD) simulations can elucidate and optimize these materials, provided that accurate interatomic potentials are available. In this work, we compare active and passive machine learning approaches for developing TiAlNb interatomic potentials using both deep potential molecular dynamics (DeePMD) and the moment tensor potential (MTP) methods. Our comprehensive evaluation encompasses elastic constants, equilibrium volume, lattice parameters, and finite-temperature behavior, as well as simulated tension tests and generalized stacking fault energy calculations to assess the impact of Nb on the thermo-mechanical properties of γ -TiAl and α_2 -Ti₃Al phases. Active learning consistently outperformed passive learning for both methods while requiring only a fraction of the training samples. Notably, active learning with DeePMD yielded a single potential capable of predicting the properties of both phases, whereas MTP exhibited limitations that necessitated separate training for each phase. Although active learning potentials excelled in predicting high-temperature behavior, their room-temperature property predictions were less accurate due to a sample selection bias toward higher temperatures. Overall, our thermomechanical analysis demonstrates that Nb incorporation enhances ductility while simultaneously reducing strength.

KEYWORDS

TiAlNb alloy, machine-learning interatomic potentials, deep learning, moment tensor, active learning, molecular dynamics, density functional theory

1 Introduction

Intermetallic alloys based on γ -titanium aluminide (γ -TiAl) have emerged as superior lightweight materials for aerospace and automotive applications. Their unique combination of low density, excellent oxidation resistance, and strong creep

resistance at elevated temperatures makes them particularly attractive (Appel et al., 2011). Typically, these alloys consist of two intermetallic phases: the γ -TiAl phase, which adopts an ordered face-centered tetragonal structure ($L1_0$, P4/mmm), and the α_2 -Ti₃Al phase, characterized by an ordered hexagonal structure ($D0_{19}$, P6₃/mmc). Significant progress has been made in developing γ -TiAl alloys for high-temperature applications, notably within the TNM (TiAl-Nb-Mo) and TNB (TiAl-Nb-B) systems (Appel et al., 2011; Zhang et al., 2016; Li et al., 2014; Clemens and Mayer, 2012; Klein et al., 2016). These systems generally contain 5–10 at.% Nb along with minor additions of elements such as Mo, B, C, Si, W, Cr, and Ta. Although Nb is known to enhance mechanical performance by improving both strength and ductility (Zhu et al., 2018; Chen and Zhang, 2002; Cheng et al., 2016; Liu et al., 2022; Song et al., 2020; Zhang et al., 2023; Appel et al., 2011, Appel et al., 2006; Clemens and Mayer, 2012; Appel et al., 2000; Liu et al., 2002), its precise role in these alloys remains somewhat ambiguous.

In our previous work Chandran et al. (2024a), we investigated the effect of Nb on the thermo-mechanical properties of TiAl-based alloys using atomistic simulations with Farkas' ternary interatomic potential (Farkas and Jones, 1996). We examined a range of models—from single-phase structures and lamellar interfaces to complex microstructure-informed atomistic models (MIAMs) featuring nano-polycolonies. Our study revealed that Farkas' potential (Farkas and Jones, 1996) is limited in handling Nb concentrations above 1 at.% in MIAMs and above 2 at.% in certain lamellar interfaces, a significant drawback given that Nb levels of 5–10 at.% are most promising for enhancing strength and ductility. To overcome this limitation, our subsequent work Chandran et al. (2024b) focused on developing machine learning (ML)-based interatomic potentials for TiAlNb in molecular dynamics (MD) simulations. We employed moment tensor potential (MTP) and deep potential molecular dynamics (DeePMD) methods, performing a comparative evaluation based on error analysis, performance metrics, and calculations of key material properties such as elastic constants, equilibrium volume, and lattice parameters. Finite temperature properties, including specific heat capacity and thermal expansion, were also computed, and mechanical properties were assessed through simulated uniaxial tension tests and generalized stacking fault energy calculations. However, these approaches required extensive datasets (333,340 configurations for DeePMD and 40,000 for MTP) and struggled to accurately capture the α_2 -Ti₃Al phase characteristics. In the current work, we implement an advanced active learning strategy to develop a robust interatomic potential with reduced reliance on large datasets. We also compare active and passive learning procedures to determine the effective approach for developing the interatomic potentials for TiAlNb alloys.

The development of such an effective potential is a crucial step, as it provides the foundation for bridging the gap between atomistic mechanisms and macroscopic mechanical behavior. The ultimate failure of polycrystalline materials is mainly governed by processes initiated at the atomic and microstructural scales, such as dislocation activity and nano-void formation (Maruschak et al., 2023). Key properties calculated from atomistic models, including the generalized stacking fault energy (GSFE) and elastic constants, directly inform these fundamental deformation mechanisms. Therefore, an accurate potential is an essential first step toward

building predictive, multi-scale models of the structural integrity and service life of Ti-Al-Nb components.

Various ML techniques have been developed for generating interatomic potentials. Among these, the deep potential (DP) and MTP methods have distinguished themselves through their success in modeling both ordered and disordered systems. DP-based potentials have been effectively applied to diverse systems, including MgCl₂-NaCl and MgCl₂-KCl (Xu et al., 2023), LiF and FLiBe (Rodriguez et al., 2021), AlN (Li et al., 2024) and Cu (Du et al., 2022). Similarly, MTP has been utilized in studies on Ti_{0.5}Al_{0.5}N (Tasnádi et al., 2021). In addition, the work by Lu et al. (2023) is particularly noteworthy, as it developed a DeePMD potential for TiAlNb that not only validates the bulk material properties of TiAl-based alloys but also provides accurate evaluations of the stacking fault energy and tensile properties of γ -TiAl. Notably, Zhao et al. (2024) applied the neuroevolution potential (NEP) method to train a general-purpose Ti-Al-Nb potential that both explains the high-temperature mechanical properties of TiAl-based alloys and reproduces their fundamental material properties.

Active learning is a sophisticated approach that initiates the training process with a small dataset. As learning proceeds, the algorithm selectively incorporates the most informative data points, thereby incrementally enhancing the model's performance. This strategy facilitates the development of high-quality interatomic potentials without the need for extensive datasets, substantially reducing computational costs—especially considering that dataset generation often relies on numerous computationally intensive *ab initio* molecular dynamics (AIMD) calculations. AIMD integrates the atomic equations of motion in real time using forces derived from first-principles, thereby capturing the dynamic evolution of a system at finite temperatures. In contrast, density functional theory (DFT) is a static approach that calculates the ground-state electronic structure for a fixed atomic configuration, yielding highly accurate energies and forces. Thus, while AIMD utilizes DFT to generate realistic dynamical trajectories, DFT itself provides precise, time-independent property evaluations. Active learning has been successfully implemented within the MTP framework for various materials, including Rosenbrock et al. (2021), Ta-V-Cr-W alloys (Gubaev et al., 2023), Cu-Ni-Si-Cr alloys (Ángel et al., 2023; Attarian et al., 2022), Mo-Nb-Ta systems (Novikov et al., 2021), as well as Cu-Pd, Co-Nb-V, and Al-Ni-Ti systems (Gubaev et al., 2019), along with Zr (Luo et al., 2023) and U (Chen et al., 2023). Similarly, active learning in DeePMD has been applied to materials such as Ag₂S (Balyakin and Sadovnikov, 2022), B₄C (Ghaffari et al., 2024; Du et al., 2022), HfO₃ (Wu et al., 2021), Al (Cheng et al., 2021), MgAlSi (Zhu et al., 2024), MgCl₂-NaCl-KCl (Feng and Lu, 2024), SnSe (Guo et al., 2021), and SrTiO₃ (He et al., 2022).

In this paper, we compare passive and active learning strategies using the MTP and DeePMD methods. Section 2.1 describes the generation of the dataset for potential training. Here, the term “dataset” refers to the collection of atomic configurations, along with their corresponding energies and forces derived from AIMD simulations, which serve as the input for training the interatomic potentials. We also introduce the formulations of the MTP (Section 2.2) and DeePMD (Section 2.3) active learning strategies, as well as the methodology for simulated tension tests (Section 2.4). In Section 3.1, we present a detailed analysis of active learning performance—illustrated through root mean square error

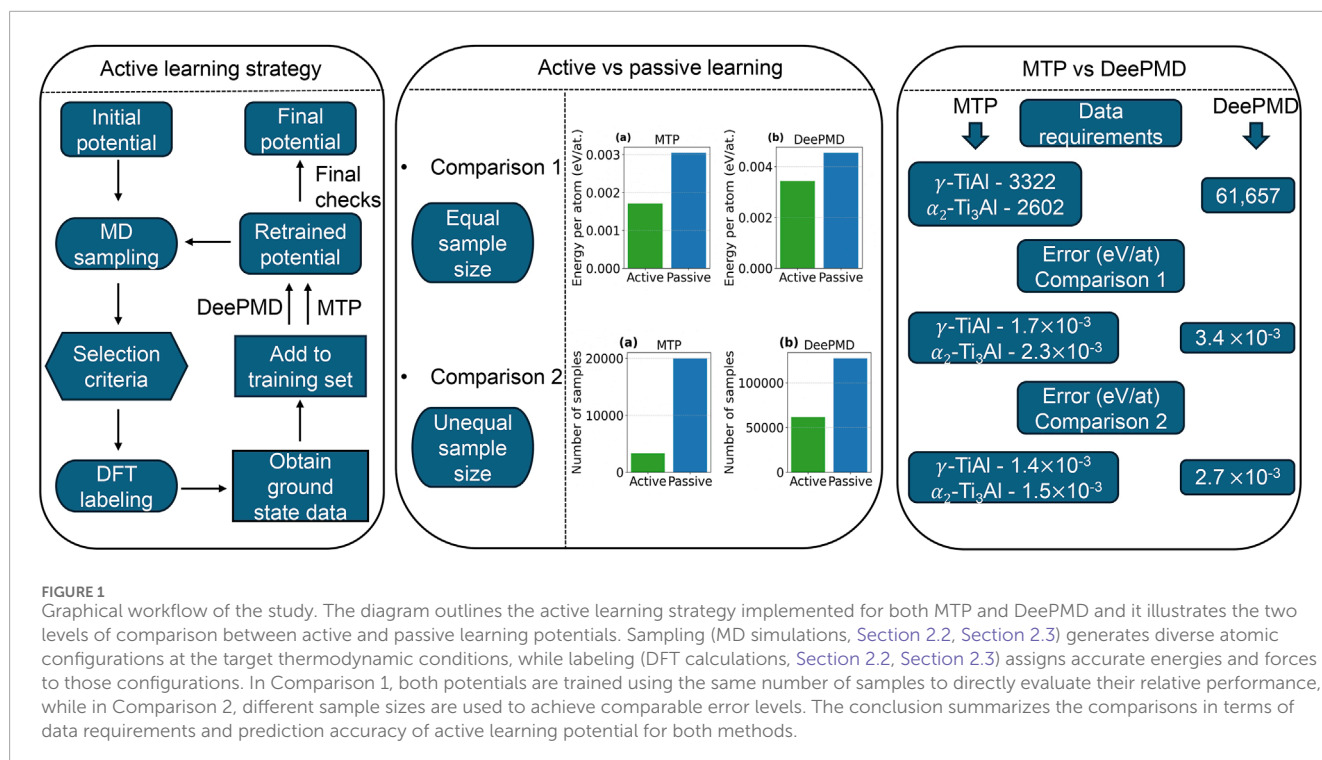


FIGURE 1

Graphical workflow of the study. The diagram outlines the active learning strategy implemented for both MTP and DeePMD and it illustrates the two levels of comparison between active and passive learning potentials. Sampling (MD simulations, Section 2.2, Section 2.3) generates diverse atomic configurations at the target thermodynamic conditions, while labeling (DFT calculations, Section 2.2, Section 2.3) assigns accurate energies and forces to those configurations. In Comparison 1, both potentials are trained using the same number of samples to directly evaluate their relative performance, while in Comparison 2, different sample sizes are used to achieve comparable error levels. The conclusion summarizes the comparisons in terms of data requirements and prediction accuracy of active learning potential for both methods.

(RMSE) energy/atom and force curves—and compare these metrics with those obtained via passive learning. Finally, MD simulations were conducted to evaluate the predictive accuracy of the developed potentials, as demonstrated by equilibrium lattice parameters, equilibrium volume and elastic constants (Section 3.2.1), finite-temperature properties (Section 3.2.2), and thermo-mechanical behavior under uniaxial tension, including GSFE calculations (Section 3.2.3). Figure 1 depicts a comprehensive workflow that outlines both the active learning strategy employed in our study and the approach used to compare passive and active learning potentials.

2 Materials and methods

2.1 Dataset generation

The dataset generation began by constructing a series of unique structural configurations for γ -TiAl and α_2 -Ti₃Al alloys with varying Nb concentrations. These concentrations spanned from 1 at.% to 14 at.%, resulting in 28 unique structural configurations. Each configuration consisted of 108 atoms for γ -TiAl and 128 atoms for α_2 -Ti₃Al. Here, a configuration refers to a distinct atomic arrangement within the simulation cell, defined by the specific positions and chemical identities of the atoms. While the primary focus was on Nb concentrations between 1 and 10 at.%, additional configurations with concentrations up to 14 at.% were included to strengthen the dataset for training. The introduction of Nb atoms into titanium lattice sites was carefully executed, accounting for the preferential site occupancy of Nb in these materials. The methodology for generating these configurations is detailed in our prior work Chandran et al. (2024a), as well as in other related studies (Holec et al., 2016; Ouadah et al., 2020; Ouadah et al., 2021;

Song et al., 2000; Wei et al., 2012). Precise structural configurations were generated using the Atomsk tool (Hirel, 2015).

To generate the dataset, AIMD simulations were performed on these configurations using the Vienna ab-initio simulation (VASP). The AIMD calculations employed projector augmented wave (PAW) potentials (Blöchl, 1994) to accurately model electron-ion interactions, while exchange-correlation effects were treated via the generalized gradient approximation (GGA) using the Perdew–Burke–Ernzerhof (PBE) functional (Perdew et al., 1996; Perdew et al., 1997). An energy cutoff of 510 eV was adopted, with the Brillouin zone sampled by a Γ -centered $1 \times 1 \times 1$ k-point mesh and a time step of 0.5 femtoseconds to ensure temporal precision.

For active learning, short AIMD simulations were conducted at 300 K using the NVT ensemble (constant particle number, volume, and temperature) for 2.5 ps. These runs provided the initial training set for the DeePMD and MTP active learning models. The dataset containing the training and testing data for both MTP and DeePMD are available under the link: [Active learning dataset](#). In contrast, the passive learning models were trained on the TiAlNb dataset from our previous work Chandran et al. (2024b), which comprises additional AIMD simulations carried out with both the NVT ensemble and the NPT (which maintains constant particle number, pressure, and temperature while allowing volume fluctuations) ensemble for approximately 10 ps across all configurations. For further details on the TiAlNb dataset, please refer to Chandran et al. (2024b).

For DeePMD potential training, the DeePMD-kit (Wang et al., 2018) was employed in conjunction with the LAMMPS package (Thompson et al., 2022) for MD simulations. Similarly, MTP potentials were trained using the MLIP package (Novikov et al., 2021), integrated with LAMMPS (Thompson et al., 2022) to

streamline MD simulations. These tools facilitated efficient and accurate training for both DeePMD and MTP models.

2.2 Moment tensor potential (MTP) and active learning strategy

The theoretical foundation of MTP and its associated moment tensor descriptors has been extensively discussed in our previous work [Chandran et al. \(2024b\)](#) and in other publications [Novikov et al. \(2021\)](#), [Podryabinkin et al. \(2023\)](#). We refer the reader to these resources for a detailed understanding of these concepts. This section will concentrate on the active learning formulations concerning MTP.

Traditional methods for constructing interatomic potential training sets often involve manual trial-and-error. However, by defining extrapolation mathematically, this process can be automated. The D-optimality criterion ([Novikov et al., 2021](#)), which maximizes the information matrix, introduces an extrapolation grade, denoted as $\gamma(\text{cfg})$. This grade correlates with prediction errors without requiring *ab initio* data. In active learning simulations, configurations exceeding a specified γ threshold are computed using an *ab initio* method and then added to the training set, thus reducing extrapolation.

The D-optimality criterion is particularly straightforward for linear regression, where MTP is parameterized by linear variables $\xi = \{\xi_1, \dots, \xi_m\}$. For a given configuration, the energy can be expressed as [Novikov et al. \(2021\)](#):

$$E^{\text{mtp}}(\text{cfg}; \xi) = \sum_i \sum_{\alpha=1}^m \xi_{\alpha} B_{\alpha}(\eta_i) = \sum_{\alpha=1}^m \xi_{\alpha} \sum_i B_{\alpha}(\eta_i) = \sum_{\alpha=1}^m \xi_{\alpha} \underbrace{\sum_i B_{\alpha}(\eta_i)}_{b_{\alpha}(\text{cfg})}. \quad (1)$$

Here B_{α} represents the basis functions and η_i corresponds to local atomic neighborhood of i^{th} atom. When fitting to energy values, an overdetermined system of K linear equations for ξ is solved, which can be represented in the following matrix form:

$$\mathbf{B} = \begin{pmatrix} b_1(\text{cfg}_1) & \cdots & b_m(\text{cfg}_1) \\ \vdots & \ddots & \vdots \\ b_1(\text{cfg}_K) & \cdots & b_m(\text{cfg}_K) \end{pmatrix}. \quad (2)$$

Each equation corresponds to a specific configuration. The D-optimality criterion selects m configurations that form the most linearly independent system of equations. This is equivalent to finding an $m \times m$ submatrix A with the largest determinant, $|\det(A)|$, which corresponds to the maximum volume. These configurations constitute the active set. Within the MLIP code, the active learning state is characterized by this active set, along with A and its inverse, A^{-1} . From the MTP perspective, this D-optimal active set contains the most “extreme” and diverse configurations. The Maxvol algorithm efficiently constructs this active set by choosing the submatrix with the greatest volume from a pool of configurations.

The extrapolation grade $\gamma(\text{cfg})$ for a given configuration is computed as:

$$\gamma(\text{cfg}) = \max_{1 \leq j \leq m} |c_j|, \quad (3)$$

where

$$(c_1 \dots c_m) = (b_1 \dots b_m) A^{-1}. \quad (4)$$

If $\gamma(\text{cfg}) > 1$, this indicates that $|\det(A)|$ could be increased. In such cases, the Maxvol algorithm replaces the row in A indexed by

$$k = \arg \max_{1 \leq j \leq m} |c_j|, \quad (5)$$

with the corresponding row from $(b_1 \dots b_m)$, and the active set is updated accordingly.

When the active set and the training set are the same, the parameters ξ can be determined by solving the system:

$$\xi = (E^{\text{qm}}(\text{cfg}_1) \dots E^{\text{qm}}(\text{cfg}_m)) A^{-1}. \quad (6)$$

Therefore, the energy of any configuration can be written as:

$$E^{\text{mtp}}(\text{cfg}) = \sum_{j=1}^m c_j E^{\text{qm}}(\text{cfg}_j). \quad (7)$$

This mathematical formulation shows that MTP is extrapolating if $\gamma(\text{cfg}) > 1$, and interpolating otherwise. Here E^{qm} corresponds to the quantum mechanical energy of configurations obtained using AIMD calculations. Thus, γ effectively serves as the extrapolation grade.

To extend the D-optimality criterion to MTP with nonlinear parameters, energies E^{mtp} are linearized with respect to the parameters $\bar{\theta}$, assuming these are close to their optimal values. In this scenario, the matrix $\mathbf{B}$ becomes a tall Jacobi matrix:

$$\mathbf{B} = \begin{pmatrix} \frac{\partial}{\partial \theta_1} E^{\text{mtp}}(\text{cfg}_1; \bar{\theta}) & \cdots & \frac{\partial}{\partial \theta_m} E^{\text{mtp}}(\text{cfg}_1; \bar{\theta}) \\ \vdots & \ddots & \vdots \\ \frac{\partial}{\partial \theta_1} E^{\text{mtp}}(\text{cfg}_K; \bar{\theta}) & \cdots & \frac{\partial}{\partial \theta_m} E^{\text{mtp}}(\text{cfg}_K; \bar{\theta}) \end{pmatrix}. \quad (8)$$

Each row of this matrix corresponds to a specific configuration in the training set, while the matrix $\mathbf{A}$ has a similar structure. The rest of the active learning algorithm remains the same as in the linear case. [Equations 1–8](#) pertain to the active learning strategy of MTP.

To assess the effectiveness of our active learning strategy, we conducted two types of comparisons. In Comparison 1, we evaluated the performance of active versus passive learning using an identical number of training configurations, thereby isolating the impact of the learning strategy on model accuracy. In Comparison 2, we varied the dataset sizes to determine the number of configurations required by each approach to achieve comparable RMSEs. While working with MTP, we found that attempting to train a single active learning model for both γ -TiAl and α_2 -Ti₃Al phases yielded unsatisfactory results. Consequently, we opted to perform active learning separately for each phase to better fine-tune the models.

For each phase, we employed a consistent active learning workflow that comprises two main phases: sampling and labeling. During the sampling phase, MD simulations are performed using the initial potential (trained on a limited sample size) to explore the configuration space—the set of possible atomic arrangements and states of the system. Configurations where the model fails to accurately predict energies or forces are identified. In the labeling phase, these selected configurations are accurately annotated using

DFT simulations, and the new data is incorporated into the training set, thereby refining the potential.

The initial training set comprised 170 configurations extracted from short NVT simulations of 2.5 ps at 300 K. We initiated the training with a 10.mtp (potential model with level 10) model and divided the active learning process into four steps. Steps 1, 2, and 3 corresponded to Comparison 1, using an equal number of samples for both learning strategies, while Step 4 extended the process for Comparison 2 by incorporating datasets of varying sizes. In step 1, MD simulations were conducted using the NVT ensemble at 300 K across different Nb concentrations, with iterations continuing until no new configurations were added to the training set. In Step 2, the exploration temperature range was expanded to 300 K, 500 K, 700 K, and 900 K, and the iterative process was repeated until no new configurations were chosen. In Step 3, we upgraded the model to 20.mtp, refined the k-point mesh to $2 \times 2 \times 2$ for DFT labeling, and pruned half of the configurations obtained with a coarser k-point mesh during the previous steps. This procedure follows the methodology outlined in Luo et al. (2023), Novikov et al. (2021), Bock et al. (2024). The iterations continued until no new configurations were selected. Finally, in Step 4, we further upgraded the model to 24.mtp and repeated the active learning iterations across the full temperature spectrum (300 K–900 K), again until the training set stabilized.

For MD exploration, we utilized LAMMPS (Thompson et al., 2022), while VASP (Kresse and Furthmüller, 1996b; Kresse and Furthmüller, 199a) was employed for the *ab initio* calculations.

2.3 DeePMD and active learning strategy

The theoretical foundations of DeePMD have been discussed extensively in our previous work Chandran et al. (2024b) and elsewhere Zhang et al. (2019). Here, we focus on the active learning aspects. As with MTP, we performed two types of comparisons: in Comparison 1, we evaluated the performance of active versus passive learning using an identical number of training configurations; in Comparison 2, we varied the dataset sizes to determine how many configurations each approach requires to achieve comparable RMSEs. Unlike the MTP approach—where separate active learning processes were conducted for γ -TiAl and α_2 -Ti₃Al—the DeePMD method incorporated both phases together.

For DeePMD, we began with a larger initial dataset of 40,000 configurations (20,000 for γ -TiAl and 20,000 for α_2 -Ti₃Al), since deep neural networks typically require more training data than linear regression-based models. This dataset was assembled from snapshots of short AIMD simulations (2.5 ps at 300 K using the NVT ensemble), which provided the initial training data for the active learning procedure.

During the sampling phase, MD simulations were conducted at 300 K, 500 K, 700 K, and 900 K using both the NVT and NPT ensembles for all Nb concentration cases across both phases. For Comparison 1, active learning iterations were performed over 80 cycles. The process began with 300 K NVT simulations, then transitioned to 300 K NPT simulations, with the number of simulation steps increasing as the iterations progressed, and eventually extending to higher temperatures. The production model of potential for Comparison 1 was obtained with a training run of $2 \times$

10^6 steps. By the end of these iterations, the RMSEs for energy/atom and force were within acceptable limits. For Comparison 2, the exploration was extended by an additional 20 iterations, focusing on high-temperature configurations until the potential error was further reduced. As with Comparison 1, the production model of potential for Comparison 2 was obtained using a training run of 2×10^6 steps.

To assess the reliability of atomic force predictions, we used the concept of model deviation (Zhang et al., 2019), denoted as $\mathcal{E}$ (Equation 9), which is defined as the maximum standard deviation of the predicted atomic forces across multiple models initialized with different parameters:

$$\mathcal{E} = \max_i \sqrt{\|\mathbf{f}_i - \hat{\mathbf{f}}_i\|^2}, \quad \mathbf{f}_i = \langle \hat{\mathbf{f}}_i \rangle. \quad (9)$$

Here, $\mathbf{f}_i$ represents the atomic force prediction for atom i from a specific model, and $\hat{\mathbf{f}}_i$ is the average prediction over all models. The notation $\langle \dots \rangle$ represents the ensemble average, which is computed across the set of models in the ensemble. Using the maximum standard deviation rather than an average is crucial because atomic forces are highly sensitive to local variations, making this metric more effective at identifying localized prediction failures that might be overlooked by global energy evaluations (Zhang et al., 2019).

Selected configurations exhibiting high model deviation were subsequently labeled using DFT simulations. These DFT calculations employed projector augmented wave (PAW) potentials (Blöchl, 1994) to accurately model electron–ion interactions, along with the generalized gradient approximation (GGA) (Perdew et al., 1996; Perdew et al., 1997) using the Perdew–Burke–Ernzerhof (PBE) functional. A cutoff energy of 510 eV and a k-spacing of $0.2 \text{ \AA}^{-1}$ were used to ensure precision.

The DeePMD model architecture comprised radial and angular embedded-atom neural networks, each with three hidden layers containing 25, 50, and 100 nodes, respectively, to effectively capture complex interatomic interactions. Additionally, the fitting networks were constructed with three hidden layers of 240 nodes each. The training process began with an initial learning rate of 0.001, which was gradually reduced to 3.51×10^{-8} to allow for fine adjustments and improved predictions. Weighting factors were also carefully calibrated: energy weights were initially set at 0.02 and increased to 1, while force weights started at 1,000 and were reduced to 1, thereby prioritizing force accuracy in the early stages before shifting focus toward energy accuracy.

2.4 Simulated tension tests

To investigate the influence of Nb on the thermomechanical properties of the alloy, we performed uniaxial tension tests using MD models of both the γ -TiAl and α_2 -Ti₃Al phases at varying Nb concentrations. In these simulations, periodic boundary conditions were imposed in all directions, and uniaxial tension was applied along the Z-axis at a strain rate of 10^9 s^{-1} . Prior to loading, each system was equilibrated at 300 K using an NVT ensemble, after which the uniaxial stress was applied along the Z-direction.

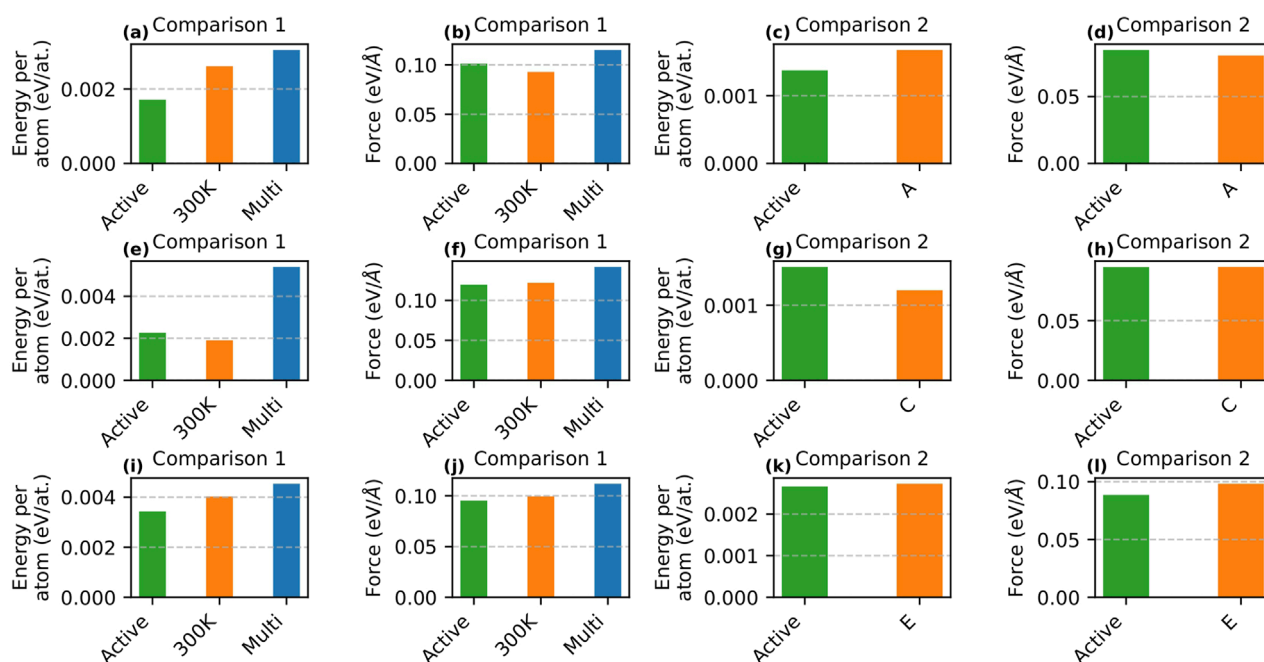


FIGURE 2

Comparison of RMSE energy/atom and force on the testing dataset during active learning using MTP and DeePMD. Panels (a–d) show MTP results for the γ -TiAl phase, and panels (e–h) for the α_2 -Ti₃Al phase; panels (i–l) display DeePMD results. “Active” refers to the active learning approach. In Comparison 1, the active learning potential is evaluated against two passive learning potentials trained with the same number of samples: one at 300 K (labeled 300 K) and one with multi-temperature data (300 K, 500 K, 700 K, and 900 K, labeled Multi). For Comparison 1, MTP used 3,094 samples for γ -TiAl and 1,665 for α_2 -Ti₃Al, while DeePMD used 57,180 samples. Comparison 2 examines the effect of dataset size by comparing the active learning potential with passive learning potentials trained on Dataset-A and Dataset-C (20,000 samples) for MTP, and on Dataset-E (127,212 samples) for DeePMD. For Comparison 2, active learning for MTP used 3,322 samples for γ -TiAl and 2,602 for α_2 -Ti₃Al, while DeePMD required 61,657 samples.

3 Results and discussion

3.1 Error analysis

This section presents the error analysis for the machine learning potentials developed for γ -TiAl and α_2 -Ti₃Al, comparing their performance against passive learning approaches.

3.1.1 MTP

Figures 2a–h presents the detailed error analysis for the active learning performed using MTP. Panels a, b, c, and d show the RMSEs for energy/atom and force for Comparison 1 and Comparison 2 in the γ -TiAl phase, while panels e, f, g, and h display the corresponding RMSEs for the α_2 -Ti₃Al phase using MTP. The learning curves for MTP in both phases are shown in Supplementary Figure S1 in the supplementary section.

For the γ -TiAl phase, we began with a simple 10.mtp model and performed MD sampling up to 15 iterations. During iterations 1–5, only NVT simulations at 300 K were employed. From iteration 6, NVT simulations covering a temperature range of 300–900 K were included. At iteration 15, no additional configurations were added to the training set. Starting from iteration 16, we transitioned to a more complex 20.mtp model and continued exploring configuration space with NVT simulations at various temperature ranges. A finer k-point mesh of $2 \times 2 \times 2$ was used for DFT labeling. Iterations up to 33 were considered for Comparison 1. Subsequently, from

iteration 34, we switched to a 24.mtp model (Comparison 2) and continued until iteration 36, where no new configurations were selected. The transition to 20.mtp and the finer k-point mesh led to a noticeable drop in RMSE for energy and force values of γ -TiAl, as shown in Supplementary Figure S1 in the supplementary section. By iteration 33, the training set contained 3,094 configurations, which increased to 3,322 configurations by iteration 36.

To evaluate the effectiveness of the active learning strategy, we performed passive learning for Comparison 1 using 3,094 configurations under two scenarios. In the first scenario, only 300 K configurations were included in the training set to replicate the initial conditions of active learning. In the second scenario, configurations spanning a range of temperatures (300 K–900 K) were used. The RMSEs for energy/atom and force for all three cases (active learning, passive learning with 300 K configurations, and passive learning with multi-temperature configurations) are shown in Figures 2a,b. The results indicate that active learning achieved the lowest RMSE for energy/atom (1.71×10^{-3} eV/atom), followed by passive learning with 300 K configurations (2.61×10^{-3} eV/atom) and passive learning with multi-temperature configurations (3.04×10^{-3} eV/atom). In contrast, the RMSE for force from active learning (101.35×10^{-3} eV/Å) was slightly higher than that from passive learning with 300 K configurations (92.83×10^{-3} eV/Å), but lower than that from passive learning with multi-temperature configurations (115.18×10^{-3} eV/Å). The higher RMSE

for passive learning with multi-temperature configurations is attributed to thermodynamic noise from high-temperature frames.

For Comparison 2, we examined whether a passive learning model trained on a larger dataset could match or exceed the predictive accuracy (in terms of RMSE) of active learning. At this stage, the active learning model was trained using 3,322 samples. Initially, passive learning was performed using Dataset-A (20,000 samples). Although the errors obtained from Dataset-A were comparable to those from active learning, we further investigated whether increasing the sample size with Dataset-B (80,000 samples) could reduce the error. Figures 2c,d compare the performance of active learning and passive learning using Dataset-A, while the RMSE values for Dataset-B are provided in Supplementary Table S1 in the supplementary section. Note that the passive learning models for Comparison 2 utilized a 24.mtp model. As shown in Figures 2c,d, active learning produced a lower RMSE for energy/atom (1.37×10^{-3} eV/atom) than passive learning with Dataset-A (1.67×10^{-3} eV/atom). However, the force RMSE from active learning (84.72×10^{-3} eV/Å) was slightly higher than that from passive learning (80.59×10^{-3} eV/Å). Furthermore, passive learning with Dataset-B resulted in errors similar to those of both active learning and passive learning with Dataset-A, yielding RMSE values of 1.32×10^{-3} eV/atom for energy/atom and 80.14×10^{-3} eV/Å for force, indicating that expanding the sample size to 80,000 did not significantly reduce the error values.

For the α_2 -Ti₃Al phase, a similar active learning strategy as that of γ -TiAl was applied. The learning curves for Comparison 1 and Comparison 2 are shown in Supplementary Figure S1 in the supplementary section. Initial training involved 300 K samples starting with a 10.mtp model up to iteration 18. From iteration 19 onward, configurations from a broader temperature range (300 K–900 K) were included, continuing until iteration 48 when no new configurations were selected. At this point, we transitioned to a 20.mtp model, applied a finer k-point mesh ($2 \times 2 \times 2$), and resumed exploration. A significant drop in RMSE for energy/atom was observed, although the decrease in RMSE for force was less pronounced. At iteration 61, the training set contained 1,665 configurations, which were used for Comparison 1. From iteration 62 onward, we adopted a 24.mtp model and concluded at iteration 65 with 2,602 configurations in the training set, used for Comparison 2.

For Comparison 1, we compared active learning with passive learning using 1,665 configurations. The passive learning scenarios included both 300 K-only and multi-temperature samples. As shown in Figures 2e,f, active learning yielded a slightly higher RMSE for energy/atom (2.26×10^{-3} eV/atom) than passive learning with 300 K samples (1.91×10^{-3} eV/atom), yet a markedly lower RMSE compared to multi-temperature samples (5.39×10^{-3} eV/atom). Similarly, active learning produced a lower RMSE for force (119.56×10^{-3} eV/Å) than both passive learning with 300 K samples (121.90×10^{-3} eV/Å) and multi-temperature samples (141.81×10^{-3} eV/Å). The higher RMSE observed for passive learning with multi-temperature configurations is because of the thermodynamic noise from high-temperature frames.

For Comparison 2, passive learning was conducted using Dataset-C (comprising 20,000 samples), while active learning was performed with 2,602 samples. Similar to the γ -TiAl case, although Dataset-C produced errors comparable to those obtained from active learning, Dataset-D (comprising 80,000 samples) was also

employed to assess how the errors change with an increased sample size. The RMSE comparisons between active learning and passive learning using Dataset-C are presented in Figures 2g,h, while the errors from Dataset-D are tabulated in Supplementary Table S1 in the supplementary section. As illustrated in Figures 2g,h, active learning resulted in a slightly higher RMSE for energy/atom (1.51×10^{-3} eV/atom) compared to passive learning using Dataset-C (1.20×10^{-3} eV/atom). However, for force, active learning (94.94×10^{-3} eV/Å) achieved a slightly lower RMSE than passive learning using Dataset-C (95.06×10^{-3} eV/Å). Furthermore, Dataset-D produced errors similar to those of both active learning and passive learning with Dataset-C (energy/atom: 1.23×10^{-3} eV/atom; force: 94.44×10^{-3} eV/Å), indicating that increasing the sample size to 80,000 did not substantially improve the error values.

3.1.2 DeePMD

Here, we explore the comparison between active learning of DeePMD and passive learning using Comparison 1 and Comparison 2. The learning curves are presented in Supplementary Figure S2 in the supplementary section. Initially, we used 20,000 samples each from the γ -TiAl and α_2 -Ti₃Al phases. Exploration began with NVT simulations for both phases at 300 K for 10 iterations, followed by NPT simulations at 300 K for another 10 iterations. Subsequently, higher temperature simulations were introduced, combining NVT and NPT simulations for MD exploration every 20 iterations. By the end of 80 iterations, the potential exhibited sufficiently low RMSE for energy/atom and force, was selected for Comparison 1. At this stage, the training data contained 57,180 samples. We continued with 20 more iterations using higher-temperature NVT and NPT simulations to ensure the potentials were adequately trained on high-temperature samples until the error values of the potential becomes significantly lower than that in Comparison 1. The training data then contained 61,657 samples.

Figures 2i–l show the RMSEs for energy/atom and force for active learning potentials and their passive learning counterparts for Comparison 1 and Comparison 2, respectively. Similar to the case of MTP, for Comparison 1, we performed passive learning with the same number of samples as active learning (57,180 samples). Two scenarios were considered: one with only 300 K samples (matching the starting conditions of active learning) and the other with multi-temperature samples. The RMSE values for the active learning potential (RMSE energy/atom = 3.43×10^{-3} eV/atom, RMSE force = 95.27×10^{-3} eV/Å) were lower than those for both passive learning cases (300 K samples: RMSE energy/atom = 4.03×10^{-3} eV/atom, RMSE force = 99.41×10^{-3} eV/Å; multi-temperature samples: RMSE energy/atom = 4.54×10^{-3} eV/atom, RMSE force = 111.89×10^{-3} eV/Å). Similar to the MTP case, the increased RMSE observed in passive learning with multi-temperature configurations can be due to thermodynamic noise arising from high-temperature frames.

For Comparison 2, we conducted passive learning using Dataset-E, which comprises 127,212 samples. As in the MTP approach, although Dataset-E produced errors similar to those from active learning, we increased the sample size to create Dataset-F (333,340 samples) to examine whether a larger dataset would yield further error reduction. Note that active learning was performed using 61,657 samples. The RMSE comparisons are presented in Figures 2k,l, while the errors for Dataset-F are provided in Supplementary Table S1 in the supplementary section.

The RMSE energy/atom values for active learning (2.66×10^{-3} eV/atom) and passive learning with Dataset-E (2.73×10^{-3} eV/atom) are nearly identical, whereas the RMSE force for active learning (88.53×10^{-3} eV/Å) is slightly lower than that for passive learning with Dataset-E (98.22×10^{-3} eV/Å). In contrast, passive learning with Dataset-F clearly outperforms both active learning and passive learning with Dataset-E, achieving an RMSE energy/atom of 1.07×10^{-3} eV/atom and an RMSE force of 93.14×10^{-3} eV/Å. In the case of DeePMD, further increasing the sample size results in an additional decrease in the error.

3.2 MD results

In this section, we summarize the results of MD simulations performed using the potentials derived from both active and passive learning. In Section 3.1, we assessed the RMSEs of the active and passive learning potentials under two scenarios—Comparison 1 and Comparison 2. Here, we present the MD simulation results obtained with these potentials to evaluate their effectiveness in predicting the fundamental material and thermo-mechanical properties of the two phases.

For Comparison 1, we utilized the passive learning potential trained on multi-temperature samples, which provides a fair comparison with active learning when exploring high-temperature properties. For Comparison 2, we selected the active learning cases using Dataset-A (γ -TiAl) and Dataset-C (α_2 -Ti₃Al) in MTP, as well as Dataset-E in DeePMD, all of which achieved performance comparable to that of active learning in terms of RMSE for energy/atom and force.

3.2.1 Energy volume curve and elastic constants

In this section, we evaluate the energy-volume (EV) curves obtained via the Murnaghan fit to compare the performance of passive and active learning. Figures 3, 4 display the EV curves for Comparison 1 and Comparison 2, respectively, alongside the corresponding DFT curves for MTP and DeePMD.

Here we focus on MTP. For Comparison 1, the passive learning potential failed to find a minimum volume, and therefore, the results for passive learning are not included here. For γ -TiAl, the equilibrium volume predicted by the active learning potential was 16.91 Å^3 , compared to 16.22 Å^3 obtained from DFT. The equilibrium lattice parameters were $a = 3.99 \text{ Å}$ and $c = 4.18 \text{ Å}$ for the active learning potential, while the DFT values were $a = 3.95 \text{ Å}$ and $c = 4.14 \text{ Å}$. For α_2 -Ti₃Al, the equilibrium volume predicted by the active learning potential (17.48 Å^3) deviated more from the DFT value (15.88 Å^3). The predicted lattice parameters were $a = 5.79 \text{ Å}$ and $c = 4.73 \text{ Å}$, whereas DFT values were $a = 5.62 \text{ Å}$ and $c = 4.59 \text{ Å}$.

For Comparison 2, the equilibrium volume for γ -TiAl for active learning, though over-predicted, remained the same as in Comparison 1, while the passive learning potential predicted 15.97 Å^3 , which under-predicted the DFT value. For α_2 -Ti₃Al, the equilibrium volume predicted by active learning was 16.95 Å^3 , while passive learning predicted 16.61 Å^3 , which was closer to the DFT value. The equilibrium lattice parameters predicted by active learning for γ -TiAl were $a = 3.99 \text{ Å}$ and $c = 4.18 \text{ Å}$,

while passive learning predicted $a = 3.95 \text{ Å}$ and $c = 4.14 \text{ Å}$. For α_2 -Ti₃Al, the predicted lattice parameters were $a = 5.79 \text{ Å}$ and $c = 4.73 \text{ Å}$ for active learning, and $a = 5.74 \text{ Å}$ and $c = 4.68 \text{ Å}$ for passive learning.

When analyzing DeePMD, Figure 4 compares the EV curves for active and passive learning for both Comparison 1 and Comparison 2. Similar to the case of γ -TiAl the passive learning potential failed to accurately find a minimum volume. For γ -TiAl, the equilibrium volume predicted by the active learning potential was 17 Å^3 , and the equilibrium lattice parameters were $a = 4.03 \text{ Å}$ and $c = 4.22 \text{ Å}$. These values are farther from the DFT predictions compared to the MTP case. For α_2 -Ti₃Al, the equilibrium volume predicted by active learning (16.13 Å^3) was closer to the DFT value (15.88 Å^3) than in the MTP case, with lattice parameters $a = 5.68 \text{ Å}$ and $c = 4.63 \text{ Å}$.

In Comparison 2, the equilibrium volume predicted for γ -TiAl by active learning was 15.80 Å^3 , and that by passive learning was 15.55 Å^3 . Active learning was closer to the DFT prediction. The equilibrium lattice parameters for γ -TiAl predicted by active learning were $a = 3.91 \text{ Å}$ and $c = 4.10 \text{ Å}$, matching passive learning predictions. For α_2 -Ti₃Al, the equilibrium volume predicted by active learning was 16.27 Å^3 , closer to DFT than the passive learning value of 14.95 Å^3 . The lattice parameters predicted by active learning were $a = 5.68 \text{ Å}$ and $c = 4.63 \text{ Å}$, while those predicted by passive learning were $a = 5.51 \text{ Å}$ and $c = 4.49 \text{ Å}$, which are closer to DFT values than the active learning predictions.

Along with evaluating equilibrium volume and lattice parameters, a comparative analysis of elastic constants was performed for both γ -TiAl and α_2 -Ti₃Al using DFT, active learning, and passive learning potentials at 300 K. These analyses were carried out with both MTP and DeePMD (refer Figure 5) models. For reference the additional plots showing the absolute differences between the elastic constants predicted by the passive and active learning potentials with respect to those predicted by DFT are included in Supplementary Figure S3 in the supplementary section.

For MTP, in the case of γ -TiAl, active learning outperformed passive learning in predicting higher elastic constants such as C_{11} , C_{12} and C_{66} , with values closer to those from DFT in both Comparison 1 (where both learning methods were trained on the same number of samples) and Comparison 2 (where different dataset sizes were used to achieve comparable errors). However, passive learning showed better agreement for constants like C_{33} especially in Comparison 1. In Comparison 2, while both methods improved their alignment with DFT (see Supplementary Figure S3 in the supplementary section), active learning provided more balanced performance across all constants, even though passive learning occasionally achieved closer predictions for C_{33} . For α_2 -Ti₃Al, active learning generally predicted elastic constants further from DFT values compared to passive learning, as highlighted in Supplementary Figures S3c, d in the supplementary section. Nevertheless, active learning and passive learning results aligned with DFT in the case of C_{33} and C_{44} .

For DeePMD, in the case of γ -TiAl, passive learning exhibited significant deviations from DFT in Comparison 1—most notably, C_{11} was overpredicted and active learning struggled with elastic constants such as C_{13} and C_{33} . In Comparison 2, both learning approaches showed improved agreement with DFT; active learning was more consistent for C_{33} and C_{13} . For α_2 -Ti₃Al, active learning outperformed passive learning in Comparison 1 for

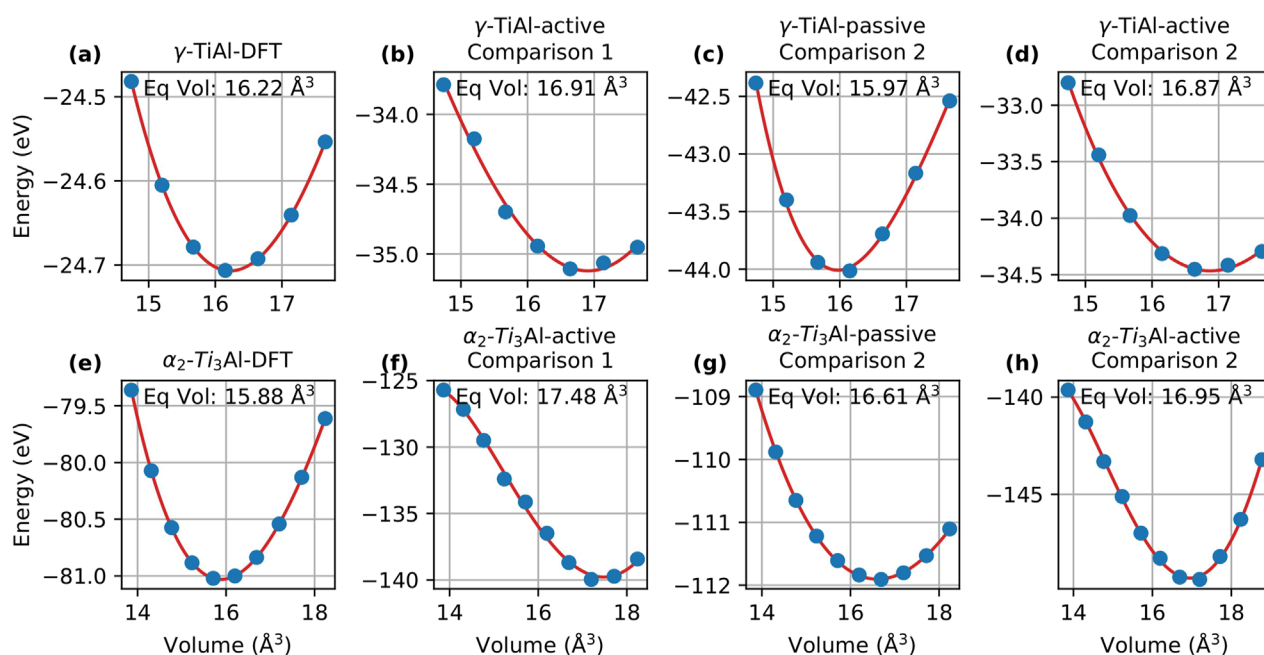


FIGURE 3 Energy-volume curves for γ -TiAl (a–d) and α_2 -Ti₃Al (e–h) obtained using DFT, an active learning potential, and a passive learning potential developed with MTP. The curves correspond to Comparison 1 and Comparison 2. In Comparison 1, both potentials were trained with the same number of samples to enable a direct performance comparison, whereas in Comparison 2, different training dataset sizes were employed to achieve comparable error levels.

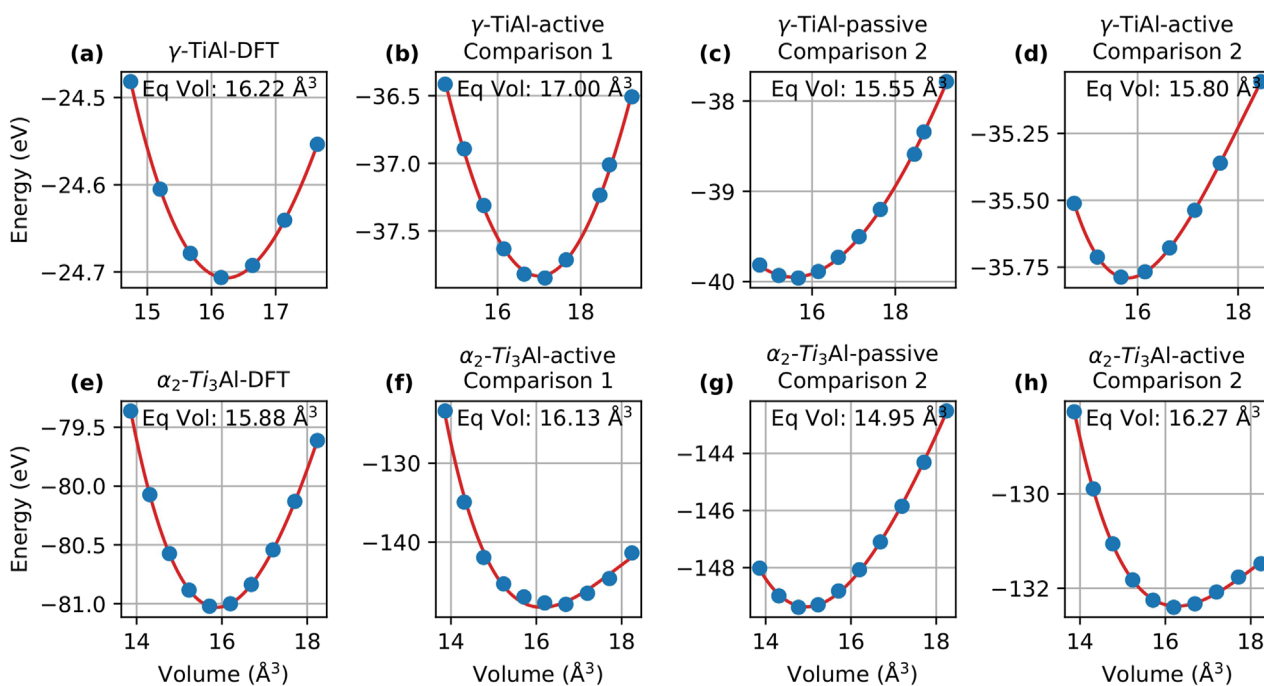


FIGURE 4 Energy-volume curves for γ -TiAl (a–d) and α_2 -Ti₃Al (e–h) obtained using DFT, an active learning potential, and a passive learning potential developed with DeePMD. The curves correspond to Comparison 1 and Comparison 2. In Comparison 1, both potentials were trained with the same number of samples to enable a direct performance comparison, whereas in Comparison 2, different training dataset sizes were employed to achieve comparable error levels.

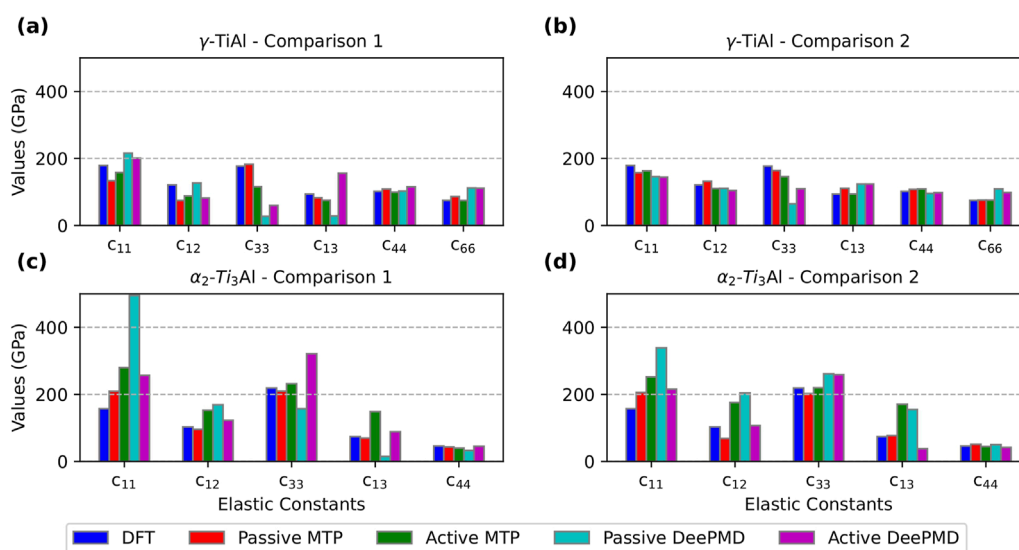


FIGURE 5

Comparison of elastic constants calculated using DFT, passive learning, and active learning potentials developed with MTP and DeePMD. (a,b) correspond to γ -TiAl, while (c,d) correspond to α_2 -Ti₃Al. In Comparison 1, both active and passive learning potentials were trained with the same number of samples to enable a direct performance evaluation. In Comparison 2, the training dataset sizes were adjusted to achieve comparable error levels. Note that the unit for elastic constants is GigaPascal (GPa) and they are computed at 300 K.

elastic constants such as C_{12} and C_{13} , whereas passive learning overpredicted higher constants like C_{11} . Moreover, compared to the MTP results, the active learning potentials developed with DeePMD provided predictions for α_2 -Ti₃Al that were closer to DFT (see [Supplementary Figure S3](#) in supplementary section). In Comparison 2, both active and passive learning approaches exhibited improved accuracy. Active learning achieved superior predictions for C_{12} , while for C_{44} both methods demonstrated marginally better agreement.

Overall, active learning consistently demonstrated robust predictive capability for fundamental material properties—including lattice parameters, equilibrium volume, and elastic constants—compared to passive learning. However, passive learning exhibited greater variability and a tendency to overpredict, particularly in Comparison 1. It is also noteworthy that active learning produced some outliers, especially for the MTP active learning potential in α_2 -Ti₃Al elastic constants and for certain equilibrium volume and lattice parameter predictions. This may be attributed to the fact that these basic properties were not the primary focus during the active learning sample selection process, leading to a bias towards higher temperature configurations that deviate from the base state, thus affecting the overall prediction accuracy.

3.2.2 Finite temperature results

To evaluate the finite-temperature properties and compare the performance of active and passive learning for MTP and DeePMD, we computed the radial distribution functions (RDFs) and compared them with AIMD results. The RDFs were evaluated for both γ -TiAl and α_2 -Ti₃Al using configurations that were not included in the training set.

The RDF comparisons for γ -TiAl obtained with MTP and DeePMD (Comparison 1) are shown in [Figure 6](#), while [Figure 7](#) presents the corresponding comparisons for α_2 -Ti₃Al. Similarly, the RDF comparisons for Comparison 2 are provided in the [Supplementary Material](#) (see [Supplementary Figure S4](#); [Supplementary Figure S5](#)).

RDFs provide a quantitative measure of how accurately a potential predicts the atomic arrangement in a material. In this study, RDFs were computed over a range of temperatures to evaluate the performance of the potentials from low to high temperatures. As shown in [Figure 6](#) for γ -TiAl, the RDFs predicted by both passive and active learning using MTP and DeePMD are nearly identical, which is expected since γ -TiAl is relatively straightforward for the potentials to model. In contrast, [Figure 7](#) indicates that α_2 -Ti₃Al poses a greater challenge for accurate prediction, particularly for passively trained potentials. This is most evident in the noticeable discrepancies that appear at 700 K ([Figure 7g](#)). This intermediate temperature allows the system to access complex, high-energy atomic configurations that may be statistically underrepresented in a randomly sampled passive learning dataset. The failure of the passively trained MTP potential to maintain the correct structure suggests it is extrapolating poorly into these unlearned regions. In contrast, the comparatively good performance of the active learning potentials ([Figure 7c](#)) demonstrates their ability to identify and specifically learn from these challenging configurations, resulting in a more stable and physically accurate model across the wider temperature range.

For Comparison 2 (refer [Supplementary Figure S4](#); [Supplementary Figure S5](#) in the supplementary section), the RDF curves show a good match with AIMD results for both passive and active learning. This is in line with our expectations, as our

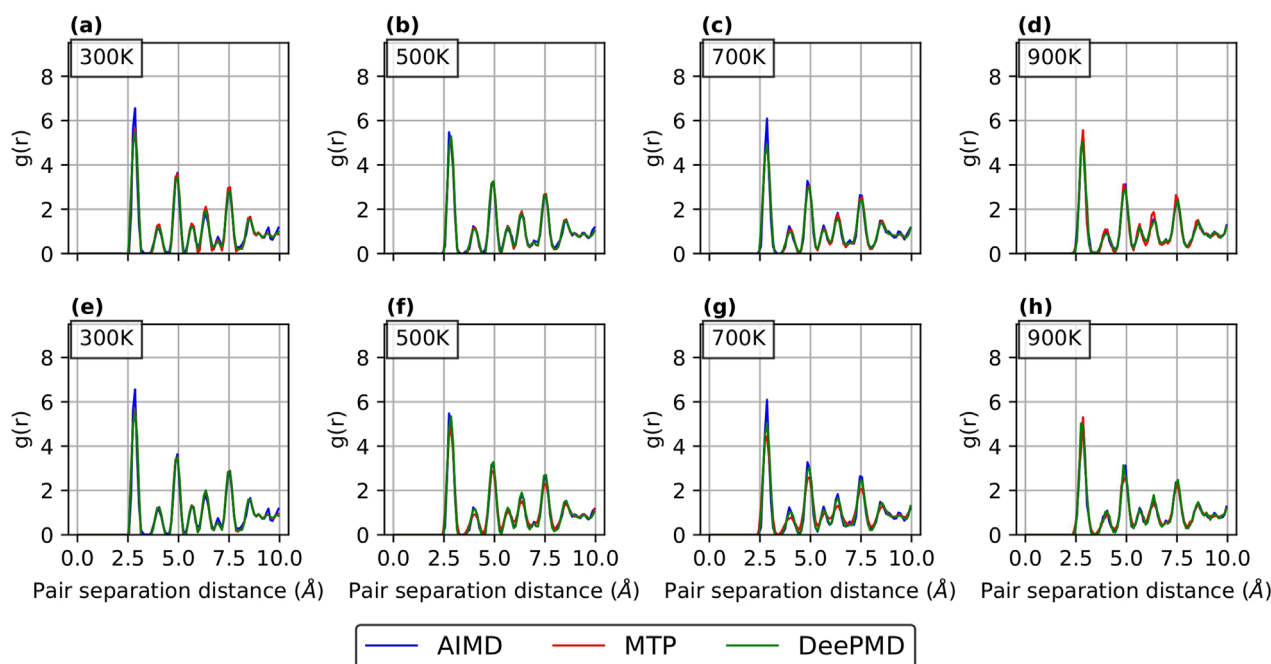


FIGURE 6

RDF curves for γ -TiAl with a 5.6 at.% Nb configuration obtained from passive and active learning potentials, compared with AIMD results at 300 K, 500 K, 700 K, and 900 K for both MTP and DeePMD. The top row (a–d) shows the AIMD and active learning potential results, while the bottom row (e–h) presents the curves from the passive learning potential. These results correspond to Comparison 1, where both active and passive learning potentials were trained using the same number of samples, allowing for a direct evaluation of their relative performance.

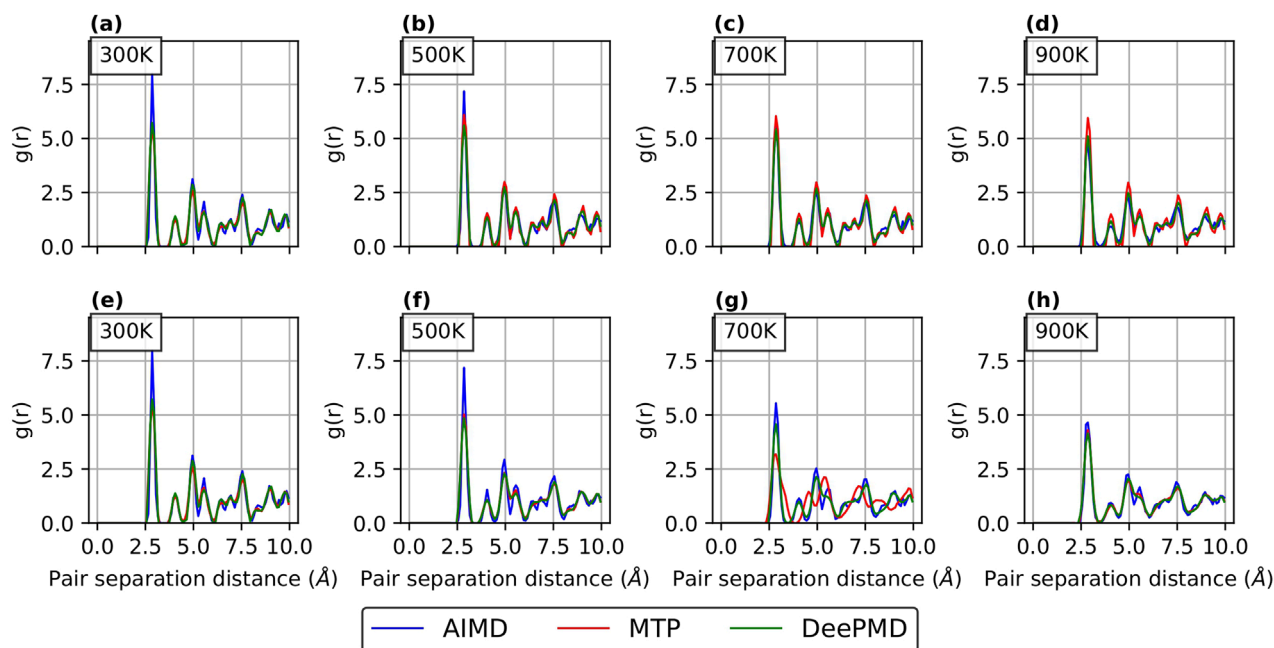


FIGURE 7

RDF curves for α_2 -Ti₃Al with a 2.3 at.% Nb configuration obtained from passive and active learning potentials, compared with AIMD results at 300 K, 500 K, 700 K, and 900 K for both MTP and DeePMD. The top row (a–d) shows the AIMD and active learning potential results, while the bottom row (e–h) presents the curves from the passive learning potential. These results correspond to Comparison 1, where both active and passive learning potentials were trained using the same number of samples, allowing for a direct evaluation of their relative performance.

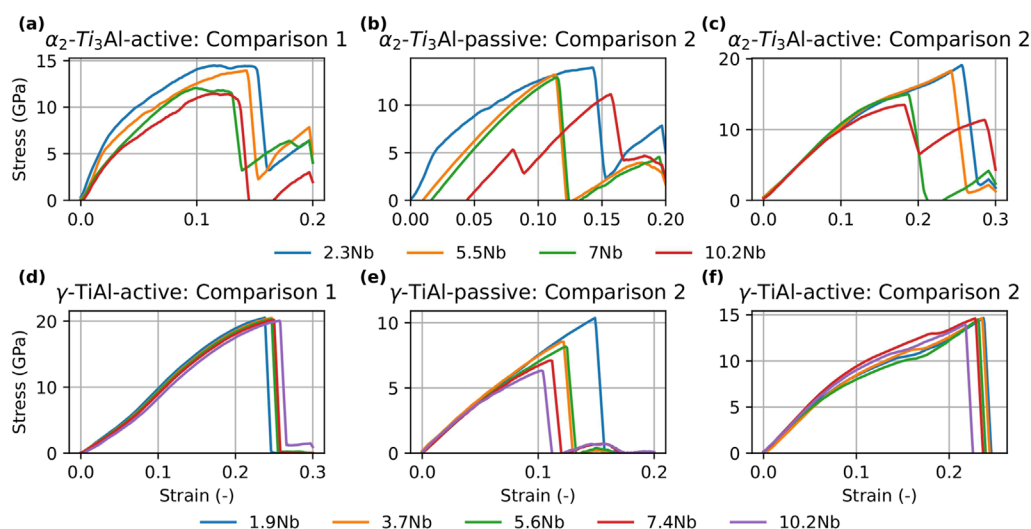


FIGURE 8

Stress-strain curves for α_2 -Ti₃Al (a–c) and γ -TiAl (d–f) at varying Nb concentrations, obtained using active and passive learning potentials developed with MTP. All simulations were conducted at a strain rate of 10^9 s^{-1} . Results are shown for both Comparison 1 and Comparison 2. In Comparison 1, both potentials were trained with the same number of samples to enable a direct performance evaluation, while in Comparison 2 the training dataset sizes were adjusted to achieve comparable error levels.

primary aim in Comparison 2 was to employ different sample sizes for active and passive learning while achieving comparable errors.

3.2.3 Tension tests and generalized stacking fault energy

In this study, we evaluated the performance of MTP and DeePMD potentials in capturing the thermo-mechanical behavior of our target phases (γ -TiAl and α_2 -Ti₃Al). To this end, we simulated uniaxial tension tests on selected samples at a strain rate of 10^9 s^{-1} . The detailed method is already discussed in Section 2.4. The stress-strain curves obtained via active learning for both the γ -TiAl and α_2 -Ti₃Al phases for Comparison 1 and Comparison 2 are illustrated in Figures 8, 9. Our analysis focused primarily on ultimate tensile strength (UTS) to evaluate the accuracy of the mechanical property predictions. For the MTP potentials, a consistent decrease in UTS with increasing Nb concentration was observed in both Comparison 1 and Comparison 2, a trend evident for both γ -TiAl and α_2 -Ti₃Al phases using MTP and DeePMD potentials. Notably, under active learning in Comparison 1 for MTP, the UTS values for γ -TiAl at 3.7 at.% Nb, 5.6 at.% Nb, and 7.4 at.% Nb were very similar (Figure 8d), and a similar pattern was observed in Comparison 2 (Figure 8f) for 1.9 at.% Nb, 3.7 at.% Nb, and 5.6 at.% Nb. Overall, a significant reduction in UTS with increasing Nb concentration was found for both phases, and this trend was consistent across active and passive learning cases. These observations align with our earlier findings Chandran et al. (2024a) and with results reported in other studies Dumitraschkewitz et al. (2017), Lu et al. (2023), Qi et al. (2023). The stress-strain curves of DeePMD (Figure 9) reveal that the α_2 -Ti₃Al phase at high Nb concentrations is particularly challenging to model accurately. The unphysical fluctuations seen in Figure 9a and c are the most severe for the highest Nb content. This suggests that the combination of the complex D0₁₉ lattice and the

significant local distortions introduced by a high concentration of Nb atoms creates unique, high-energy configurations under strain. The relative stability of the γ -TiAl phase across all concentrations underscores the difficulty of modeling the α_2 -Ti₃Al phase with high Nb concentration.

In addition to the tension tests, we computed the generalized stacking fault energy (GSFE) for both γ -TiAl and α_2 -Ti₃Al using active learning potentials. For the MTP approach, the active learning GSFE results were compared with those reported in Qi et al. (2023), where MTP was employed to calculate the stacking fault energies for these phases, and were further benchmarked against the corresponding DFT values from the same study. In the case of DeePMD, due to the absence of previous GSFE studies using this method, our results were compared solely with the DFT values reported in Qi et al. (2023).

For γ -TiAl, dislocation glide occurs on the {111} close-packed planes. Here, the complex stacking fault (CSF) is associated with ordinary $\langle \bar{1}10 \rangle/2$ dislocations, while the antiphase boundary (APB) is linked to $\langle 11\bar{2} \rangle/2$ super-dislocations. (Note that we did not compute the intrinsic stacking fault (SISF) for γ -TiAl.) The GSFE values obtained from active learning using both MTP and DeePMD, at two stages (Comparison 1 and Comparison 2), are provided in Tables 1, 2. It is evident that the energy values from Comparison 2 are closer to the reference and DFT values. This improvement in accuracy can be attributed to the lower energy/atom and force RMSEs of the active learning potential in Comparison 2 versus Comparison 1, as shown in Figure 2. Moreover, an increase in Nb concentration leads to a noticeable reduction in the energies for both CSF and APB, consistent with the findings of Dumitraschkewitz et al. (2017). Generally, a higher GSFE indicates greater resistance to dislocation motion and reduced dislocation mobility, which directly enhances the strengthening behavior in nanomaterials.

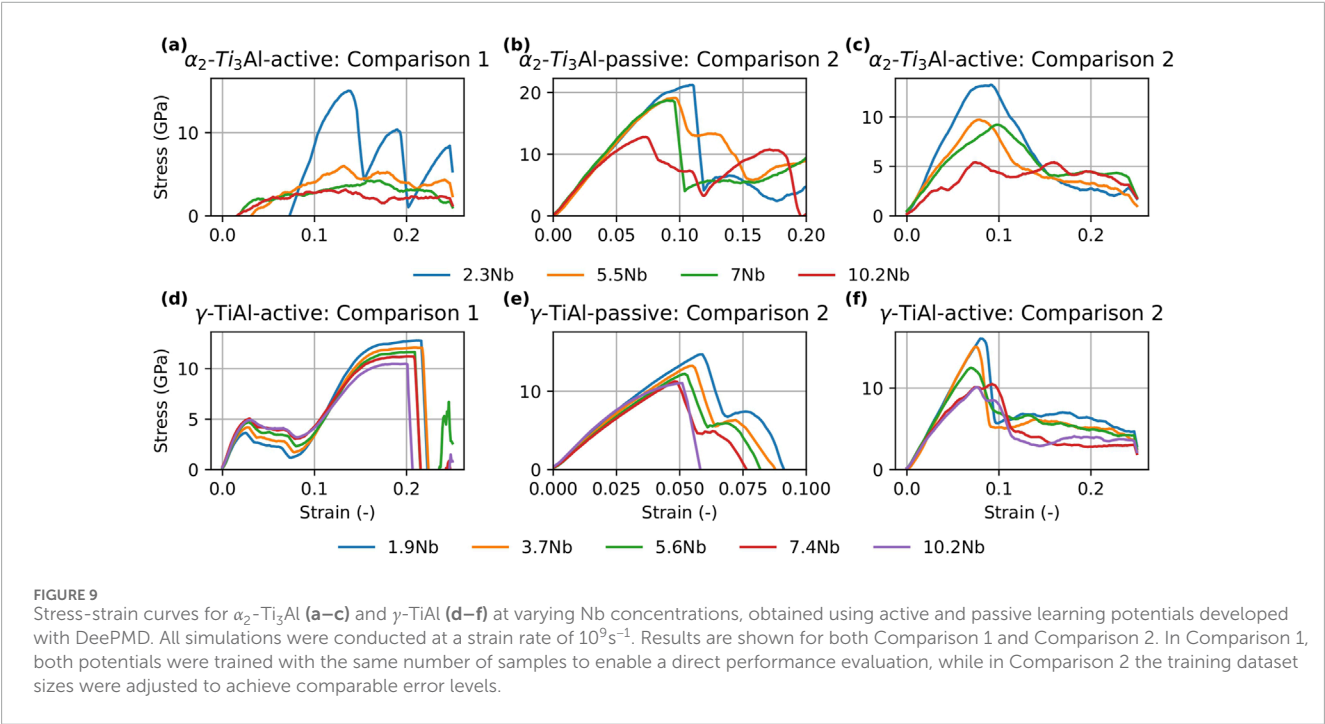


FIGURE 9 Stress-strain curves for α_2 -Ti₃Al (a–c) and γ -TiAl (d–f) at varying Nb concentrations, obtained using active and passive learning potentials developed with DeePMD. All simulations were conducted at a strain rate of 10^9s^{-1} . Results are shown for both Comparison 1 and Comparison 2. In Comparison 1, both potentials were trained with the same number of samples to enable a direct performance evaluation, while in Comparison 2 the training dataset sizes were adjusted to achieve comparable error levels.

TABLE 1 Comparison of generalized stacking fault energy values for γ -TiAl and α_2 -Ti₃Al using MTP. The figure compares two scenarios: (i) Comparison 1, where both active and passive learning potentials were trained on the same number of samples for a direct performance evaluation, and (ii) Comparison 2, where training dataset sizes were varied to achieve comparable error levels.

Phases	Fault energy	Nb conc (at.%)	Comparison 1	Comparison 2	Ref ¹	DFT ¹
γ -TiAl	APB (mJ/m^2)	0Nb	680	580	611	600
		8Nb	540	532	—	—
	CSF (mJ/m^2)	0Nb	376	343	372	356
		8Nb	229	222	—	—
α_2 -Ti ₃ Al	SISF (mJ/m^2)	0Nb	237	72	84	93
		6Nb	108	52	—	—
	APB (mJ/m^2)	0Nb	311	264	213	256
		6Nb	165	228	—	—
	CSF (mJ/m^2)	0Nb	285	346	308	320
		6Nb	73	215	—	—

¹Qi et al. (2023).

Conversely, a lower GSFE implies increased dislocation mobility and improved ductility.

Turning to the α_2 -Ti₃Al phase, which is characterized by hexagonal symmetry and multiple slip systems, our analysis primarily focused on the fundamental {0001} plane. Here we computed CSF, APB and SISF. As shown in Tables 1, 2, the GSFE values obtained from Comparison 2 for this phase also align more closely with the DFT benchmarks. Additionally, similar to γ -TiAl, an increase in Nb concentration results in a decrease in stacking fault

energies, indicating that Nb alloying enhances the ductility of the α_2 -Ti₃Al phase.

In summary, the combined results of the tension tests and GSFE calculations suggest that Nb addition generally decreases the strength of both phases while simultaneously increasing ductility. These findings are in agreement with our previous work Chandran et al. (2024a), which demonstrated that Nb alloying enhances dislocation density and ductility at the cost of reduced strength.

TABLE 2 Comparison of generalized stacking fault energy values for γ -TiAl and α_2 -Ti₃Al using DeePMD. The figure compares two scenarios: (i) Comparison 1, where both active and passive learning potentials were trained on the same number of samples for a direct performance evaluation, and (ii) Comparison 2, where training dataset sizes were varied to achieve comparable error levels.

Phases	Fault energy	Nb conc (at.%)	Comparison 1	Comparison 2	DFT ¹
γ -TiAl	APB (mJ/m ²)	0Nb	655	624	600
		8Nb	455	392	—
	CSF (mJ/m ²)	0Nb	271	365	356
		8Nb	86	243	—
α_2 -Ti ₃ Al	SISF (mJ/m ²)	0Nb	315	77	93
		6Nb	153	48	—
	APB (mJ/m ²)	0Nb	450	272	256
		6Nb	399	289	—
	CSF (mJ/m ²)	0Nb	410	307	320
		6Nb	266	254	—

¹Qi et al. (2023).

4 Conclusion

In our study, we performed a comprehensive evaluation of passive and active learning strategies for developing interatomic potentials using both MTP and DeePMD methods. The analysis was structured into two stages. In Comparison 1, both approaches were trained on the same number of samples, allowing for a direct assessment of their performance on a fixed dataset. In Comparison 2, the number of training configurations was varied to determine how many samples are required to achieve comparable RMSE values for energy/atom and force. Under Comparison 1, the active learning potential outperformed the passive learning potential. In Comparison 2, where the primary goal was to achieve comparable accuracies for passive and active learning potentials, the active learning potentials required significantly fewer samples. Specifically, for MTP, the active learning potential used only 3,322 samples for γ -TiAl and 2,602 samples for α_2 -Ti₃Al, compared to 20,000 samples for passive learning in both phases. Similarly, for DeePMD, the active learning potential was trained on 61,657 samples, whereas the passive learning approach required 127,212 samples.

The MTP approach required separate potentials for γ -TiAl and α_2 -Ti₃Al, while DeePMD enabled the training of a single potential for both phases. Both methods commenced with relatively small datasets—170 samples for MTP and 40,000 samples for DeePMD—generated solely at 300 K, which substantially reduced the need for computationally expensive AIMD simulations. In contrast, our passive learning work relied on 180 AIMD simulations of 10 ps each (Chandran et al., 2024b), whereas the active learning approach required only 32 AIMD simulations of 2.5 ps duration.

The predictive accuracy of the active learning potentials was further verified by comparing key material properties—such as elastic constants, equilibrium volume, and lattice parameters—against DFT results for both γ -TiAl and α_2 -Ti₃Al.

Although active learning generally produced predictions close to DFT, some outliers were observed, particularly for the elastic constants of the α_2 -Ti₃Al phase (in Comparison 1) and in certain equilibrium volume estimates. These discrepancies likely arise because these basic material properties were not the primary targets during the active learning sample selection, leading to a bias toward higher temperature configurations that deviate from the base state.

Finite-temperature properties, as assessed using radial distribution functions, showed good agreement with AIMD for both phases, with slightly better results for γ -TiAl. Moreover, simulated tension tests and generalized stacking fault energy calculations provided insights into the thermomechanical behavior of the alloy. Analysis of the stress–strain curves revealed a consistent decrease in UTS with increasing Nb concentration. Similarly, the GSFE analysis indicated that higher Nb content reduces stacking fault energies, which correlates with enhanced ductility. Overall, these findings confirm that Nb incorporation improves ductility at the expense of reduced strength, in agreement with our previous work Chandran et al. (2024a) and other studies Dumitraschkewitz et al. (2017).

The accuracy and reliability of the developed potentials were validated through a comprehensive procedure. Accuracy was established by achieving low RMSEs on an independent test set and by good agreement of key physical properties like equilibrium volume, elastic constants, and GSFEs with reference DFT data. The potential's reliability was confirmed by its ability to maintain thermal stability up to 900 K and to produce physically realistic stress-strain responses under large deformation. While some instabilities were observed, they were confined to the most demanding conditions: the α_2 -Ti₃Al phase with high Nb content. This demonstrated success under both thermal and mechanical extremes confirms that these potentials are accurate and reliable tools for future large-scale simulations.

Finally, we acknowledge that the scope of this study was intentionally limited to defect free, single crystal models to isolate the properties of the γ -TiAl and α_2 -Ti₃Al phases. While this approach was necessary to isolate the intrinsic effect of Nb on the thermomechanical properties of these alloys, it consequently excludes the complex influence of microstructural features like grain boundaries and phase interfaces, which are crucial to the behavior of real-world alloys.

In summary, our study demonstrates that active learning provides a more efficient and accurate approach for interatomic potential development, significantly reducing data requirements and computational cost while maintaining robust predictive performance across various material properties and thermomechanical behaviors.

Data availability statement

The original contributions presented in the study are publicly available. This data can be found here: Zenodo under the link: <https://zenodo.org/uploads/14990265>

Author contributions

AC: Conceptualization, Data curation, Investigation, Methodology, Software, Writing – original draft, Resources. AS: Validation, Writing – review and editing. CP: Validation, Writing – review and editing. PJ: Validation, Writing – review and editing. RA: Investigation, Project administration, Supervision, Writing – review and editing. Validation. CC: Funding acquisition, Investigation, Project administration, Supervision, Writing – review and editing, Validation.

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Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

Generative AI statement

The author(s) declare that Generative AI was used in the creation of this manuscript. During the preparation of this work the author(s) used ChatGpt (ChatGpt 4o-mini) in order to improve the language and readability of the text. After using this tool/service, the author(s) reviewed and edited the content as needed and take(s) full responsibility for the content of the publication.

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Supplementary material

The Supplementary Material for this article can be found online at: <https://www.frontiersin.org/articles/10.3389/fmats.2025.1591955/full#supplementary-material>

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Supplementary Material

In the supplementary materials, we provide the learning curves for active learning using MTP and DeePMD (see Section 0.1), the absolute differences in elastic constants predicted by MTP and DeePMD relative to DFT for both Comparison 1 and Comparison 2 (see Section 0.2), and RDF comparisons for cases not presented in the main text (see Section 0.3).

0.1 Active learning curves

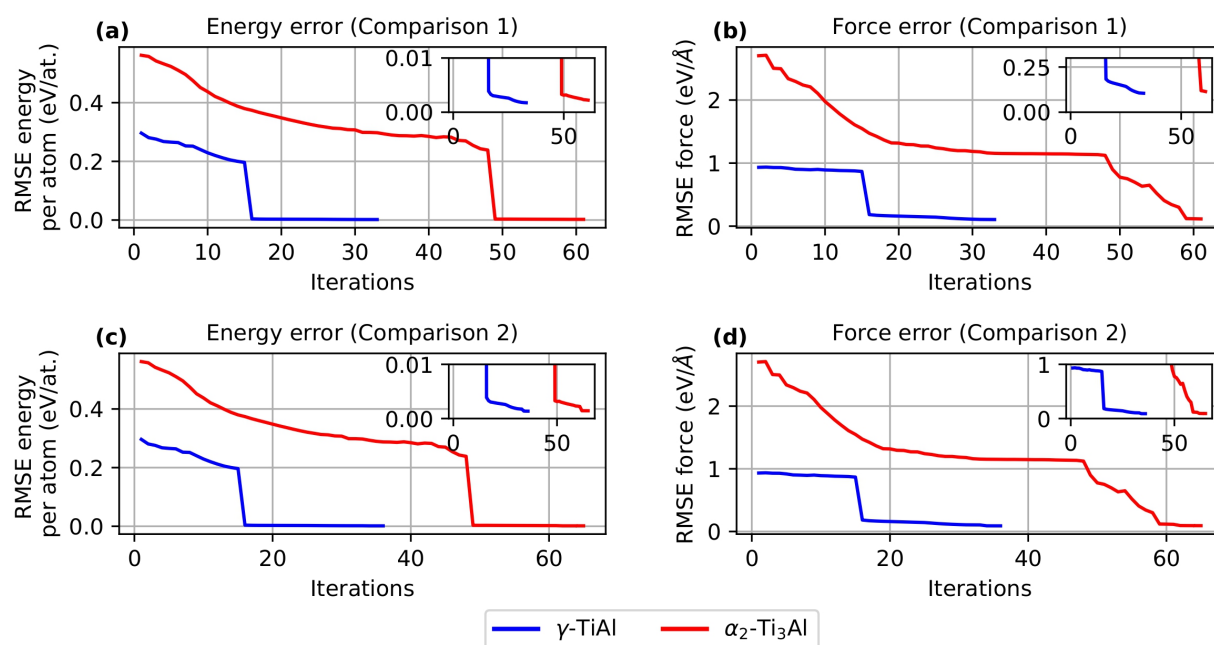


Figure S1. Learning curves for MTP showing the RMSE in energy per atom and force for γ -TiAl and α_2 -Ti₃Al. The RMSE values are plotted as a function of the active learning iterations. Panels (a) and (b) correspond to Comparison 1, where both active and passive learning potentials were trained using the same number of samples, while panels (c) and (d) correspond to Comparison 2, where the training dataset sizes were varied to achieve comparable error levels.

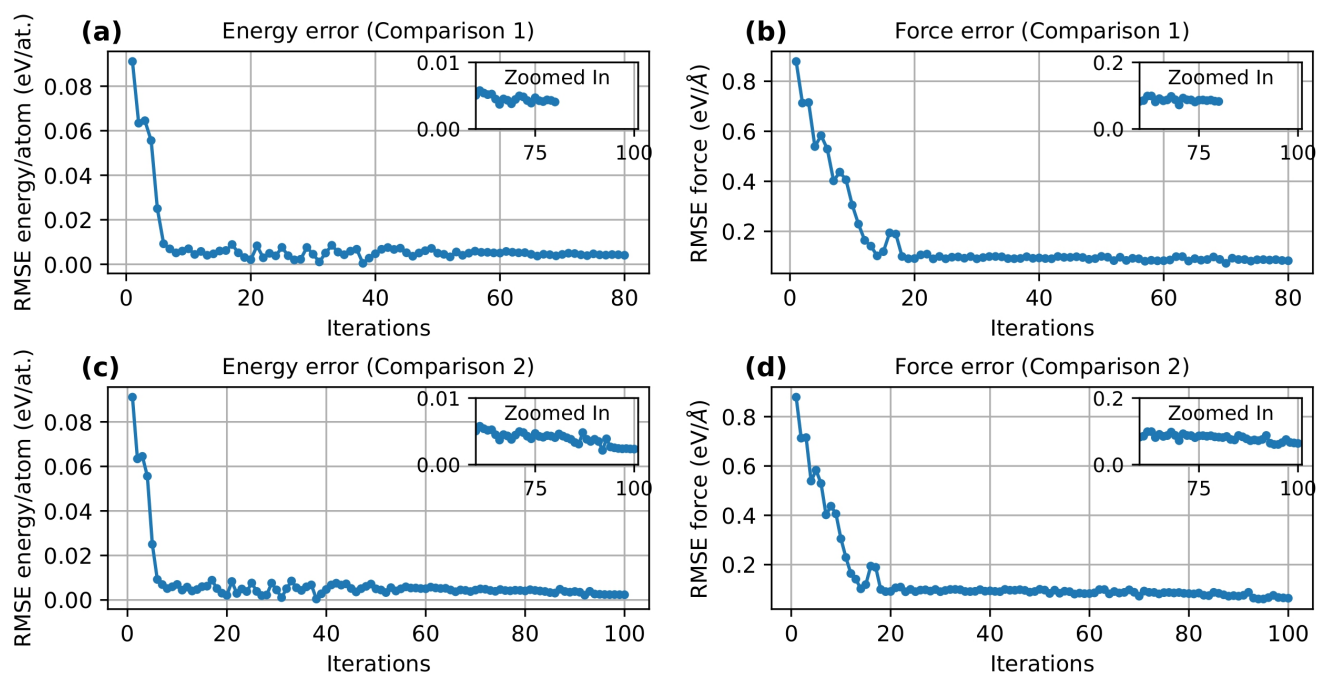


Figure S2. Learning curves for DeePMD showing the RMSE in energy per atom and force. The RMSE values are plotted as a function of the active learning iterations. Panels (a) and (b) correspond to Comparison 1, where both active and passive learning potentials were trained using the same number of samples, while panels (c) and (d) correspond to Comparison 2, where the training dataset sizes were varied to achieve comparable error levels.

Table S1. Comparison of RMSE values of energy/atom and force for MTP (using Dataset-B for γ -TiAl and Dataset-D for α_2 -Ti₃Al) and DeePMD (using Dataset-F).

Potential	Energy/atom (eV/atom)	Force (eV/Å)
MTP (γ -TiAl / α_2 -Ti ₃ Al	1.32×10^{-3} / 1.23×10^{-3}	80.14×10^{-3} / 94.44×10^{-3}
DeePMD	1.07×10^{-3}	93.14×10^{-3}

0.2 Absolute difference of elastic constants.

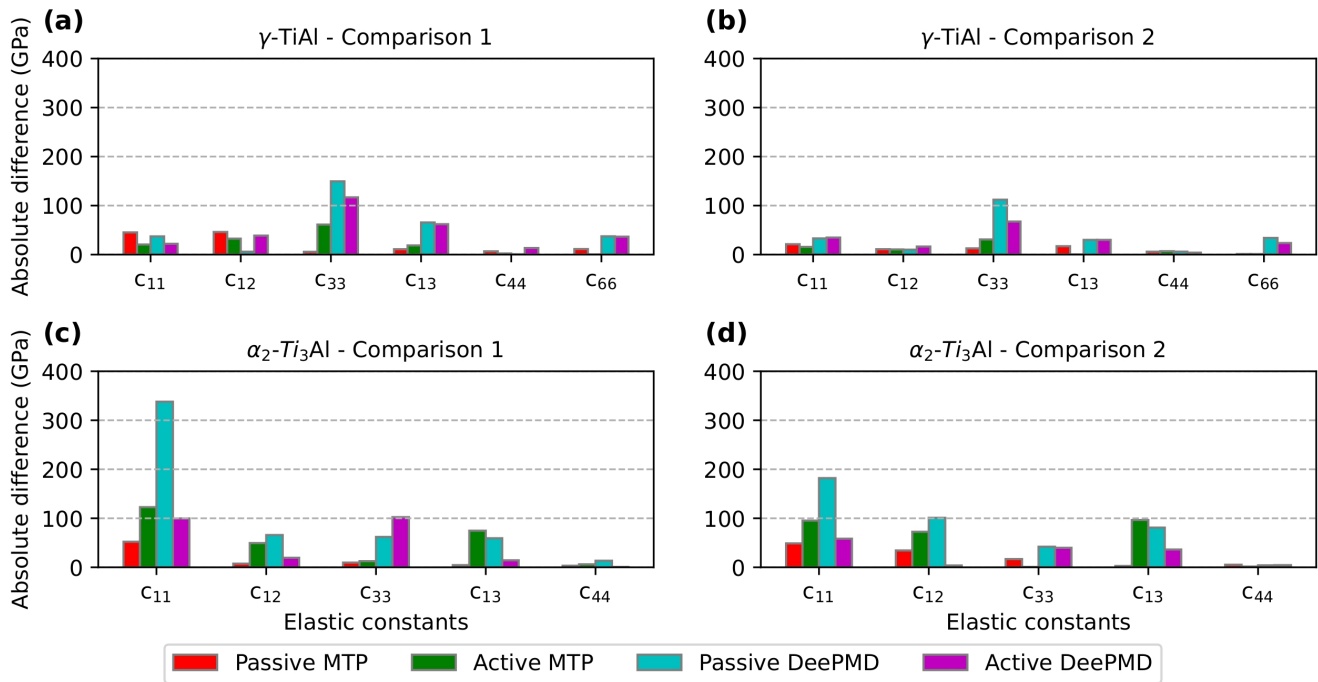


Figure S3. Absolute differences in elastic constants relative to DFT for active and passive learning potentials developed using MTP and DeePMD. Panels (a) and (b) correspond to γ -TiAl, while panels (c) and (d) correspond to α_2 -Ti₃Al. For Comparison 1, both potentials were trained using the same number of samples to enable a direct performance evaluation, whereas in Comparison 2 the training dataset sizes were varied to achieve comparable error levels. Note the unit of elastic constant is Gigapascal (GPa) and they are computed at 300 K.

0.3 Radial distribution function

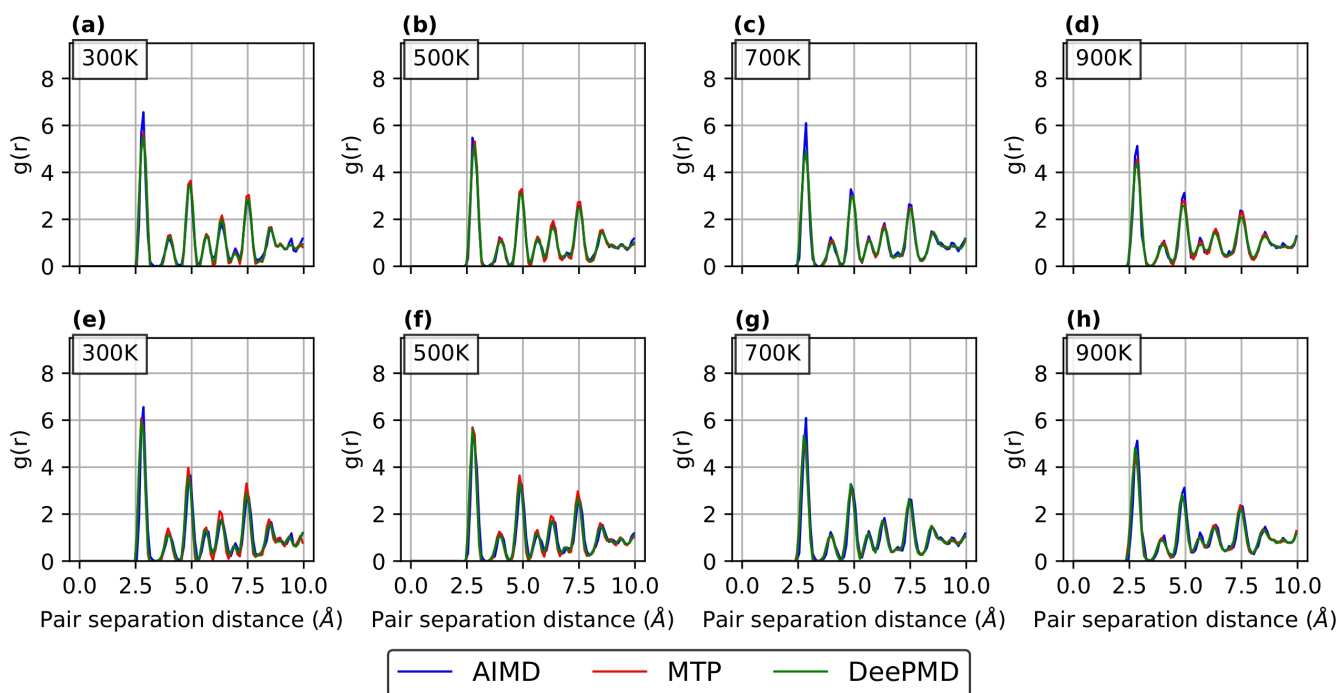


Figure S4. RDF curves for γ -TiAl with a 5.6 at% Nb configuration obtained from passive and active learning potentials, compared with AIMD results at 300 K, 500 K, 700 K, and 900 K for both MTP and DeePMD. The top row (panels a–d) displays the AIMD and active learning potential results, while the bottom row (panels e–h) shows the curves from the passive learning potential. These results correspond to Comparison 2, where the training dataset sizes were adjusted to achieve comparable error levels.

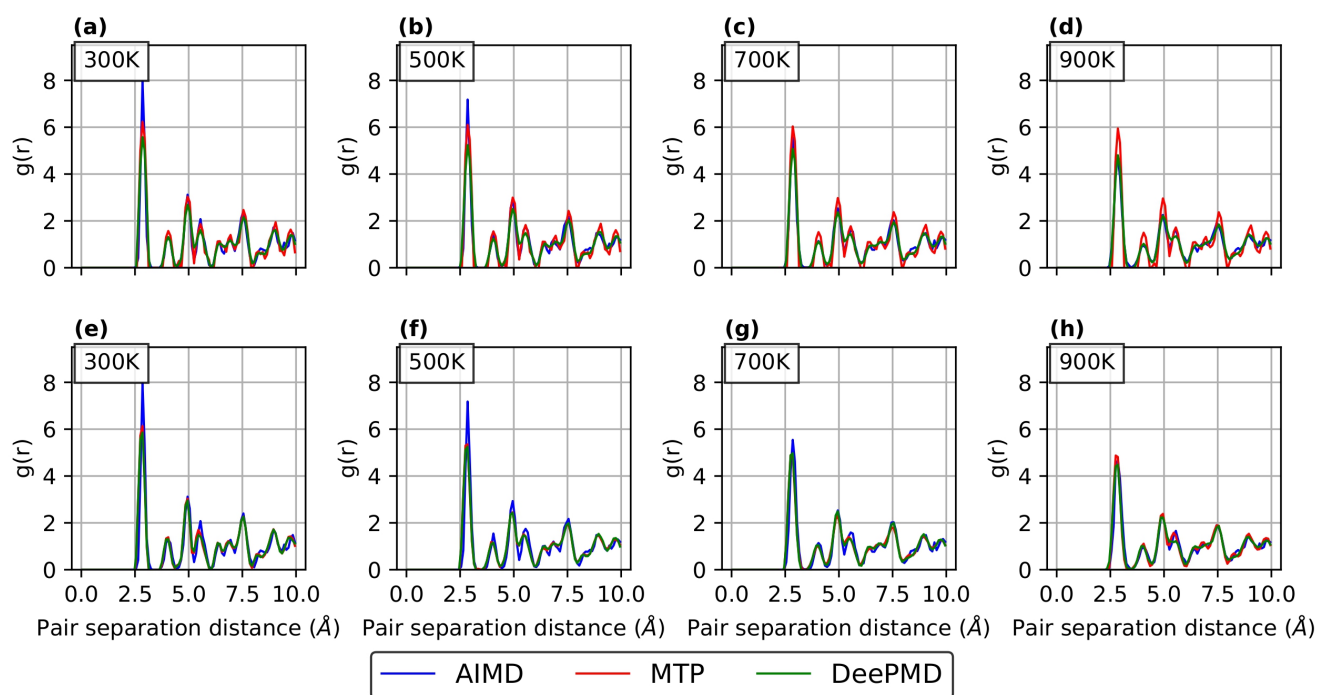


Figure S5. RDF curves for α_2 -Ti₃Al with a 2.3 at% Nb configuration obtained from passive and active learning potentials, compared with AIMD results at 300 K, 500 K, 700 K, and 900 K for both MTP and DeePMD. The top row (panels a–d) displays the AIMD and active learning potential results, while the bottom row (panels e–h) shows the curves from the passive learning potential. These results correspond to Comparison 2, where the training dataset sizes were adjusted to achieve comparable error levels.