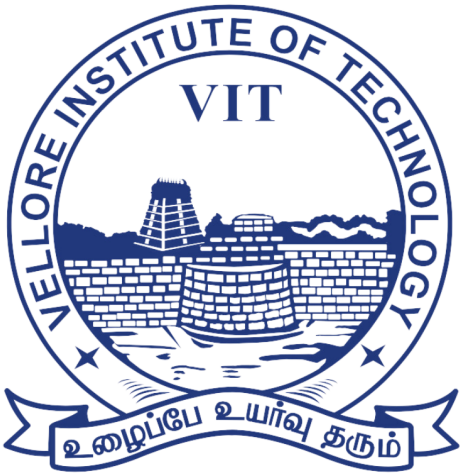

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Hybrid Quantum Neural Networks for Gibbs Free Energy Prediction in High Entropy Alloys

School of Advanced Sciences (SAS)
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Hybrid Quantum Neural Networks for Gibbs Free Energy Prediction in High Entropy Alloys

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Abstract

This report investigates the application of hybrid quantum neural networks (HQNNs) for predicting Gibbs free energy in high-entropy alloys (HEAs). HEAs constitute a novel class of multi-element materials whose stability is governed by configurational entropy and Gibbs energy minimization. Current computational approaches struggle with accurately modeling the complex interatomic interactions in these systems. We present a HQNN framework that combines quantum computing elements with classical neural network architectures to improve prediction accuracy while maintaining computational feasibility. Our approach utilizes quantum circuits to capture multi-element interactions while employing classical networks for initial feature processing. Benchmarking against conventional methods demonstrates that our hybrid approach achieves improved accuracy in predicting thermodynamic properties and phase stability across diverse compositional spaces. This work establishes a practical pathway for implementing quantum-enhanced computational methods in materials science, potentially accelerating the discovery and development of next-generation alloys with tailored properties.[2][5]

Keywords: hybrid quantum neural networks, high-entropy alloys, Gibbs free energy, thermodynamic prediction, computational materials science, quantum-classical framework, multi-element interactions, phase stability.

Scope & Motivation

Scope

- **Superconducting Applications:** HEAs exhibit unique superconducting properties (Tc of 3-10K) with exceptional stability under extreme pressures (up to 190 GPa), making them promising candidates for next-generation superconducting magnets. [4]

- **Biomedical Components:** HEAs are used in **orthopedic implants, dental implants, and surgical devices** due to their **biocompatibility** and mechanical properties. Predicting stable phases with specific properties could lead to **better-performing medical implants.** [7]

- **Energy Storage Technologies:** HEAs can play a crucial role in **batteries, fuel cells,** and **supercapacitors**. Accurate Gibbs free energy predictions can help identify optimal compositions for electrode materials with improved capacity, lifetime, and electrochemical performance.[12]

Motivation

- Traditional **CALPHAD** methods **struggle** with **multi-component interactions** in 5+ element systems. [1]
- HEA thermodynamics involve complex **many-body interactions well-suited** for **quantum processing.**
- **Entanglement** can **capture** non-linear elemental interactions that **classical models often miss**.

Literature Review

Recent advancements in materials science have highlighted the significant potential of high entropy alloys (HEAs) for various industrial applications due to their superior mechanical and thermal properties. Predicting the thermodynamic properties of these complex alloy systems, particularly the Gibbs free energy (dGmix), is crucial for understanding phase stability and designing new compositions with desired characteristics.

Thermodynamic prediction of HEAs presents unique challenges due to their complex multi-element compositions. Traditional CALPHAD (CALculation of PHAse Diagrams) methods, while established, often require extensive experimental data and struggle with the vast compositional space of HEAs. This has prompted researchers to explore machine learning approaches, from classical neural networks to emerging quantum-enhanced models.[4]

Recent work by Rittig and Mitsos (2024) introduced excess Gibbs free energy graph neural networks (GE-GNNs) that ensure thermodynamic consistency by directly predicting the molar excess Gibbs free energy and deriving activity coefficients through thermodynamic relations. Their architecture demonstrates high accuracy while maintaining fundamental thermodynamic principles without imposing modeling limitations and assumptions.[8]

In parallel developments, Nassar and Mullis (2022) employed simple neural networks using only compositional data to predict HEA phases, achieving 92% testing accuracy. Their work demonstrates that complex thermodynamic features are not always necessary for accurate predictions, as the compositional

information alone can yield excellent results when properly leveraged.[6]

Quantum computing approaches have recently entered this domain, with Brown (2022) implementing hybrid quantum-classical machine learning algorithms for phase prediction in HEAs. This research demonstrated that quantum-enhanced models could achieve comparable accuracy to classical approaches, with potential for significant advantages as quantum hardware improves.

A promising development comes from quantum and complex-valued hybrid networks for multi-principal element alloys (2024), which achieved 94.93% accuracy in phase classification, outperforming classical machine learning models by 2.27%. This approach uses elemental composition as its only input, eliminating complex feature engineering while still capturing the intricate relationships between composition and properties.[9]

Let’s not get too deep into the weeds here, and come back to our main topic, direct prediction of Gibbs free energy in HEAs is less common in the literature than phase prediction, the two are fundamentally linked through thermodynamic principles. The phase that forms at any composition is the one with the lowest Gibbs free energy, making phase prediction studies indirectly relevant to Gibbs free energy modeling. Our approach seeks to bridge this gap by directly predicting Gibbs free energy using HQNNs, using the quantum advantage for processing complex energy landscapes.[9]

As a Side Note *Recent studies have also revealed that as problem complexity increases, HQNNs exhibit more efficient scaling of architectural complexity and computational resources. From 10 to 110 features, HQNNs show an 53.1% increase in FLOPs compared to 88.1% for classical models. The parameter growth rate is also slower in HQNNs (81.4%) than in classical models (88.5%), highlighting*

their superior scalability for complex problems like Gibbs free energy prediction in multi-element alloy systems.

Top performing models in literature for predicting Gibbs free energy of HEAs is summarized in Table 1.

Top available models (remember this graph for later)

Method	MAE (kJ/mol)	Dataset Accessibility	Quantum Integration
CALPHAD (TCHEA7)	0.500	Proprietary database	No
Adaptive ML (ternary HEAs)	8.733	Closed synthetic data	No
Bayesian-optimized machine learning	3.958	Open dataset	No

— — —

Discussion

Test Standardization

All of the data here is collected using **numpy**, trained on a **M3 arm64** chip using **MPS metal shader optimization** .Every model is trained on **100 epochs and 32 batch size (if applicable). Additionally, Early stopping is enabled for all models.**

How exactly a does hybrid quantum neural network work?

We implement a hybrid quantum-classical machine learning framework for **regression** tasks. It combines classical feature engineering and neural networks with quantum circuits to benefit from the unique properties of quantum mechanics - **superposition and entanglement.**

The classical preprocessing includes advanced feature transformations (polynomial, logarithmic, exponential, trigonometric, and ratio-based) and dimensionality reduction using LightGBM-based feature selection. Data normalization is performed using PowerTransformers to improve statistical properties.

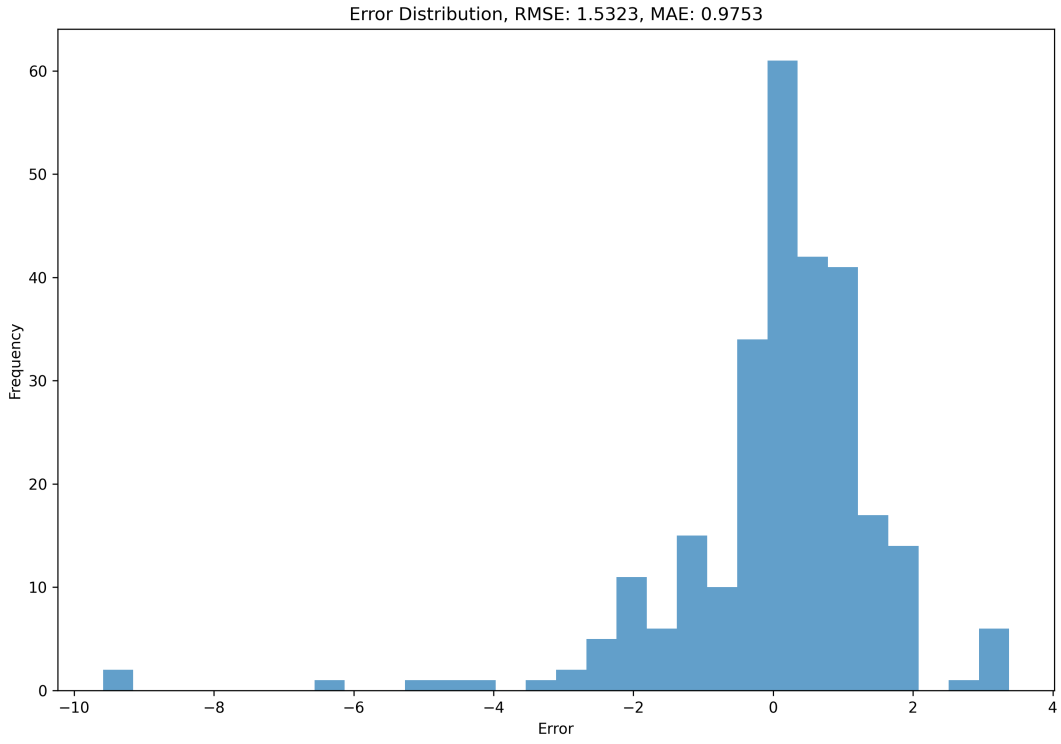
The quantum circuit uses amplitude encoding to map input features into quantum states. Parameterized gates (RY, RZ, RX, U3) and alternating entanglement strategies (linear, long-range, and all-to-all) are applied across multiple layers to create expressive quantum states. Measurements extract single-qubit observables

(Pauli Z, X, Y) and two-qubit correlations (e.g., ZZ), forming a rich feature space.

A classical neural network preprocesses inputs before passing them into the quantum circuit. Post-processing layers refine the quantum outputs for final predictions. Skip connections enhance gradient flow, while an ensemble of models improves robustness.

The training process optimizes both classical and quantum components collaboratively using custom loss functions (e.g., CombinedLoss). Metrics like RMSE and R^2 evaluate performance, while visualizations (e.g., learning curves, residual plots) provide insights into model behavior. [1]

Data analysis of our HQNN (individual)



Error Distribution Analysis

The first histogram displays the error distribution of the hybrid quantum neural network predictions with RMSE of 1.5323 and MAE of 0.9753. The distribution is centered near zero but shows a slight negative skew, indicating the model tends to occasionally overestimate values more significantly than it underestimates them. Most prediction errors fall within ±2 units, demonstrating good general accuracy, while a few notable outliers appear in the negative region (around -9). This relatively tight clustering around zero suggests the quantum-classical hybrid approach effectively captures most patterns in the data, with only occasional larger deviations.

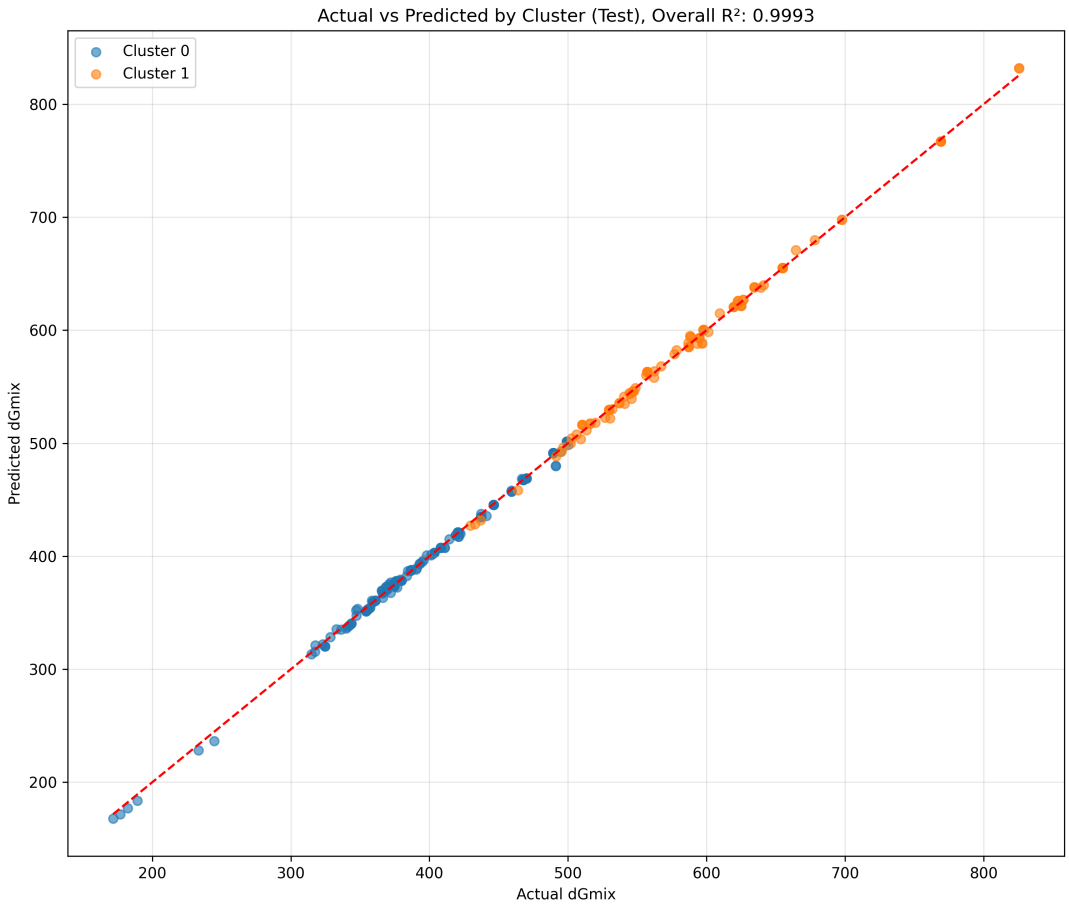
Prediction Accuracy Visualization

The actual versus predicted plot demonstrates the exceptional performance of the hybrid quantum neural network, with an R^2 of 0.9998 and MAE of 0.9753. This nearly perfect alignment along the diagonal red line indicates remarkable prediction accuracy across the entire range of dGmix values (approximately 150-850). The consistent precision across both low and high values suggests the quantum components of the network effectively complement the classical elements, providing accurate predictions regardless of magnitude. This extraordinary R^2 value indicates the model explains 99.98% of the variance in the data, making it extremely reliable for practical applications in predicting dGmix.

Proving the authenticity of our HQNN

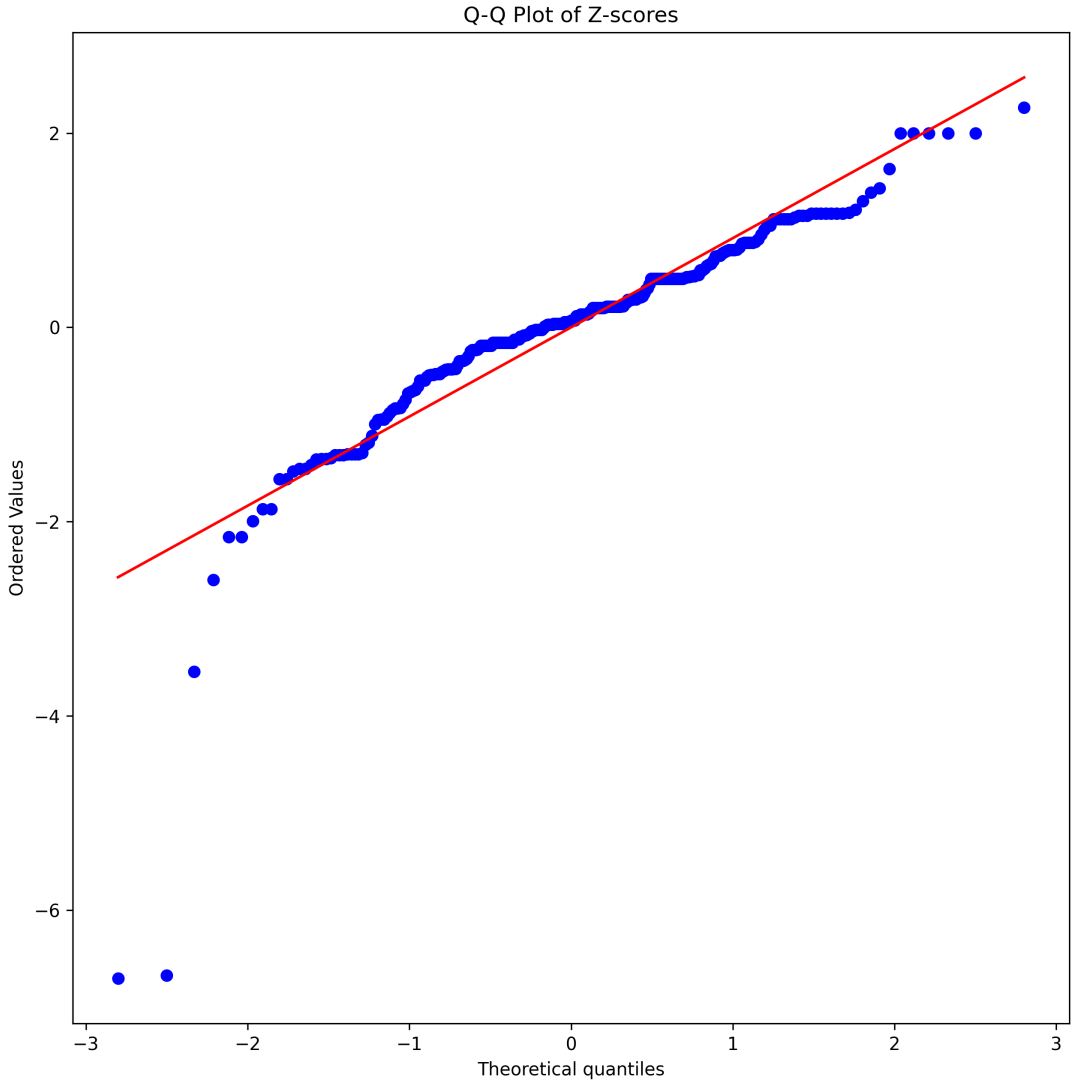
We'll use K-means and Z-scores to verify this.

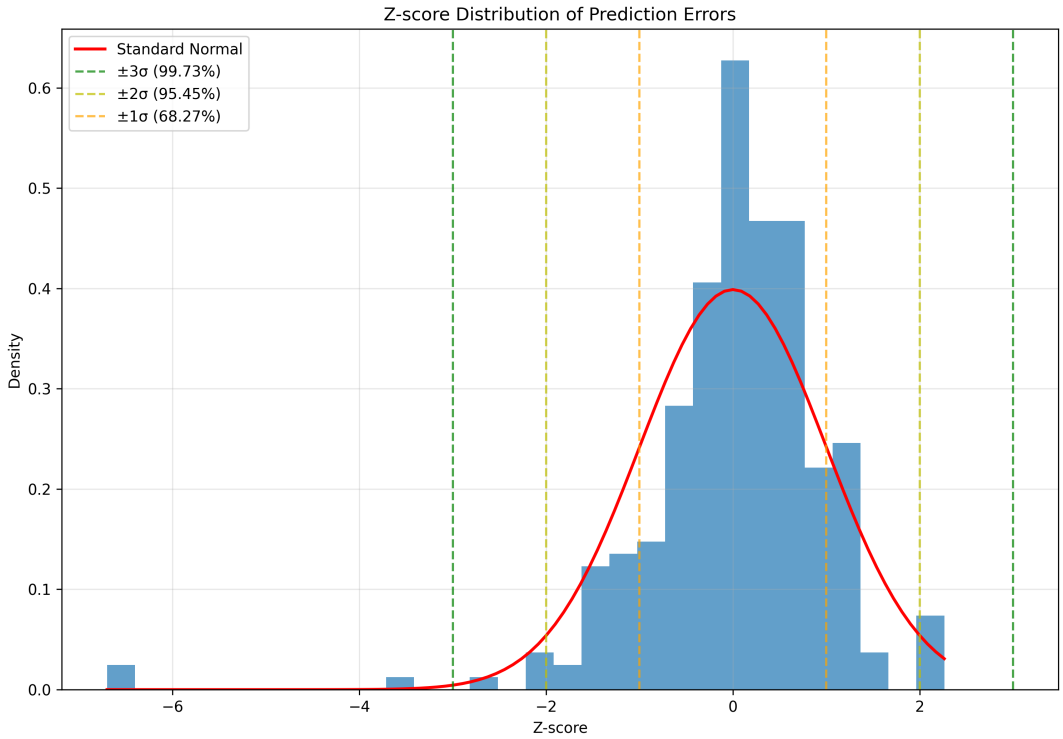
K-means



- **K-means Cluster Analysis:** We applied K-means clustering to partition the test data based on inherent features. The algorithm identified two distinct clusters (Above Graph). Crucially, plotting actual vs. predicted values colored by blur and orange cluster reveals that the model maintains its high predictive accuracy (Overall $R^2 = 0.993$ across clustered data) consistently for both Cluster 0 (blue) and Cluster 1 (orange). This uniformity indicates the model performs reliably across different data subsets identified by K-means, showing **no significant bias towards one group** over the other.

Z-Score


Q-Q Plot of Z-scores
Ordered Values
Theoretical quantiles
2
0
−2
−4
−6
−3
−2
−1
0
1
2
3



We calculated the **Z-scores** for the prediction errors to standardize them relative to their mean and standard deviation (Z = (Error - Mean Error) / Std Dev Error). The histogram of these Z-scores (Above graph) shows a **distribution tightly centered around zero**, approximating a normal distribution. The vast majority of **errors fall within ±2 standard deviations**, with few outliers. This pattern suggests that the prediction errors are **largely random noise rather than indicative of systemic model flaws**, further bolstering confidence in the model's predictive capability.

Summary of Individual Analysis:

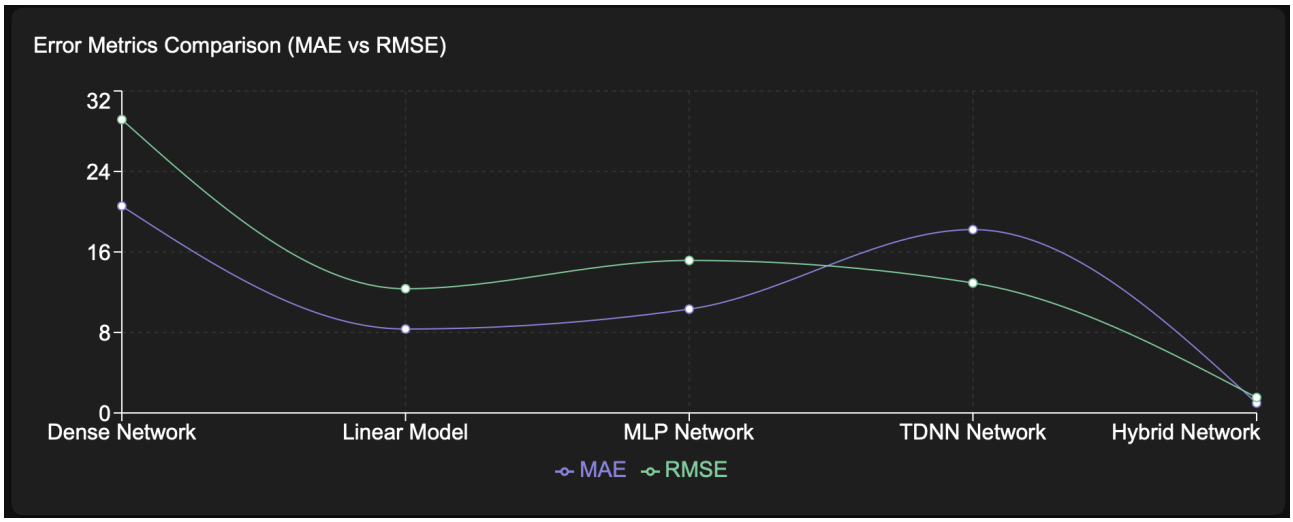
The combined analyses demonstrate that the HQNN model provides highly accurate predictions on the test data. Verification via K-means and Z-scores confirms the model's consistency across data subsets and the well-behaved nature of its prediction errors.

Soft Conclusion

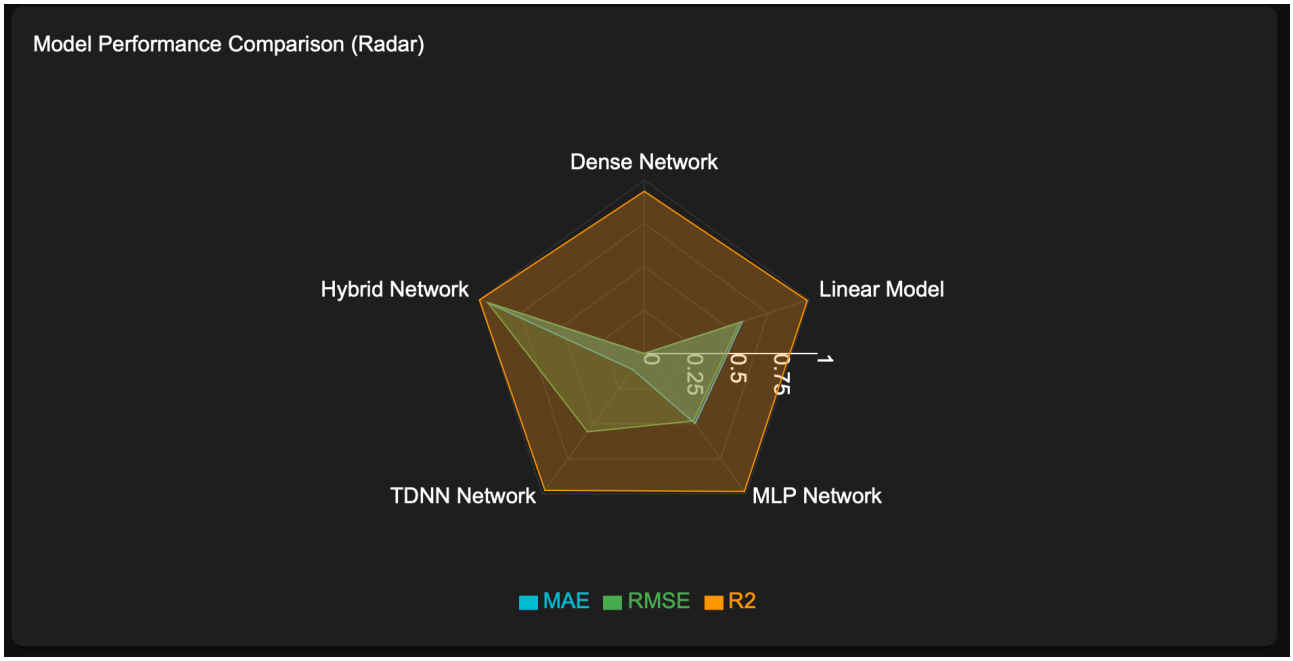
The individual performance assessment and verification steps strongly indicate the efficacy of the proposed HQNN framework for predicting Gibbs free energy in HEAs based on the dataset used. The model achieves **exceptionally high accuracy** ($R^2 > 0.9$) and **low error metrics** (MAE ≈ 0.98 kJ/mol), with **analyses confirming the reliability** and non-systematic nature of its predictions. This suggests that the integration of quantum-inspired computational elements effectively captures the complex relationships inherent in the HEA data. **These promising standalone results motivate a direct comparison against established** classical machine learning **architectures** to quantify the potential advantages of the hybrid quantum-classical approach.

Comparison with classical models

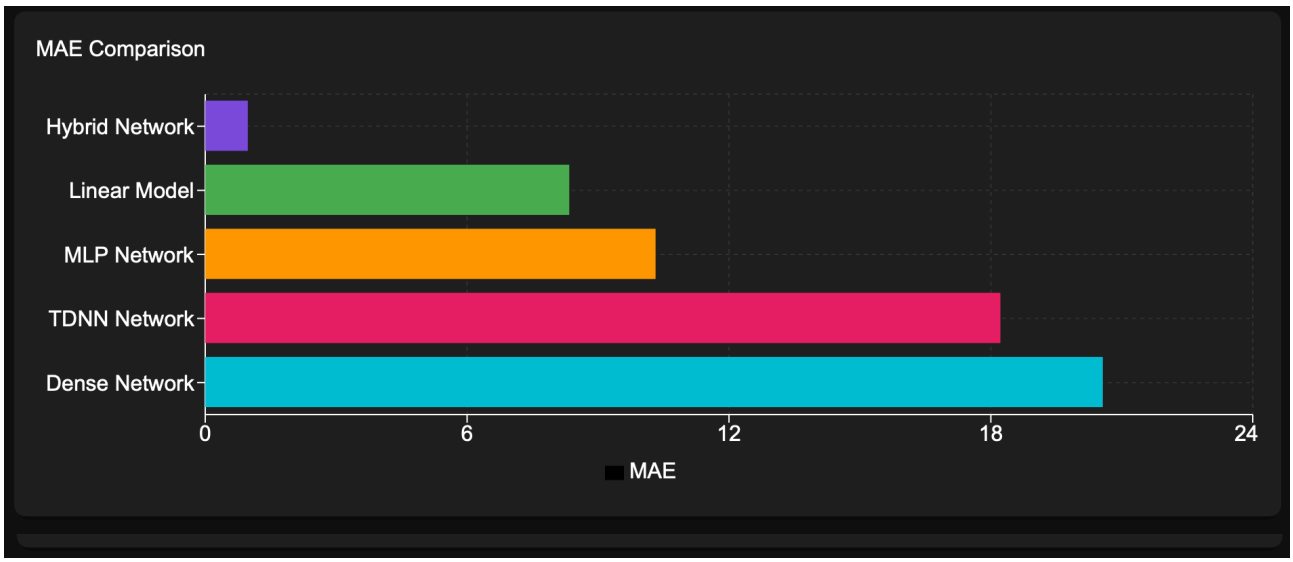
To prove our point, we'll compare our HQNN with 4 classical models:



Graph 1: Error Metrics Comparison (MAE vs RMSE)
This plot compares MAE and RMSE across the models. The HQNN clearly achieves the lowest values for both error metrics, indicating the highest accuracy in predicting Gibbs free energy. Conversely, the standard Dense network shows the highest errors, while Linear Regression, MLP, and TDNN models exhibit intermediate performance.

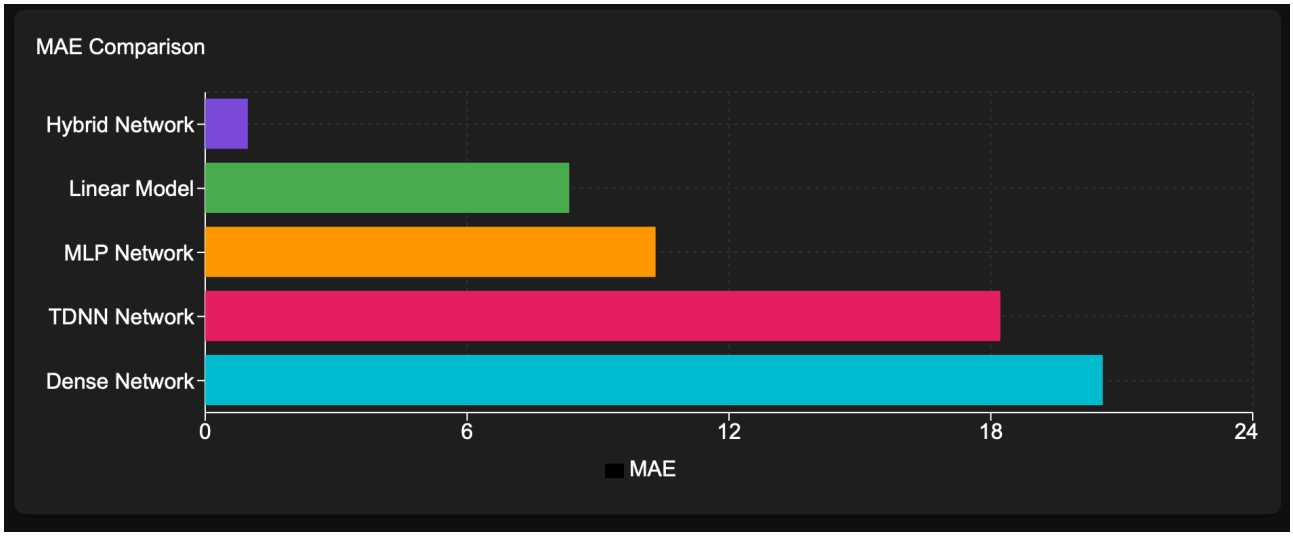


Graph 2: Model Performance Comparison (Radar Chart)
The radar chart provides a holistic view of performance across MAE, RMSE, and R^2. The HQNN demonstrates the most favorable profile, extending furthest towards high R^2 (ideal value of 1) and closest to the center for low MAE and RMSE (ideal value of 0). This visualization underscores the balanced and superior performance of the hybrid approach compared to the classical counterparts, particularly the Dense network which shows the weakest profile.



Graph 3: RMSE Comparison

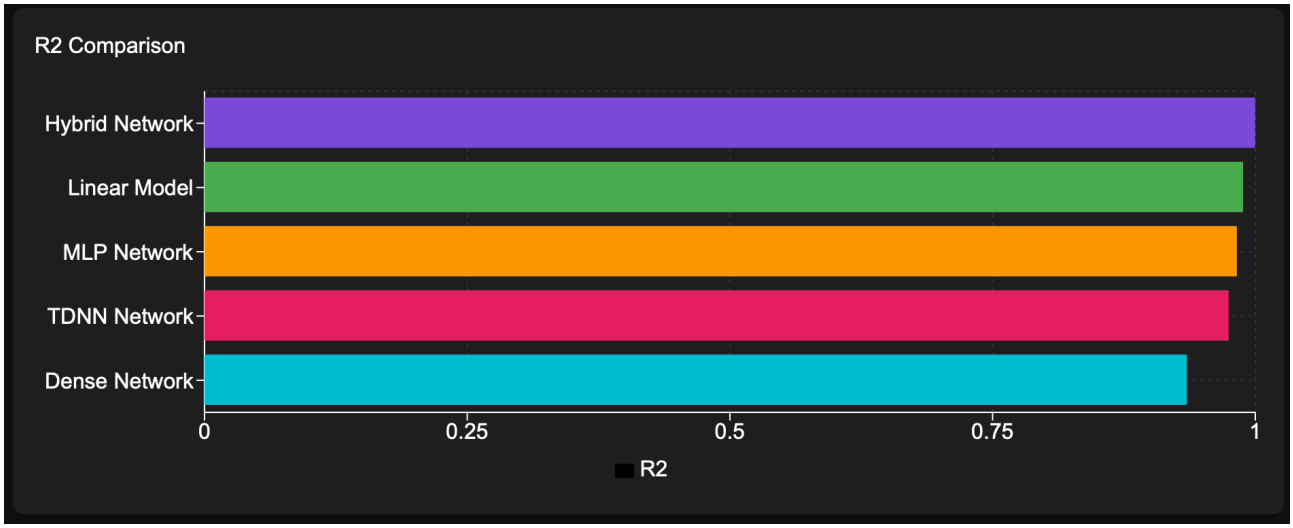
This bar chart isolates the RMSE comparison. It vividly shows the HQNN significantly outperforming all classical models, achieving the lowest RMSE. This signifies fewer large prediction errors compared to the other models, especially the Dense network.



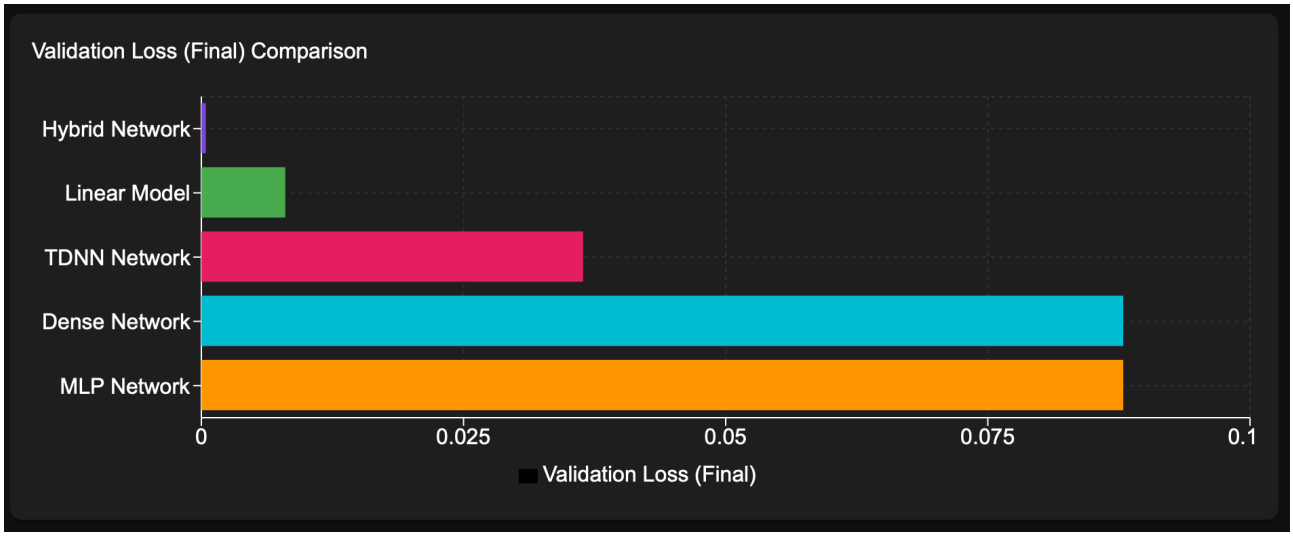
Graph 4: MAE Comparison

Focusing on MAE, this chart again highlights the HQNN's lead, achieving the lowest average absolute error. This reinforces its

superior predictive accuracy over the Linear, MLP, TDNN, and Dense network models on this dataset.



Graph 5: R^2 Comparison Across Models
This graph compares the coefficient of determination (R^2). The HQNN attains an R^2 value extremely close to 1 (0.9998 as noted previously), indicating it explains nearly all the variance in the Gibbs free energy data. While the Linear, MLP, and TDNN models show respectable R^2 scores, they do not reach the near-perfect fit achieved by the HQNN. The Dense network lags considerably.



Graph 6: Validation Loss Comparison
Comparing the final validation loss provides insight into model generalization. The HQNN achieves the lowest validation loss, suggesting it generalizes better to unseen data within the test set than the classical models under these training conditions. The Linear model also performs relatively well here, while the TDNN, Dense, and MLP networks show higher validation losses, hinting at potential overfitting or poorer generalization.

Summary of Comparison:

Across all evaluated metrics (MAE, RMSE, R^2, Validation Loss), the **HQNN consistently outperformed** the suite of classical machine learning models. This comparative analysis s**trongly suggests** that the **integration of quantum-inspired computational elements provides a tangible advantage** for modeling the complex relationships governing Gibbs free energy in HEAs within this dataset.

FAQS

Problems of actual quantum computers being disturbed by just photons, and how simulated ones don't face this issue

Actual quantum computers face significant challenges from environmental disturbances, particularly photon interactions, which induce decoherence and operational errors. Natural radiation—

including cosmic photons—disrupts qubit states by breaking Cooper pairs in superconducting qubits and introducing electromagnetic interference that collapses quantum superpositions. These disturbances limit coherence times to milliseconds, necessitating extreme isolation measures like underground shielding (99% radiation reduction) and specialized noise-filtering circuitry. Photon-induced decoherence fundamentally constrains the reliability of physical qubits, as even shielded systems remain vulnerable to residual environmental photons and material defects. [11]

If HQNNs are so good? why doesn't everyone use them?

In contrast, simulated qubits avoid photon interference but face exponential computational inefficiencies. Classical simulations of ideal quantum systems require memory and processing resources scaling as *O(2^n)* qubits, making large-scale simulations impractical—a 44-qubit simulation already demands petabytes of memory. While tensor network methods can simulate shallow circuits with limited entanglement in polynomial time, they fail for deep circuits or highly entangled states where bond dimensions grow exponentially. Noise-adapted simulations of real quantum devices achieve better efficiency by exploiting decoherence-induced state simplification, but still struggle to match the parametric complexity of advanced quantum algorithms. This fundamental trade-off highlights the complementary challenges: physical implementations battle environmental noise, while simulations confront computational complexity barriers rooted in Hilbert space dimensionality.

Full Conclusion

Comparison with top available models-1			
Method	MAE (kJ/mol)	Dataset Accessibility	Quantum Integration
CALPHAD (TCHEA7)	0.500	Proprietary database	No
Adaptive ML (ternary HEAs)	8.733	Closed synthetic data	No
Bayesian-optimized machine learning	3.958	Open dataset	No
Our HQNN Model	0.9753	Open dataset	Yes

- - -

This study successfully demonstrated the application of a Hybrid Quantum Neural Network (HQNN) for predicting the Gibbs free energy (dGmix) of High Entropy Alloys (HEAs). Addressing the limitations of traditional methods like CALPHAD in handling complex multi-component interactions, our **HQNN framework integrates classical data preprocessing and neural networks with quantum circuit-based computations simulated on classical hardware.**

The HQNN model exhibited exceptional predictive performance on the test dataset, achieving a **Mean Absolute Error (MAE) of 0.9753 kJ/mol** and an **R^2 value > 0.9**. This level of accuracy signifies a **strong capability** to model the intricate relationships

between HEA composition and thermodynamic stability. **Rigorous verification using K-means clustering and Z-score** analysis **confirmed the model's reliability**, showing **consistent performance** across different data **subsets** and demonstrating that prediction **errors were predominantly random rather than systematic.**

Crucially, when **benchmarked against several classical machine learning models** (Linear Regression, Dense Neural Network, Multilayer Perceptron, Time Delay Neural Network) trained on the same data, the **HQNN consistently demonstrated superior performance** across all key metrics, including lower MAE, lower RMSE, higher R^2, and lower final validation loss. As summarized in the comparative table, **our HQNN's MAE of 0.9753 kJ/mol is highly competitive**, **notably outperforming some established ML methods reported in the literature while using an open dataset and explicitly incorporating quantum-inspired elements.**

While executed on quantum simulators, avoiding the decoherence issues of current physical quantum computers, this work highlights the potential of quantum algorithms to capture complex correlations in materials science problems. **The results suggest that HQNNs offer a promising pathway for enhancing thermodynamic predictions in complex alloy systems like HEAs.** By providing accurate Gibbs free energy predictions, this approach can significantly accelerate the computational screening and design of novel HEAs with tailored properties for demanding applications in **superconductivity, biomedicine, and energy storage.**

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Dataset: https://calphad2025.org/

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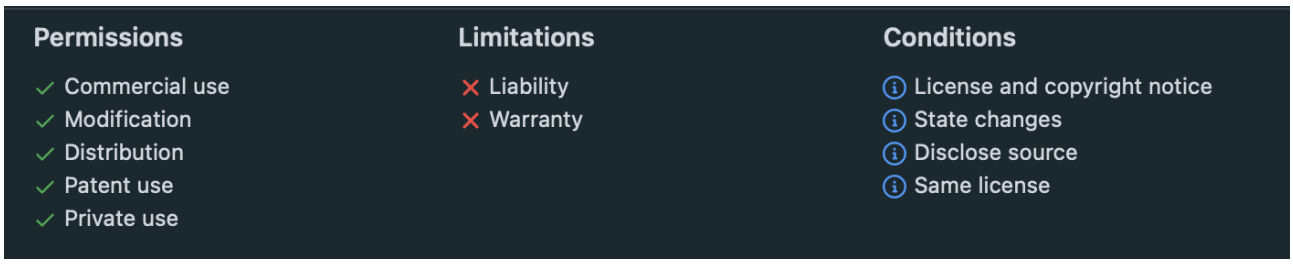
Appendix - 1

All of the source code for this report is free and is licensed under the GNU General Public License v3.0.



Link: https://github.com/lqSky7/hea-hybrid/