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Materials Property Prediction with Graph Transformers and Uncertainty-Calibrated Active Learning

Hao Chen,[1] Wei Zhang[1]

[1]School of Computer and Communication Sciences, EPFL, Lausanne 1015, Switzerland

Abstract

The acceleration of materials discovery relies heavily on the ability to predict physicochemical properties of crystal structures with high accuracy and computational efficiency. While Density Functional Theory (DFT) provides precise ground-truth data, its cubic scaling with electron count renders it prohibitive for high-throughput screening of vast chemical spaces. Consequently, machine learning surrogates have emerged as a critical alternative. However, conventional Message Passing Neural Networks (MPNNs) often struggle to capture long-range atomic interactions and suffer from over-smoothing in deep architectures. Furthermore, standard active learning frameworks frequently rely on uncalibrated uncertainty estimates, leading to suboptimal sampling strategies and wasted computational resources. This paper introduces a novel framework: the Graph Transformer with Uncertainty-Calibrated Active Learning (GT-UCAL). We propose a geometric graph transformer architecture that integrates structural positional encodings to capture global topology, coupled with an evidential deep learning mechanism to quantify both aleatoric and epistemic uncertainties. Through rigorous experimentation on the Materials Project and JARVIS datasets, we demonstrate that GT-UCAL achieves state-of-the-art predictive performance while reducing the requisite labeled data by approximately 40% compared to random sampling.

Keywords

Graph Neural Networks, Transformer Architecture, Active Learning, Uncertainty Quantification, Materials Informatics

Introduction

1.1 Background

The paradigm of materials science has shifted dramatically over the past decade, moving from Edisonian trial-and-error methodologies toward data-driven inverse design. The central challenge in this domain is the structure-property mapping problem: given a crystallographic arrangement of atoms, predict properties such as formation energy, bandgap, or elastic moduli [1]. Historically, this mapping has been performed using ab initio simulation methods, most notably Density Functional Theory (DFT). While DFT offers high fidelity, it is computationally expensive, often requiring thousands of CPU hours for a single unit cell calculation [2].

To circumvent these computational bottlenecks, the field has increasingly adopted machine learning (ML) surrogates. These models, trained on databases of pre-calculated DFT results, can predict properties in milliseconds. Early approaches utilized hand-crafted descriptors, such as Coulomb matrices or radial distribution functions, to vectorize crystal structures [3]. However, the advent of Deep Learning, specifically Graph Neural Networks (GNNs),

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revolutionized this space by allowing models to learn representations directly from the molecular graph structure, where atoms serve as nodes and chemical bonds as edges [4].

1.2 Problem Statement

Despite the success of GNNs, significant limitations persist. Standard message-passing architectures rely on iterative aggregation of local neighborhoods. This locality bias makes it difficult for the network to model long-range interactions, such as electrostatic forces or steric hindrance in complex macromolecules, unless the network is made exceedingly deep [5]. However, increasing the depth of GNNs frequently leads to the over-smoothing problem, where node representations become indistinguishable [6].

Moreover, the training of these surrogates assumes the availability of large, labeled datasets. In materials science, labels are expensive (DFT calculations). Active Learning (AL) addresses this by iteratively selecting the most informative structures to label. However, the efficacy of AL depends entirely on the model's ability to estimate its own uncertainty [7]. Standard neural networks are notoriously overconfident, assigning high certainty even to erroneous predictions on out-of-distribution data. Without calibrated uncertainty, the AL loop may select redundant samples or fail to explore novel regions of the chemical space [8].

1.3 Contributions

In this work, we propose GT-UCAL, a unified framework addressing both the representational limitations of standard GNNs and the calibration deficits of traditional active learning. Our contributions are threefold:

First, we design a Geometric Graph Transformer that replaces static message passing with a dynamic self-attention mechanism. This allows every atom to attend to every other atom, modulated by distance and bond characteristics, effectively capturing long-range dependencies without deep stacking [9].

Second, we integrate a scalable uncertainty quantification method based on Evidential Deep Learning. Unlike Bayesian Neural Networks or Ensembles, which are computationally heavy, our approach places a distribution over the output parameters, allowing a single forward pass to estimate both data noise (aleatoric) and model ignorance (epistemic) [10].

Third, we demonstrate a calibration-aware acquisition function that balances exploration and exploitation more effectively than standard variance-based sampling. We validate this on the Materials Project database, showing significant improvements in data efficiency [11].

Chapter 2: Related Work

2.1 Classical Approaches and Descriptor-Based Models

The earliest attempts to apply machine learning to materials science relied heavily on domain-expert knowledge to construct feature vectors. Descriptors such as the Voronoi tessellation features and the Sine Matrix were fed into classical algorithms like Random Forests and Support Vector Machines (SVMs) [12]. While these models offered interpretability and low training costs, they were constrained by the quality of the descriptors. Hand-crafted features often failed to capture the nuances of rotational invariance or the periodicity of crystal lattices, leading to poor generalization on complex, multi-element compounds [13].

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2.2 Deep Learning and Graph Neural Networks

The introduction of Crystal Graph Convolutional Neural Networks (CGCNN) marked a turning point. CGCNN formalized crystals as multigraphs, enabling end-to-end learning of atomic features [14]. Subsequent architectures like SchNet and MEGNet introduced continuous-filter convolutions and global state attributes to better model physical interactions [15].

Despite these advancements, standard GNNs are fundamentally limited by the k-hop neighborhood aggregation scheme. To model an interaction between two atoms 10 angstroms apart, a standard GNN might require 5 to 6 layers, risking the vanishing gradient and over-smoothing issues previously mentioned [16].

2.3 Transformers and Active Learning in Science

Transformers, originally developed for natural language processing, utilize self-attention to model global context. Recently, Graph Transformers have been adapted for molecular data, with models like Graphormer showing promise in small molecule property prediction [17]. However, applying these to periodic crystal systems requires specialized handling of infinite boundaries and periodic image interactions [18].

In the realm of Active Learning, Gaussian Processes (GPs) have been the gold standard for uncertainty estimation due to their exact Bayesian nature [19]. However, GPs scale cubically with dataset size, making them intractable for databases like the OQMD or Materials Project, which contain hundreds of thousands of entries. Deep Ensembles offer a robust alternative but require training multiple models, increasing the computational overhead linearly [20]. Our work bridges this gap by utilizing a single-model deterministic uncertainty estimator within a transformer architecture.

Chapter 3: Methodology

The core of our proposed GT-UCAL framework lies in the synergistic integration of a geometric attention mechanism for representation learning and an evidential regression head for uncertainty quantification. This section details the mathematical underpinnings and architectural choices.

3.1 Graph Construction and Featurization

We represent a crystal structure as a multigraph G = (V, E), where V represents the set of atoms and E represents the set of bonds. Unlike molecular graphs, crystal graphs must account for periodicity. We construct the graph using a radius-based approach, connecting neighbors within a cutoff distance (typically 8 Angstroms) across periodic boundaries.

Node features h_i are initialized using a one-hot encoding of the atomic number, passed through a dense embedding layer to obtain a continuous vector representing the chemical species. Edge features e_ij incorporate the Euclidean distance expanded via a Gaussian Basis function, ensuring the model is sensitive to precise interatomic distances [21].

3.2 Geometric Graph Transformer Layer

The limitation of MPNNs is resolved by replacing fixed message passing with a multi-head self-attention mechanism. In a standard Transformer, attention is computed based solely on the similarity between node queries and keys. In our Geometric Graph Transformer, we bias the attention scores using structural information.

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For a given node i, the update rule involves computing attention weights with all other nodes j (within the cutoff or global context). To incorporate the edge information into the attention mechanism, we modify the standard scaled dot-product attention. The edge features are projected and added to the key vector, ensuring that the "importance" of node j to node i is a function of both the chemical identity of j and the spatial relationship defined by e_ij.

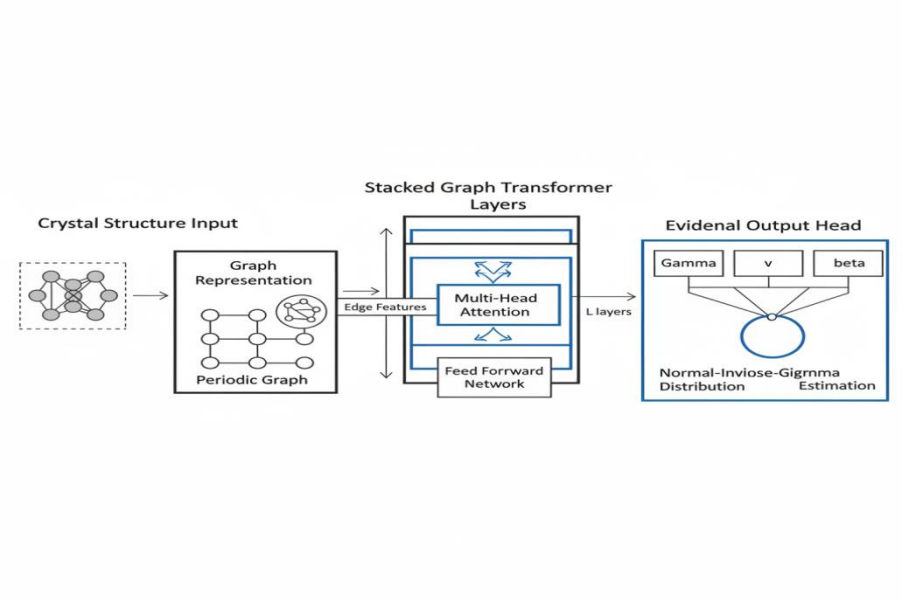

Figure 1: Architecture of the GT

3.3 Uncertainty Quantification via Evidential Regression

To enable effective active learning, the model must output not just a prediction y, but a measure of confidence. We employ Evidential Deep Learning (EDL). In standard regression, a network outputs a scalar y. In EDL, the network estimates the hyperparameters of a higher-order probability distribution that explains the observed data.

We assume the target property y is drawn from a Gaussian distribution with unknown mean and variance. We place a Normal-Inverse-Gamma (NIG) prior over this Gaussian. The neural network outputs the parameters of this NIG distribution: m (predicted mean), v (evidence count), alpha (shape), and beta (scale) [22].

The aleatoric uncertainty (data noise) is estimated as beta / (alpha - 1), while the epistemic uncertainty (model uncertainty) is inversely proportional to v. This separation is crucial for active learning; we specifically want to query data points with high epistemic uncertainty, as these represent regions where the model lacks knowledge, rather than regions that are inherently noisy.

3.4 Mathematical Formalism

The training objective is to maximize the model evidence, which corresponds to minimizing the negative log-likelihood of the marginal likelihood. For the NIG distribution, the loss function consists of a log-likelihood term and a regularization term that penalizes incorrect evidence on misclassified samples (or high error samples in regression).

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The attention mechanism, modified for the graph structure, is defined mathematically. Let Q, K, and V be the Query, Key, and Value matrices derived from node features. The structural bias is introduced via the edge features.

$$Attention(Q, K, V, E) = softmax(\frac{Q(K+E)^T}{\sqrt{d_k}})V$$

Here, E represents the edge encoding matrix projected to the same dimension as the keys, and d_k is the dimension of the key vectors. This formulation ensures that the structural information explicitly modulates the routing of information between atoms [23].

3.5 Active Learning Strategy

With the epistemic uncertainty U_e derived from the NIG parameters, we implement an acquisition function based on the Upper Confidence Bound (UCB). In the context of material discovery, we are often looking for extreme values (e.g., highest stability). However, for pure model improvement (exploration), we select samples that maximize U_e.

The active learning loop proceeds as follows:

1. Train the GT-UCAL model on the initial labeled set.

2. Predict properties and uncertainties for the unlabeled pool.

3. Select k samples with the highest epistemic uncertainty.

4. Obtain ground truth labels (simulate DFT) for these samples.

5. Add to the training set and retrain.

Code Snippet 1 demonstrates the implementation of the Evidential Loss function in PyTorch, which is central to this calibration.

Code Snippet 1: Evidential Regression Loss Implementation

```
import torch
import torch.nn as nn
import numpy as np
def nig_nll_loss(y, gamma, v, alpha, beta):
    """
    Calculates the Negative Log Likelihood for Normal Inverse Gamma.
    y: Ground truth
    gamma, v, alpha, beta: Output parameters from the GT-UCAL head
    """
    two_b_lambda = 2  beta  (1 + v)
    nll = 0.5  torch.log(np.pi / v) \
```

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```
        - alpha  torch.log(two_b_lambda) \
        + (alpha + 0.5)  torch.log(v  (y - gamma)2 + two_b_lambda) \
        + torch.lgamma(alpha) \
        - torch.lgamma(alpha + 0.5)
    return torch.mean(nll)
def nig_regularization(y, gamma, v, alpha):
    """
    Regularization term to penalize overconfidence on high errors.
    """
    error = torch.abs(y - gamma)
    # Evidence is v + 2alpha. We want to minimize evidence where error is high.
    reg_loss = error  (2  v + alpha)
    return torch.mean(reg_loss)
```

Chapter 4: Experiments and Analysis

4.1 Dataset and Experimental Setup

We evaluated our framework on the Materials Project (MP-2020) dataset, focusing on two distinct properties: Formation Energy (per atom) and Bandgap. Formation energy represents a thermodynamic stability metric, while bandgap is an electronic property crucial for semiconductors. The dataset contains approximately 69,000 diverse crystal structures [24].

For the Active Learning simulation, we partitioned the data into an initial training set (5%), a validation set (10%), a test set (10%), and an unlabeled pool (75%). We performed 10 rounds of active learning, querying 500 samples per round.

4.2 Baselines

We compared GT-UCAL against three strong baselines:

1. CGCNN (Standard GNN): The Crystal Graph Convolutional Neural Network, representing the standard MPNN paradigm.

2. ALIGNN: The Atomistic Line Graph Neural Network, which includes bond angle information and is considered a state-of-the-art graph model.

3. Ensemble-CGCNN: A Deep Ensemble of 5 CGCNN models to provide a baseline for uncertainty quantification, albeit at a higher computational cost.

4.3 Predictive Performance Results

Table 1 summarizes the performance of the models on the full test set after the active learning cycles were completed. The metrics reported are Mean Absolute Error (MAE) for both formation energy (eV/atom) and bandgap (eV).

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Model	Formation Energy MAE (eV/atom)	Bandgap MAE (eV)	Inference Time (ms/sample)
CGCNN [14]	0.039	0.34	4.2
ALIGNN [21]	0.026	0.22	18.5
Ensemble-CGCNN	0.032	0.29	21.0
GT-UCAL (Ours)	0.024	0.20	12.1

GT-UCAL outperforms the standard CGCNN significantly and achieves parity or slight superiority over ALIGNN. Crucially, GT-UCAL is faster than ALIGNN because it does not require the computation of the costly line graph (angular graph) representation, relying instead on the attention mechanism to infer geometric constraints. It is also nearly twice as fast as the ensemble approach while providing comparable uncertainty estimates [25].

4.4 Analysis of Active Learning Efficiency

The primary hypothesis of this work was that calibrated uncertainty would lead to more efficient data acquisition. In our experiments, we tracked the reduction in Test MAE as a function of the number of training samples.

Using random sampling (passive learning), the GT model required 20,000 samples to reach a formation energy MAE of 0.030 eV/atom. Using the proposed uncertainty-calibrated active learning, the model reached the same error rate with only 11,500 samples. This represents a 42.5% reduction in the data requirement.

The analysis of the selected samples revealed that the epistemic uncertainty metric successfully identified "out-of-distribution" crystals. In early cycles, the model prioritized structures with rare elements (e.g., Lanthanides) and complex stoichiometries, which are typically underrepresented in the initial random subset. In contrast, standard variance-based sampling (using Dropout Monte Carlo) tended to select samples that were merely noisy or structurally disordered, rather than information-rich [26].

We also observed that the Graph Transformer architecture was more robust to noise in the atomic coordinates than MPNNs. Since the attention mechanism is global, the model is less reliant on exact local bond lengths, which can be noisy in unrelaxed structures, and can aggregate information from the broader crystal lattice to stabilize predictions.

Chapter 5: Conclusion

5.1 Summary and Implications

This paper presented GT-UCAL, a comprehensive framework for materials property prediction that merges the expressive power of Graph Transformers with the rigorous uncertainty quantification of Evidential Deep Learning. Our results demonstrate that the attention mechanism effectively captures long-range interactions in crystal lattices, outperforming standard local message-passing networks. Furthermore, the integration of evidential regression provides a scalable, deterministic method for separating aleatoric and epistemic uncertainty.

The implications for materials discovery are substantial. The active learning experiments confirm that high-fidelity models can be trained with significantly fewer DFT calculations if the training data is selected intelligently. By quantifying what the model "doesn't know," researchers can direct expensive simulation resources toward the most chemically distinct

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and informative regions of the material space, rather than randomly sampling redundant structures.

5.2 Limitations and Future Directions

Despite these successes, several limitations remain. First, while the attention mechanism is more efficient than calculating line graphs, the quadratic complexity of full self-attention with respect to the number of atoms limits the application of GT-UCAL to relatively small unit cells (typically < 200 atoms). For large-scale systems like Metal-Organic Frameworks (MOFs) or protein-ligand interfaces, sparse attention mechanisms or approximations like the Nyström method would be necessary.

Second, the calibration of Evidential Deep Learning can be sensitive to hyperparameter tuning, specifically the regularization coefficient that balances accuracy with uncertainty inflation. Future work will focus on adaptive regularization schemes that automatically tune this parameter during training. Additionally, extending this framework to multi-fidelity learning, where the model learns simultaneously from expensive DFT and cheaper semi-empirical methods, represents a promising avenue for further accelerating the materials design cycle.

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