

Melting temperature prediction using a graph neural network model: from ancient minerals to new materials

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This PDF file includes:

Main Text
Figures 1 to 4

Abstract

The melting point is a fundamental property that is time-consuming to measure or compute, thus hindering high-throughput analyses of melting relations and phase diagrams over large sets of candidate compounds. To address this, we build a machine learning model, trained on a database of ~10,000 compounds, that can predict the melting temperature in a fraction of a second. The model, made publicly available online, features graph neural network and residual neural network architectures. We demonstrate the model's usefulness in diverse applications. For the purpose of materials design and discovery we show that it can quickly discover novel multicomponent materials with high melting points. These predictions are confirmed by density functional theory calculations and experimentally validated. In an application to planetary science and geology, we employ the model to analyze the melting temperatures of ~4,800 minerals to uncover new correlations relevant to the study of mineral evolution.

Significance Statement

High temperature materials properties, such as melting temperature, are generally challenging to measure or compute rapidly. As a result, melting points are known for less than 10 % of the ~200,000 known inorganic compounds. Here we employ machine learning methods to fill this gap by building a rapid and accurate mapping from chemical formula to melting temperature. The model, which we have made publicly available, will facilitate large-scale data analysis involving melting temperature in a wide range of areas, including the discovery of refractory materials, the design of novel extractive metallurgy processes, the modeling of mineral formation and evolution over geological time and the prediction of exoplanet structure.

Main Text

Introduction

Melting points play an important role in a wide variety of disciplines. High-performance refractory materials ¹⁻⁵ have applications ranging from gas turbines to heat shields for hypersonic vehicles. In this context, high melting points correlate with desirable mechanical properties, e.g., high-temperature materials strength as well as good ablation and creep resistance. In geology and planetary science, knowledge of the melting points of minerals provides insight into their formation and evolution in addition to shedding light into the structure of exoplanets.

In these examples, the melting points tend to be high, which considerably complicates their experimental measurement (due to containment and calibration issues). For minerals, these issues are compounded but the fact that many new species are identified from small grains and are thus accessible in limited amounts. Complex phase equilibria and incongruent melting lead to further complications. As a result, the melting temperature is known for less than 10 % of the more than 200,000 inorganic substances with known crystal structures.

In light of these limitations, it would be natural to turn to computational methods. Unfortunately, the calculation and prediction of melting temperatures also an expensive and complex procedure because it involves sampling a large number of configurations.

Numerous efficient methods have been devised to capture melting temperatures from computations ⁶. Using empirical potentials is relatively inexpensive, but it depends on availability and reliability of such potentials. It is both complicated and time-consuming to build a new classical interatomic potential for every new material, not to mention the issue of reliability regarding accuracy. Density functional theory (DFT) calculations are clearly better in terms of generalizability and reliability. However, they remain notoriously expensive, despite increasing power and capability of our computers. The large-size coexistence method ^{7, 8}, which is generally considered the gold standard and widely utilized as a benchmark, typically requires a system size too large for DFT simulations, rendering this approach prohibitively expensive in practice. The single-phase small-size “Z method” ⁹, which heats a solid until it melts, seeks to address this, but suffers from well-documented practical and conceptual problems ¹⁰. Alternatively, one can compute melting temperatures via the free energy method ^{7, 11}, which locates the intersection of the free energy curves of the solid and the liquid. This approach requires highly accurate free energy calculation of the liquid phase, because the two curves cross at a very shallow angle and thus a small free energy shift will result in a large error in melting temperature. Unfortunately, all methods for liquid state free energy computation, such as thermodynamic integration ¹², the particle insertion method ^{13, 14}, and the two-phase thermodynamics method ¹⁵, are expensive and challenging. In earlier work, we proposed the accurate and relatively more efficient small-size coexistence method ^{6, 16}, and developed the SLUSCHI (Solid and Liquid in Ultra Small Coexistence with Hovering Interfaces)

package ¹⁷ to automate the computation process. We have utilized this method to calculate melting temperatures of hundreds of materials, at the cost of several days of computations per compound. Despite such progress, these efforts still fall short of providing a comprehensive view of the melting point landscape.

To overcome these limitations, we turn to machine learning (ML) methods, which are increasingly used for the prediction of materials properties and missing thermodynamic data ^{18, 19}. We build a ML model to predict melting temperature, with an ultimate goal of integrating DFT and ML, which complement each other in terms of speed and accuracy. The ML model allows us to rapidly estimate melting temperatures, at a speed on the order of milliseconds per material, while the DFT calculation provides robustness and accuracy at a much higher cost on the order of several days of computations per material.

To provide training data for our ML procedure, we first build a melting temperature database via web crawling. Melting temperatures are collected and included in our database mostly from Ref. ²⁰. DFT melting temperature calculations are included in the database as well. Our current melting temperature database contains 9375 materials, out of which 982 compounds are high-melting-temperature materials with melting points above 2000 Kelvin. The database consists of chemical compositions, i.e., elements and concentrations, or equivalently chemical formula, of the materials, and their corresponding melting temperatures.

To illustrate the usefulness of our software tool, two application examples are described: *i)* the prediction of melting temperatures for 4828 mineral species, and *ii)* the prediction of compositions with melting temperatures above 3500 K.

Architecture of neural network model for melting temperature prediction

The majority of entries for melting temperatures used for machine learning were collected by parsing data for ~26,000 single-phase compounds from a ten volume compilation of thermodynamic constants of substances ²⁰. The values are based on experimental data from ~51,500 publications before 1982. Only congruent melting temperatures were included in the current version of the database used for machine learning. The data were complemented by results obtained from ab initio molecular dynamic calculations within the SLUSCHI framework ¹⁷.

In our machine learning model, we seek to build a mapping from chemical formula to melting temperature. The minimal requirement of input, only chemical formula here, should facilitate the model's extensive application in the future: no additional materials properties are required as input, and thus neither computational nor experiment data are needed. In our view, relying on input feature would reduce the broad applicability of the method, especially in high-throughput exploration or screening applications, where chemistry is typically the only a priori known input.

The machine learning model combines the Graph Neural Network (GNN)²¹ and residual neural network (ResNet)²² architectures within the Tensorflow²³ framework. (Fig. 1). The GNN architecture is designed to impose permutation invariance (e.g., ZrO₂ and O₂Zr are the same material), which drastically reduces model complexity and thus improves efficiency. The ResNet architecture avoids the problem of vanishing gradient by skipping connections, which also effectively simplifies the network. When a material, i.e., its elements and composition, is fed to the neural network, each element is first converted to 14 features, such as atomic radius, atomic mass, electronegativity, core and valence electrons, ionization energy, electron affinity, density, and position in the periodic table. These features are encoded and passed to the next layer, which we expect to include the key determinants of the individual atomic contributions to the melting temperature. In addition, elemental features interact with each other via the GNN connections, thus leading to contributions from the binary, ternary, and higher-order combination of elements. These encoded contributions are passed to the next layer as well. This layer, consisting of unary, binary, and ternary interactions of the elements and compositions of the material of interest, is fed into a 4-layer ResNet, which leads to the regression and the estimation of melting temperature. Currently the number of elements is limited to four, but this constraint can be removed if a larger combination of elements is needed. For instance, the limit is relaxed to five in the study of minerals later in this paper. More elements increase the complexity of the model and thus the risk of overfitting. The GNN architecture undergoes two iterations of communication among elements, as we find more rounds do not significantly improve performance of the model. Dropout layers are heavily employed in the architecture to avoid overfitting.

The 9375 materials are randomly assigned to training and testing sets, with 8635 materials in the training set, and 740 materials for testing. The training process takes 2000-4000 epochs of optimization. As shown in Fig. 2, the root mean square errors (RMSE) of melting temperature are 110 and 160 K for the training and testing sets, respectively. These surprisingly small errors (DFT error is typically 100K⁶ due to imperfect density functionals) represent the method's accuracy over the ranges of composition that are included in the database. While the testing set is a holdout dataset and thus it provides an unbiased evaluation of the final model, we note that accuracy could degrade for prediction request in relatively poorly sampled regions of composition space, which would demand considerable extrapolation. The errors for materials of different types are shown in Fig. S1 (SI Appendix).

We benchmark our GNN model with XGBoost, one of the most popular gradient boosting methods. Our model score (R²: 0.933, RMSE: 160K) outperforms that of XGBoost (R²: 0.919, RMSE: 183K). This observation is consistent with our expectation and understanding of the two methods. With only chemical formula and elemental features as input, data features are very limited in this work, and thus it favors the neural networks method, which is more capable of combining

and generating features by itself. The moderately large size of our dataset also works well with the neural networks method.

The model is currently hosted at the ASU Research Computing Facilities and available through a web page ²⁴ and Application Programming Interface (API) interface. To use the model, a user needs to visit the webpage and input the chemical compositions of the material of interest. The model will respond with a predicted melting temperature in seconds, as well as the actual melting temperatures of the nearest neighbors (i.e., the most similar materials) in the database. Thus this model serves as not only a predictive model, but a handbook of melting temperature as well. A user may also run batch calculations via command line with much shorter latency, by sending an HTTP POST request to the API server and providing JSON data (elements and compositions of multiple materials) in the body of the POST message. Detailed instruction is available at the webpage.

Mineral melting temperatures: structure and deep time correlations

There are currently more than 5700 approved minerals ²⁵, all naturally occurring compounds which include phases identified exclusively in meteorites and formed during geological processes on Earth. While the composition and structure of all minerals are known (as required for approval of new mineral species), thermodynamic properties, such as melting or decomposition temperatures, are only available for a small fraction of them. The field of mineral evolution, pioneered by Hazen ²⁶, studies occurrence of new mineral species and their increasing chemical and structural complexity as a function of geologic time ^{26, 27}. Recently, the oldest known ages were assigned for 4828 mineral species based on more than 190,000 dated mineral locality occurrences ^{28, 29}. The dataset is constantly updated and made openly available to promote data-driven discovery in mineralogy ^{30, 31}.

We employed our melting temperature database and model (based on 9375 compounds) to analyze the mineral dataset, as well as a subset of 412 minerals containing rare earth elements (lanthanides, Y and Sc). Approximately 6 % of the minerals in the dataset have direct matches in our melting temperature database. For them, experimentally measured values were used in the analysis. For the rest of the minerals, melting temperature (ML T_m) was predicted based on our ML model. Since the majority of minerals are ternary and higher order compounds, containing structural water and carbonate groups, they are unlikely to melt congruently and in this case ML T_m correlates with their decomposition temperature. We interpret ML T_m as the upper boundary of decomposition temperature, as these materials decompose before melting. The dataset is included in the Supplementary Information.

Figure 3A shows the average ML T_m versus oldest known age, grouped with an interval of 250 million years. As expected, the oldest minerals, interstellar and solar nebula condensates

predating Earth formation 4.5 billion years ago, are the most refractory, with average and median melting temperatures around 1700 K. The gradual overall decrease in ML T_m of minerals formed during Earth history is interrupted with two anomalies, which are distinctly pronounced in average and medium ML T_m using 250 or 500 Ma binning. The spike at 3.75 Ga correlates to the proposed timing of late heavy bombardment, hypothesized exclusively from dating of lunar samples and currently debated ^{32, 33}. The dip at 1.75 Ga is related to the first known occurrences of a large number of hydrous minerals and correlates with the Huronian glaciation ³⁴, the longest ice age and thought to be the first time Earth was completely covered in ice.

The rise in average ML melting temperature with increase in symmetry from triclinic to cubic structures (Fig. 3B) is consistent with the observations of the predominant stability of high symmetry structures at high temperatures, as suggested by established experimental phase diagrams. Low symmetry minerals with complex composition typically do not melt congruently, but instead decompose, often to phases with higher symmetry. Analysis of melting temperatures of 412 rare earth containing minerals gives a higher average ML T_m compared to the overall mineral dataset (1296 vs 1005 K, respectively). This is expected due to high melting temperatures of rare earth oxides. Consistent with the full dataset, cubic and tetragonal rare earth minerals represent the smallest fraction, and the highest average melting temperatures, but there is not a clear sequence among monoclinic, hexagonal and orthorhombic minerals (Fig. S2).

The approach based on Shannon's entropy ³⁵ is increasingly used in thermodynamic modeling ^{36, 37}. Krivovichev's index ³⁸ provides a quantitative evaluation of structural complexity in bits of Shannon's information per atom. In addition to symmetry, it accounts for the size of the unit cell and chemical diversity. The higher the bit/atom values, the lower the entropy of the structure ³⁹. For rare earth containing minerals, predicted melting temperature shows a strong negative correlation with the structure complexity index (Fig. 3C). The rare earth minerals with index below 2.5 bits/atom have predicted melting temperatures above 1500 K.

Discovery of new high temperature materials

Based on this moderately accurate but extremely rapid model, we run simulations to showcase its possible applications. Here we present one potential application in the design and discovery of high-melting-point materials. We run Monte Carlo (MC) simulations to generate a list of ternary compounds, which are predicted by the model as top candidates for high-melting-point materials. Since the model takes inputs in the form of elements and compositions, there are only five variables, three elements and two compositions (the sum of mole fractions must equal 1), to describe a ternary compound. Any combination of any element and composition is allowed in our simulation, i.e., any element in the periodic table. The Metropolis algorithm and

simulated annealing technique are employed to maximize melting temperature. The simulation explores the surface of melting temperature and searches for the global maximum of the surface, which is defined by the elements and composition and estimated by the GNN model. The initial MC temperature is set to 100 K, sufficiently high to allow the exploration to escape local minima. The temperature is linearly decreased to 0 K over 10,000 MC steps. We allow changes in both elements and compositions. After each MC run, we obtain a candidate, which presumably is the global maximum of the melting temperature surface as long as the MC trajectory is sufficiently long. In order to generate a list of top high-melting-temperature materials, we run a series of MC simulations in sequence, in which we exclude materials already found from the search in the next iterations, i.e., in the $(n + 1)$ th iteration, the top n material systems already discovered in the previous n iterations are excluded from the search, in order to encourage the exploration of new materials. After this series of MC simulations, we generate a list of top candidates, ranked by their melting temperatures.

As illustrated in Fig. S3, the top twenty candidates are overwhelmingly carbides and nitrides (i.e., two metallic elements plus C or N), with the only exception being the Hf-C-N system, exactly the carbonitride we previously predicted as the material with the world's highest melting temperature, based on DFT molecular dynamics (MD) calculations using the SLUSCHI package⁴⁰. This discovery was also later confirmed independently from experiment⁴¹⁻⁴³. We note that the DFT melting points of the Hf-C-N system were deliberately excluded from the database and the GNN model, in order to challenge the model and examine its predictive capability. The outcome, that the model accurately discovers the Hf-C-N system, is promising and this approach arguably outperforms our human intuition: when we found the Hf-C-N system from DFT, we searched among possible combinations of five elements, Hf, Ta, B, C, and N, and discovered Hf-C-N as the best compound. In contrast, the model correctly predicts Hf-C-N as the most promising candidates, which could have saved us significant time and effort in DFT MD simulations. This capability suggests the model's potential application in materials design and discovery. The melting temperature predicted by the GNN model is at least 500 K lower than that from DFT, which is not surprising since there is no explicit information of the new material system in the dataset and this melting temperature must be pieced together from other similar material systems.

In the next step, we will include DFT melting temperature of the Hf-C-N system, re-train and improve the ML model, and repeat the MC simulations to search for even higher melting temperatures. As summarized in Fig. 4, the new list of top candidates now suggests possibilities with other nonmetals in addition to C and N. We plan to calculate DFT melting temperatures for these materials, which will not only corroborate the discovery of high melting temperature, if indeed favorable, but also further improve our melting temperature database when we include the new DFT results. This work is beyond the scope of this paper but it will be carried out in the near future.

Summary and future directions

We built a melting temperature database and an ML GNN model to predict melting temperature from chemical formula. We demonstrated the utility of prediction of melting temperature by providing new correlations for mineral evolution and directions for further experimental and computational search for new high temperature materials. The model is openly available through web interface and will be updated as new data for neural network training will be collected. We have built a next version of the model and deployed it online at our webpage, which further improves the model's performance. The model is an ensemble model of 30 GNN models based on re-shuffling training and testing datasets using bootstrap, which further reduces overfitting.

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Figures and Tables

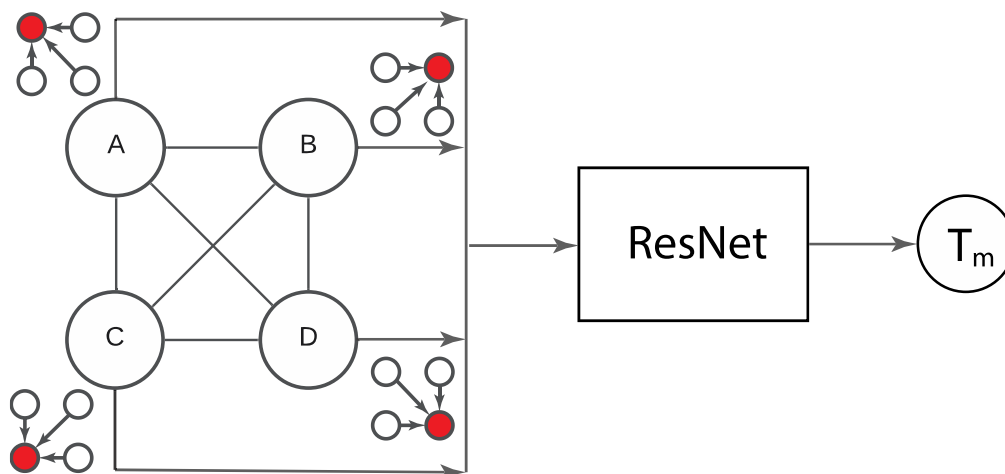


Figure 1. Architecture of the GNN model for ML melting temperature prediction. Each circle represents an element and its composition in the material. Up to four elements are connected in this graph (denoted as A, B, C, and D). First, each element and composition are converted to 14 features, which are then encoded and fed to the ResNet input layer. The circles communicate with each other in order to account for higher order contributions. For example, each circle (element and composition) pulls information from other circles via the GNN. The outputs are then sent to the ResNet. The latter consists of four fully connected layers with skipping connections and leads to the regression analysis for melting temperature prediction

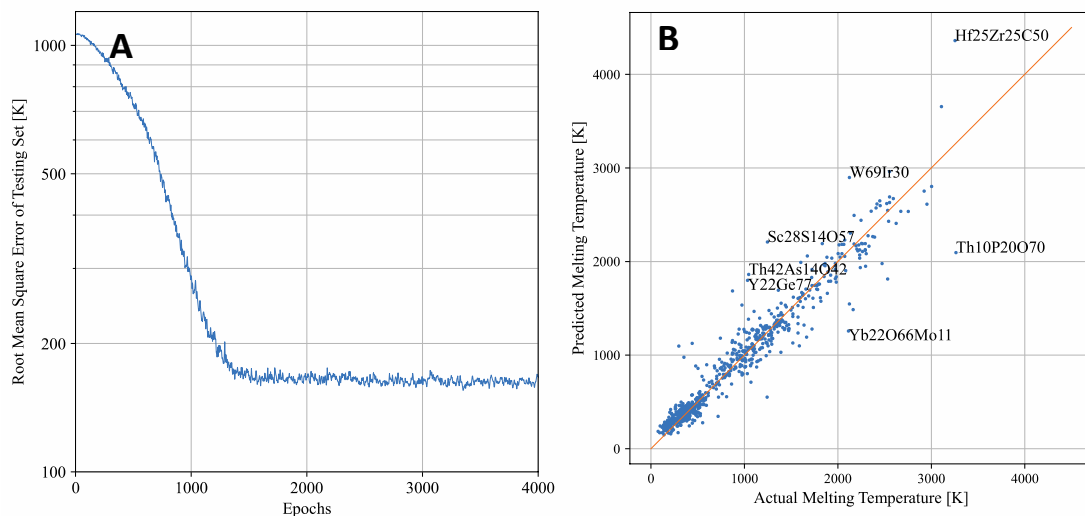


Figure 2. **A)** Root mean square error during model training. The training process was completed after 2000 epochs. **B)** Predicted vs. actual melting temperatures in the testing dataset. The root-mean-square errors are 110 and 160 K for the training and testing sets, respectively. Compounds with large errors are labeled for further investigation.

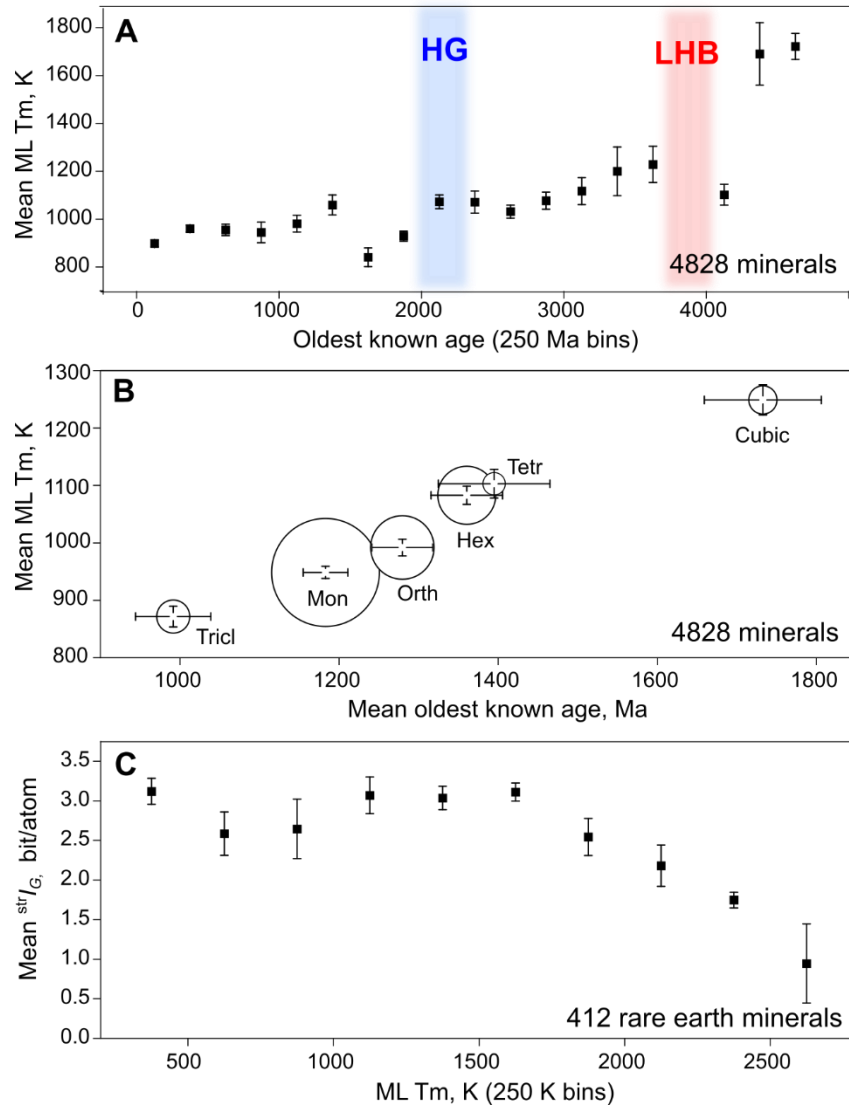


Figure 3. Machine learned melting temperature (ML Tm) for minerals from RRUFF dataset **A**) Mean ML Tm vs. oldest known age. The highlights correspond to the timing of hypothesized Late Heavy Bombardment (LHB) and Huronian Glaciation (HG) events. **B**) ML Tm vs. oldest known age for all minerals grouped by crystal systems. The size of the circles scaled with the number of minerals: (cubic: 500; tetragonal: 383; hexagonal: 855; orthorhombic: 932; monoclinic: 1600; triclinic: 509. **C**) Mean structural complexity index vs. predicted melting temperature for 412 rare earth containing minerals. The error bars correspond to standard errors.

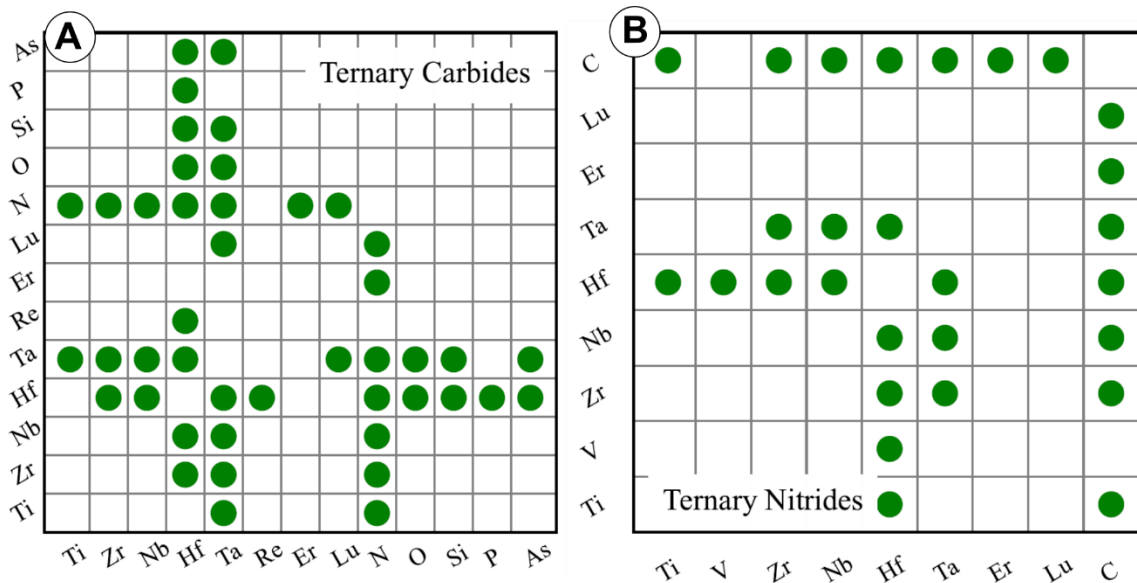


Figure 4. Constituent elements in ternary carbides (A) and nitrides (B) with predicted melting temperatures above 3500 K. Each dot represents one ternary compound, M1-M2-C, or M1-M2-N with M1 and M2 being the corresponding elements on the axes.

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Supplementary Information

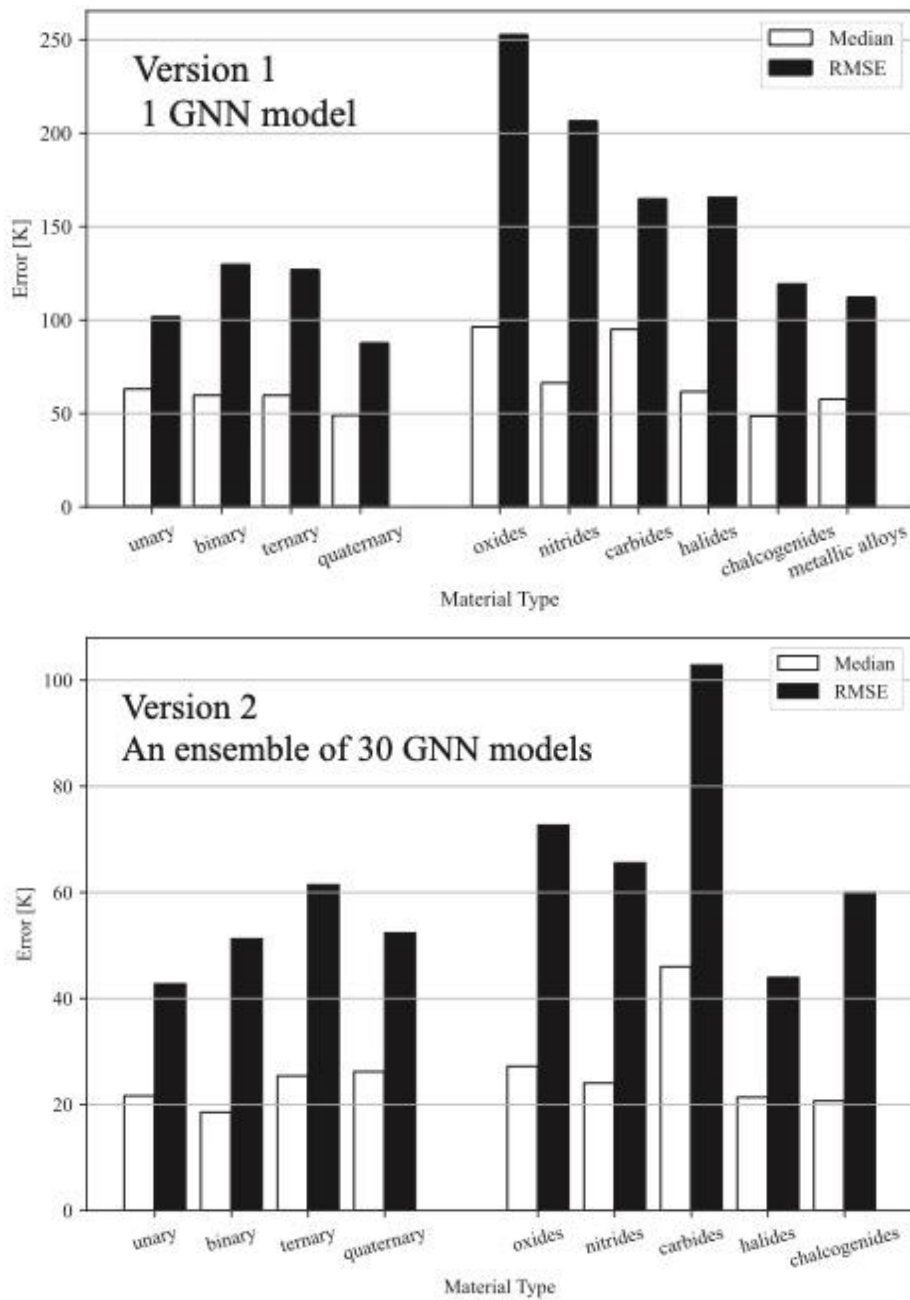


Fig. S1. Median error and root mean square error (RMSE) for materials of different types, evaluated over all data points. Stronger elemental interactions, as observed in oxides, nitrides, and carbides tend to incur relatively larger errors, since these contributions are diverse and more difficult to capture.

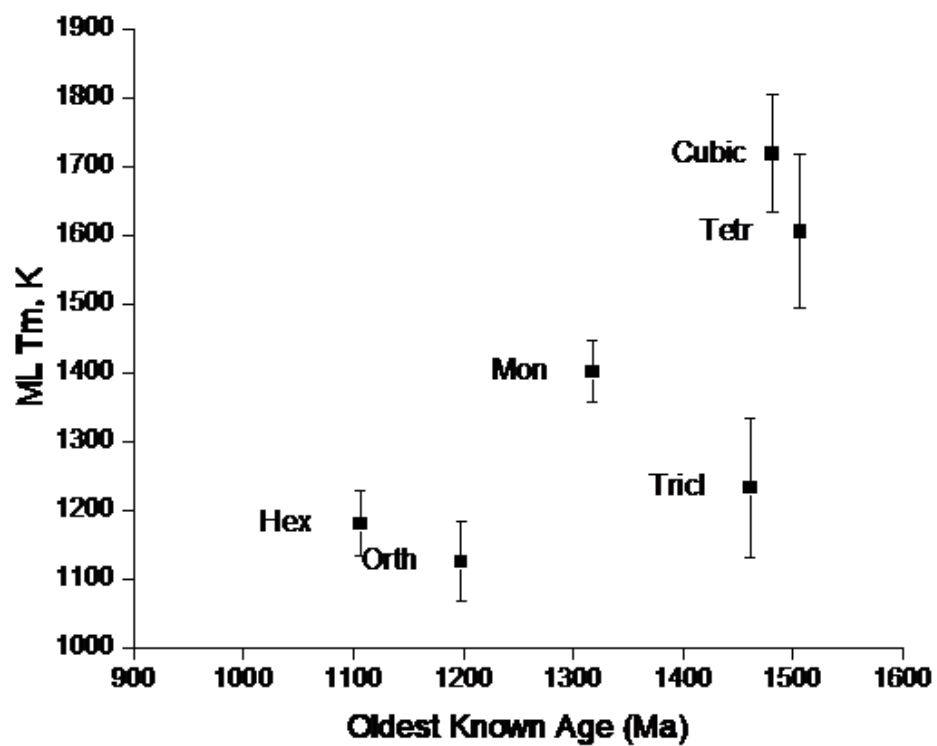


Fig. S2. Predicted melting temperature¹ vs. oldest known age for 401 rare earth containing minerals from RRUFF dataset (<https://rruff.info/ima/>)^{2,3}, averaged by crystal system (cubic: 21; tetragonal: 23; orthorhombic: 80; hexagonal: 112, monoclinic: 138). Melting temperature model was built based on a dataset of 9375 compounds.

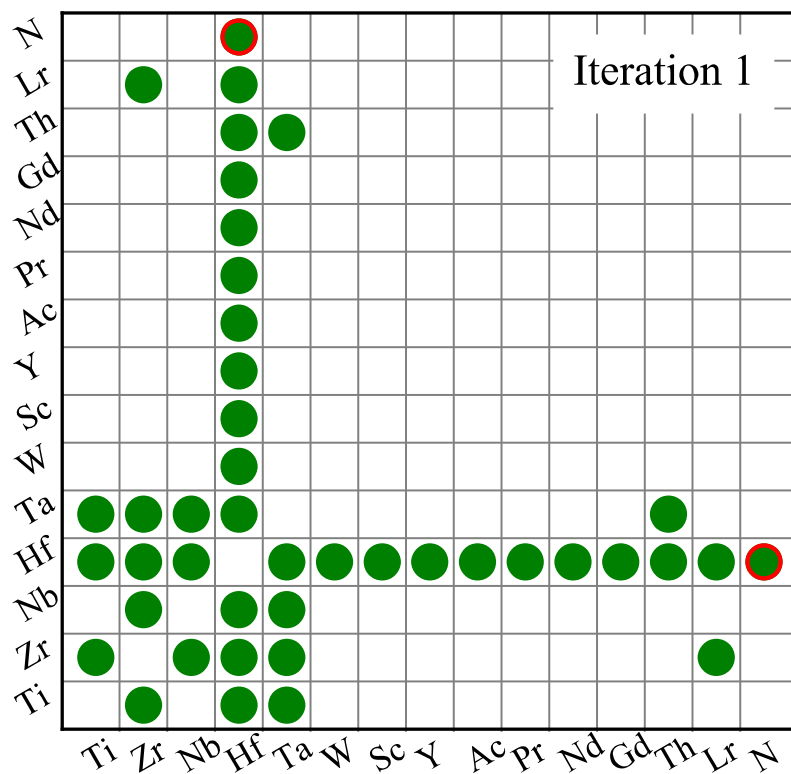


Fig. S3. Prediction of high melting temperatures in ternary systems, based on the GNN model¹ and MC simulated annealing. The top candidates are dominated by carbides and nitrides. The only exception is a carbonitride, the Hf-C-N system, which is exactly the material of the highest melting temperature predicted in 2015 based on DFT MD simulations.⁴ The DFT melting temperatures of the Hf-C-N system were deliberately excluded from the database and ML model to test the method's predictive capability.

Legends for datasets

DATASET_S1_Minerals_ML_Tm_20220503.xlsx

Microsoft Excel spreadsheet with melting temperatures for 5793 mineral species calculated on May 8, 2022 using Melting Temperature Predictor Based on Machine Learning Graph Neural Networks (GNN).¹ The list of mineral species with compositions, crystal systems, and oldest known age was downloaded from RRUFF database (<https://rruff.info/ima/>)^{2, 3} on 05/03/2022. The values of melting temperatures for compositions directly matched with the database used for training of GNN are selected in bold. Melting temperature model was built based on a dataset of 9375 compounds.

DATASET_S2_RE_minerals_ML_Tm_20220503.xlsx

Microsoft Excel spreadsheet with melting temperatures for 412 rare earth containing mineral species calculated on May 8, 2022 using Melting Temperature Predictor Based on Machine Learning Graph Neural Networks (GNN).¹ The list of mineral species with compositions, crystal systems, and oldest known age was downloaded from RRUFF database (<https://rruff.info/ima/>)^{2, 3} on 05/03/2022. The structural complexity indexes received from S.V. Krivovichev (also partially available at https://info.deepcarbon.net/vivo/gemi_minerals (Deep Carbon Observatory Data Portal)).⁵ The values of melting temperatures for compositions directly matched with the database used for training of GNN are selected in bold. Melting temperature model was built based on a dataset of 9375 compounds.

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