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Brief Report

Machine-Learning-Based Composition Analysis of the Stability of V–Cr–Ti Alloys

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Abstract: Machine learning methods allow the prediction of material properties, potentially using only the elemental composition of a molecule or compound, without the knowledge of molecular or crystalline structures. Herein, a composition-based machine learning prediction of the material properties of V–Cr–Ti alloys is demonstrated. Our machine-learning-based prediction of the stability of the V–Cr–Ti alloys is qualitatively consistent with the composition-dependent experimental data of the ductile–brittle transition temperature and swelling. Furthermore, our computational results suggest the existence of a composition region, Cr+Ti ~ 60 wt.%, at a significantly low ductile–brittle transition temperature. This outcome contrasts with a reportedly low Cr+Ti content of less than 10 wt.% in conventional V–Cr–Ti alloys. Machine-learning-based numerical stability prediction is useful for the design and analysis of metal alloys, particularly for multicomponent alloys such as high-entropy alloys, to develop materials for nuclear fusion reactors.

Keywords: materials informatics; machine learning; nuclear fusion; nuclear fission; reactor materials; vanadium; chromium; titanium; ductile–brittle transition temperature; swelling

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1. Introduction

The design or screening of alloys for specific applications has conventionally relied on a series of experiments and first-principle calculations. However, conducting experiments for a wide range of elemental compositions, particularly for higher-than-binary alloys, is extremely laborious. Furthermore, the first-principle calculations, such as density functional theory (DFT) and molecular dynamics simulations, require high computational costs and knowledge of the crystalline or molecular structure, such as the spatial configuration of atoms. The machine learning method allows the experience-based prediction of material properties, potentially only using the atomic composition of the molecule or compound without the knowledge of the molecular or crystalline structure, by utilizing the existing thorough database of the DFT calculation results for training. In the field of structural materials for nuclear fusion and fission reactors, the most common plane-wave DFT calculations are suitable for inorganic solids, particularly metal compounds.

The V–Cr–Ti alloy is a promising material for the various structural components of nuclear power plants, such as blanket, first wall, and divertor components in fusion reactors, as well as structural materials in fast breeder reactors and high-temperature gas-cooled reactors, owing to its high-temperature mechanical strength, excellent thermal stress factor, superior low-temperature ductility, high resistance to neutron irradiation, low induced activation, and compatibility with the liquid-Li environment [1–6]. Herein, composition-based machine learning prediction of the material properties of the V–Cr–Ti alloys is demonstrated as a case study. To the best of our knowledge, this study is the first attempt to apply the machine learning method to the V-based material systems.

2. Methods

An open-source code ElemNet was used to predict the stability of the V–Cr–Ti alloys based on their elemental composition. The ElemNet model is a 17-layered, fully con-

nected deep neural network developed for the prediction of formation energy (enthalpy) using only elemental composition [7,8]. The neural network architecture of ElemNet has been summarized in the literature [7]. A rectified linear unit was used as a neuronal activation function. The tuned hyperparameters of the model were as follows: number of layers: 17; momentum: 0.9; dropouts: 0.7, 0.8, and 0.9; and the optimization algorithm: stochastic gradient descent [7]. The model is trained on the enthalpies of formation of 341,000 compounds with unique elemental compositions, determined by first-principle DFT calculations available in the Open Quantum Materials Database [9]. The error analysis for the ElemNet indicated well-performed deep learning with a mean absolute error of 0.042 eV/atom in a 10-fold cross-validation [8]. ElemNet utilizes artificial intelligence to capture the essential chemistry for predicting material properties by automatically learning the chemical interactions and similarities between different elements. The ElemNet code was operated in the energy-prediction mode based on the pretrained model in a Python 3.7 environment with extension modules, including NumPy 1.21 and TensorFlow 1.14, and considering the elemental composition of the metal alloy as the only input.

3. Results and Discussion

Figure 1 presents the correlation between the enthalpy of the formation of the V–Cr–Ti alloys, computed by the ElemNet machine learning model and DFT calculations registered in the Open Quantum Materials Database. Only four DFT data points are available for ternary V–Cr–Ti compounds in the Open Quantum Materials Database as of March 2023, reflecting the difficulty of using first-principle calculations for multicomponent alloys and indicating the necessity of machine-learning-based prediction. Despite the small number of datasets, an excellent agreement between the machine learning prediction and the DFT values is observed in Figure 1, with a mean absolute error of 0.015 eV/atom.

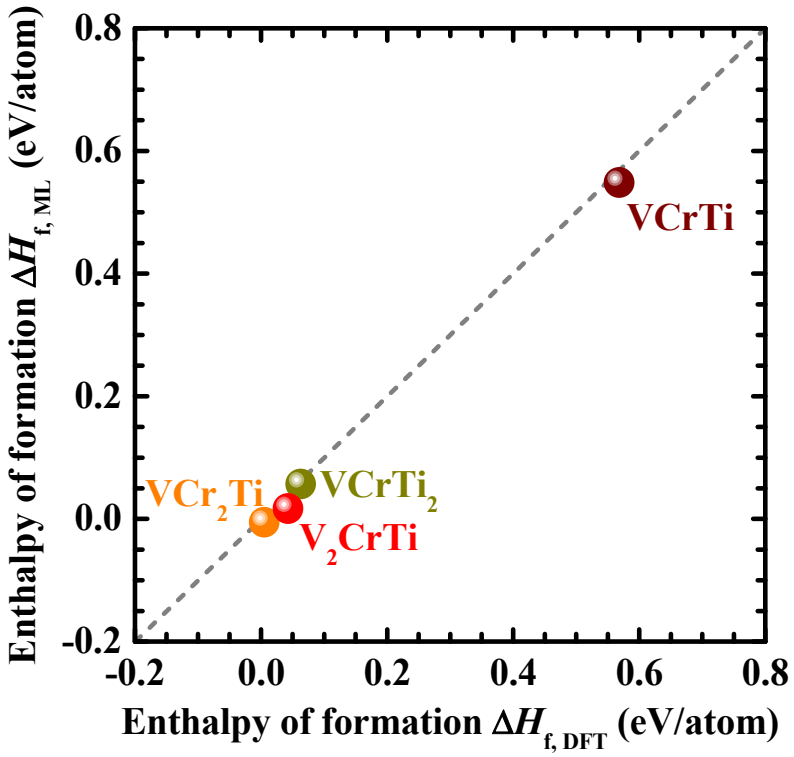

Figure 1. Correlation between the enthalpy of formation of V–Cr–Ti alloys computed by the ElemNet machine learning model, $\Delta H_{f, ML}$, and density functional theory calculations registered in the Open Quantum Materials Database, $\Delta H_{f, DFT}$.

The enthalpy of formation, ΔH_f, represents the stability of a material in the opposite direction of its value. The negative (i.e., sign-reversed) enthalpy of formation, $-\Delta H_f$,

therefore directly represents the stability. Figure 2 shows the computed $-\Delta H_f$ of the V–Cr–Ti alloys as a function of the Cr content at a fixed Ti content of 4 wt.%. In addition, a set of experimental data of the ductile–brittle transition temperature (DBTT) of the V–Cr–Ti alloys with a Ti content of 4 wt.% as a function of the Cr content are plotted in Figure 2 [10]. Brittle failure can occur at temperatures below the DBTT. Therefore, low DBTTs are preferred to be used as structural materials. In other words, a low DBTT increases the applicable operating temperature range because the structural material needs to be used at a temperature higher than its DBTT. Generally, the Cr+Ti content in the composition of the V–Cr–Ti alloys is maintained below 10 wt.% to avoid entering the high-DBTT region [1–6,11]. As shown in Figure 2, the computational result of $-\Delta H_f$ reproduced well the trend of the experimental DBTT data. In the Cr content region of less than 20 wt.%, an increase in the Cr content of the V–Cr–Ti alloys leads to an increase in the DBTT [10,12,13]. Experimentally, this trend is attributed to the effect of solid-solution strengthening and Ti-based precipitates such as Ti-oxycarbonitrides, induced by the inclusion of Cr [6,10,12,13]. In addition, an increase in the computed $-\Delta H_f$ can be attributed to the entropic stabilization effect of the elemental mixture. It may be an interesting study in future to correlate these effects in order to obtain a deeper understanding. Intuitively, the difference in energy related to the material stability corresponds to the difference in the transition temperature as $\Delta E \sim nk_B\Delta T_{trans}$, where E represents the potential energy of a molecule, n is the number of orders around unity, k_B is the Boltzmann constant, and T_{trans} is the generalized transition temperature of the molecule. In this context, the relationship observed in Figure 2, $\Delta(-\Delta H_f) = 5k_B\Delta T_{DBT}$, where T_{DBT} is the DBTT, is considered to be positioned within a feasible range.

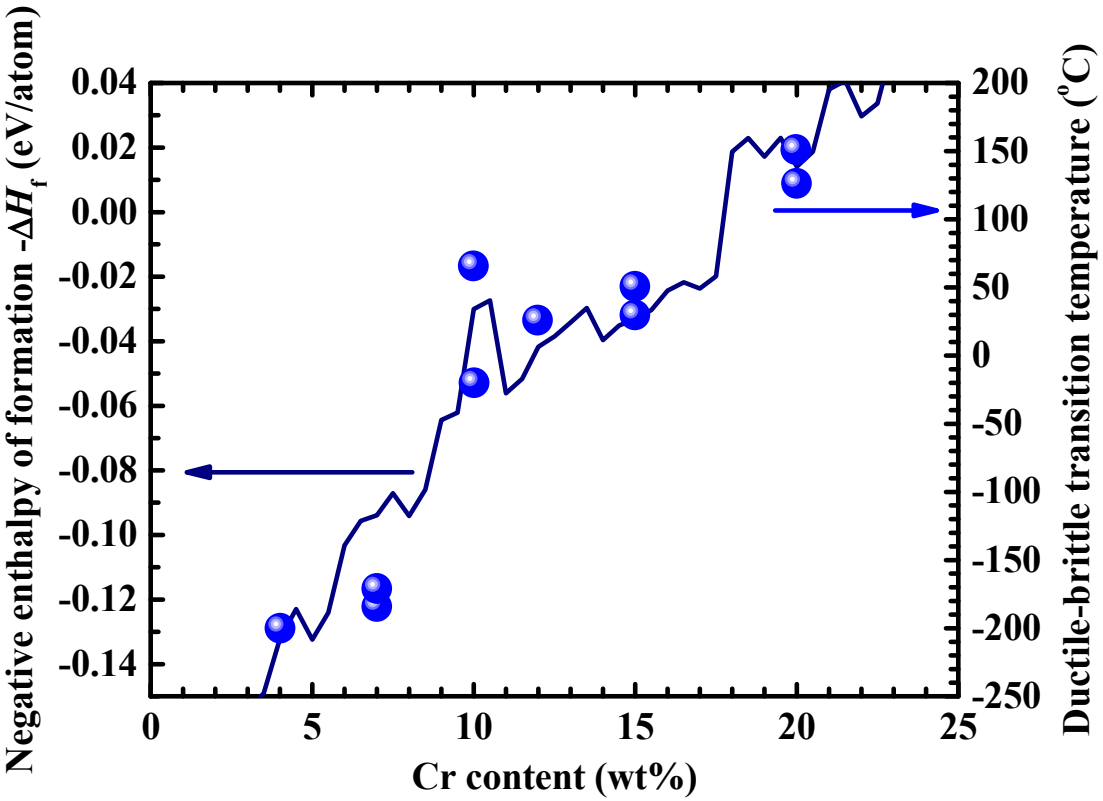

Figure 2. Computed negative enthalpy of formation (line) and experimental ductile–brittle transition temperature (dots) [10] of V–Cr–Ti alloys as a function of the Cr content at a Ti content of 4 wt.%.

As an extension of Figure 2, Figure 3 demonstrates the computed ΔH_f of the V–Cr–Ti alloys as a function of the Cr content at Ti contents of 4, 8, and 16 wt.%. The ΔH_f becomes minimum in the intermediate region of Cr content, 40–60 wt.%, for alloys with a low Ti content, such as 4 and 8 wt.%. Interestingly, an unstable regime appears around

the intermediate region of the Cr content at a higher Ti content of 16 wt.%, where DBTT decreases. This is further discussed based on the computational results, as shown in Figure 4.

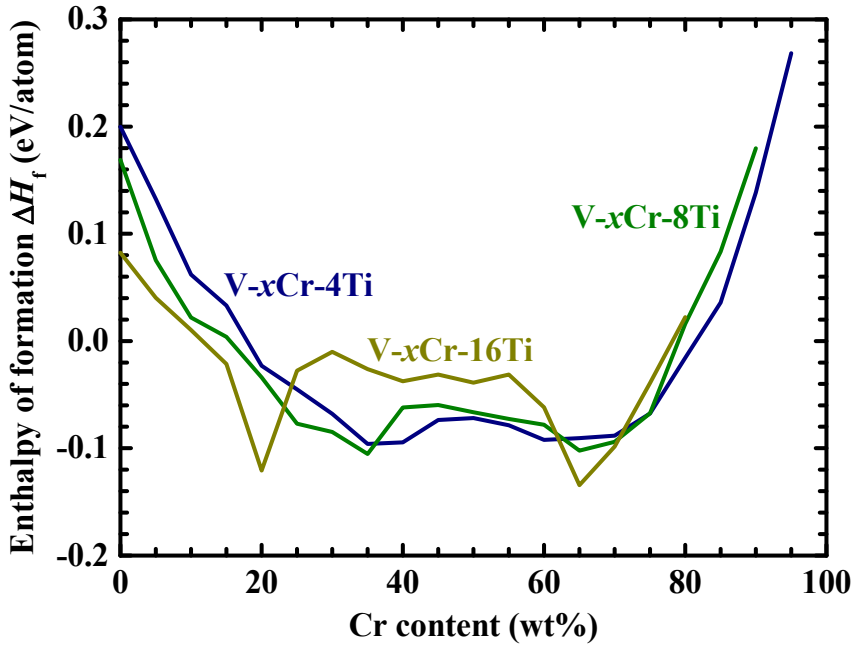

Figure 3. Computed enthalpy of formation of V–Cr–Ti alloys as a function of the Cr content at the Ti content of 4, 8, and 16 wt.%.

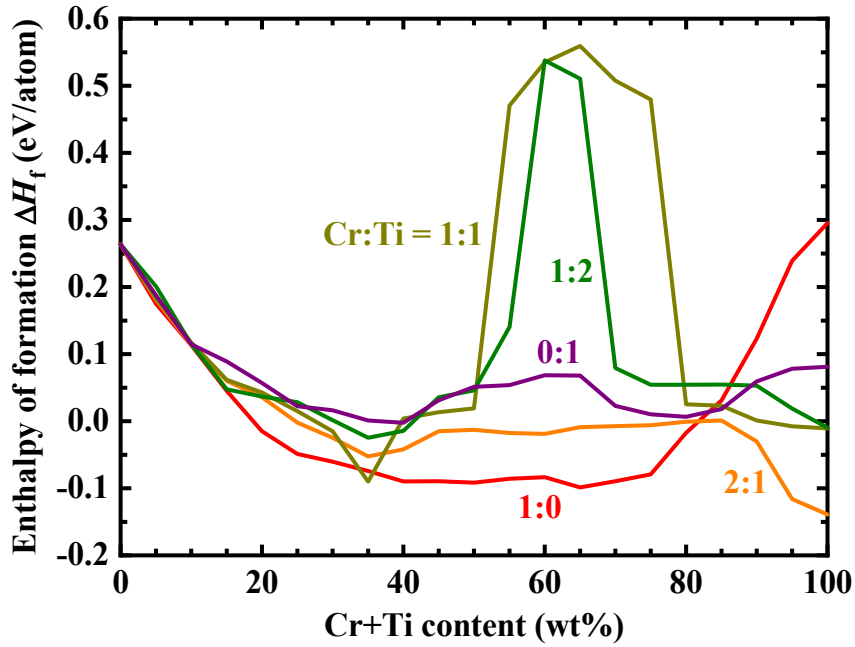

Figure 4. Computed enthalpy of formation of V–Cr–Ti alloys as a function of the Cr content for the weight ratio of Cr:Ti = 1:0, 2:1, 1:1, 1:2, and 0:1.

Figure 4 presents the computed ΔH_f values of the V–Cr–Ti alloys as a function of the Cr content at weight ratios of Cr:Ti = 1:0, 2:1, 1:1, 1:2, and 0:1. The computational results do not successfully reproduce the series of experimental DBTT data reported in [11], where the DBTT is evidently low and high when the Cr+Ti content is lesser and higher than 10 wt.%, respectively. Nevertheless, our results were qualitatively consistent with

the experimental data for a monotonic increase in DBTT below 25 wt.%. In addition, our results were consistent with the trend observed in the experimental swelling data reported in [14], confirming the higher stability of the Ti content for the region below 20 wt.% with topping out. Related to the result shown in Figure 3, Figure 4 demonstrates a decrease in ΔH_f up to a Cr+Ti content of approximately 40 wt.%; however, ΔH_f increases and reaches a maximum at a Cr+Ti content of approximately 60 wt.%, particularly for alloys with a high Ti content. This result contrasts with the conventional status, where only the low Cr+Ti content regime, <10 wt.%, in V–Cr–Ti alloys was considered for applications [1–6]. This prediction of the significantly high ΔH_f and expectedly low DBTT in the intermediate region of the Cr+Ti content can be utilized for the design of structural materials.

4. Conclusions

In this study, the composition-based machine learning prediction of the material properties of V–Cr–Ti alloys was demonstrated as a case study. The machine-learning-based prediction of the stability of the V–Cr–Ti alloys was qualitatively consistent with the experimental data of DBTT and swelling as a function of the elemental composition. In addition, our computational results suggested the existence of a composition region with high Cr and Ti contents (Cr+Ti ~ 60 wt.%) and significantly low DBTTs. This outcome contrasts with the low Cr+Ti content of less than 10 wt.% in conventional V–Cr–Ti alloys. Thus, the machine-learning-based numerical stability prediction may be useful for the design and analysis of metal alloys, particularly for multicomponent high-entropy alloys, to develop materials for nuclear fusion and fission reactors.

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Data Availability Statement: The data, supporting the plots in this study and other findings of this study, are available from the corresponding author upon reasonable request.

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Conflicts of Interest: The authors declare no competing financial interests or personal relationships that may have influenced the work reported in this study.

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