論文の内容の要旨

Thesis Summary

Material Property Predictions and Discovery Using a Novel Descriptor "Elemental Fingerprints" with Neural Networks
(新規記述子「元素フィンガープリント」を用いたニューラルネットワークによる材料物性の予測及び材料探索)

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In this dissertation, I explore how machine learning techniques can be applied to the field of material science and engineering. From simple prediction problems starting with linear regression to complicated problems like detecting a person in an image, machine learning has shown processing capability that goes beyond human ability. Machine learning is also being tried out in the field of materials science, from the simple prediction of a physical property to the interpretation of experimental results and suggestion of candidates for materials discovery. However, there are many obstacles to combine actual materials research and development with machine learning. Many previous studies are based on the use of detailed information of material (the atomic coordination, crystal structure, interatomic distance, etc.) because adding information gives better training results.

However, in testing material properties through actual experiments, it is sometimes difficult to obtain information such as atomic structures. Therefore, I focused on the situation where the independent variables available for machine learning are just chemical compositions and experimental conditions. The most intuitive descriptor that can be used in this situation is molar concentration or the presence of elements of materials. However, the molar concentration of raw material can only be applied to the composition range already tested. Some researchers

proposed to use the elemental properties of the elements in the material, such as the electronegativity of A-site element and the ionic radius of B-site element. However, it can be applied to only the case of materials with the same crystal structures.

Therefore, in this dissertation, I have proposed “elemental fingerprints” as a novel descriptor that generates input features for machine learning from limited information but can be used in various materials. Elemental fingerprints are a set of the frequency distribution of elemental properties or physical and chemical properties of raw materials. Elemental fingerprints are designed on the premise that if an elemental property is correlated with a material property, then an element with a similar elemental property will make a similar contribution to the corresponding material property. It can be described as compiling frequency distributions of each elemental property using chemical composition. Each bin will be assigned elements with similar elemental properties. Elemental fingerprints are a group of these frequency distributions created by the various elemental properties. This process is the most conspicuous feature of elemental fingerprints, which differentiates this descriptor from others. Since elemental properties required here are the data of pure elements or raw materials, not the data of materials to be analyzed, various elemental properties can be relatively easily gathered.

I also attempted to use the latest machine learning techniques in materials research. Through this dissertation, I used the neural network models for the prediction and exploration of materials. Not only did I use a simple multilayer perceptron, but also I tried to use adversarial training and neural network ensemble, which is actively discussed in the field of image recognition. The adversarial neural network ensemble makes it possible to estimate the prediction uncertainty, the exact inference of which is known to be computationally intractable. However, it is not yet tested whether the prediction uncertainty estimated by this technique is enough to perform Bayesian optimization or not. Thus, I tested the performance of Bayesian optimization with adversarial neural network ensemble in order to see its applicability to material discovery.

In Chapter 4, I tested the potential of elemental fingerprints using the viscosity of oxide glass materials. I used seven properties of oxides and their cations to predict the isokom temperature, which the temperature required to achieve a specific viscosity, i.e., $10^{6.6}$ Pa·s. It means that when I compile frequency distributions with 20 bins, elemental fingerprints have 140 features or 140-dimensional descriptor. I succeeded in predicting the isokom temperatures

within a root mean square error of 33 ℃. Not only predicts the material properties, but elemental fingerprints can also find out that the density and melting point of raw oxides are the most important physical properties to explain the viscosity of glass. Because the neural network model is one of the black box models, interpretation of prediction results is usually difficult. However, elemental fingerprints make it possible to interpret the cause of prediction even of the neural network model. That is because the selection of elemental properties is one of the hyperparameters that can be optimized and analyzed.

I also examined the possibility of the prediction of unseen data. That is, to demonstrate the further advantage of the fingerprints, I tested an extreme virtual case of material search, which cannot be performed at all by the existing descriptors such as molar concentration. For this purpose, I set a specific target oxide in raw materials and trained the neural network model using materials that do not contain the target oxide. Then, I tested the prediction accuracy of the glass materials, including the target oxide, with the neural network model trained in this manner. I succeeded in predicting the isokom temperatures within a root mean square error of 53℃ for nine target (unseen) oxides using elemental fingerprints. This prediction error is much lower than in the case of using elemental attributes (73℃) and molar fraction (62℃). This test shows that even if the chemical composition is completely different, materials can be predicted successfully in case that the elemental fingerprints are similar.

In chapter 5, I attempted the application of adversarial neural network ensemble to material discovery as well as the prediction of material properties using a large database called OQMD. When I used the descriptor set, including elemental fingerprints and other two descriptors as input features, the test prediction error of standard formation enthalpy was 31 meV/atom. This is much lower than not only the prediction error reported by Ward et al. of 88 meV/atom under the same situation, but also the DFT calculation error of 81 meV/atom. In addition, I tested whether it is possible to discover material by Bayesian optimization as well as predictions. However, no one has attempted to apply Bayesian optimization for a large number of inorganic materials, as many as 253,153. Therefore, the first I experimented on material discovery with 16,444 oxide data. However, OQMD currently lacks material properties interesting enough to search for actual materials. Therefore, in this research, I set a virtual target figure of merit as follows:

$$\text{Figure of Merit (FoM)} = \text{Band gap energy(eV)} \times \text{Density}(^{g}/_{cm^3})$$

I tried to search for the best candidate material with the Gaussian process, adversarial neural network ensemble, and random search. The average observation count required to discover the best candidate material was 59, 47, and more than 8,000, respectively. Despite the lack of theoretical background for accurately predicting posterior distribution, the neural network ensemble produces excellent results on par or even better than the results of the Gaussian process. This indicates that neural networks using elemental fingerprints as input features may also be effective in material discovery. Additionally, when I used neural networks not trained in an adversarial but traditional way, its performance was lower than the Gaussian process.

In chapter 6, I tried to predict the tensile properties of metals. I have been shown that a single neural network model could describe all the materials regardless of base elements such as Fe, Ni, and Co. However, it seems that the higher the tensile strength is, the larger the prediction error is. This may be due to the lack of heat treatment conditions in the input features on the current prediction model. The high prediction error may originate from the hardening effects of the heat treatments. However, because these high-strength metals should receive much attention from researchers and industry, the prediction accuracy should be further improved.

Even so, the test results showed the potential of a prediction model that could explain the effects of heat treatments. I tried to train a prediction model using the non-heat-treated metals. The non-heat-treated metals were well predicted. However, heat-treated metals were up to twice as strong as predicted. It is a bad result; however, if the model predicts the strength when the metal is not heat-treated, it may be a reasonable prediction. However, it is currently difficult to explain various heat process techniques such as annealing, tempering, normalizing, and quenching with a single descriptor. My suggestion is as follows: If the tensile properties and surface harness reflect the history of heat processes, we can use these simple and cheap experimental results as descriptors of heat treatment. It could be applied to creep rupture tests, in which the experimental costs are high, would significantly decrease the research and development time of the materials.