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Improving efficiency of autonomous material search via transfer learning from nontarget properties

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ABSTRACT
Recently, autonomous material search methods combining machine learning and experiments/simulations have become indispensable for exploring the extremely vast material exploration space. However, conventional autonomous material search methods focus solely on target material properties and their descriptors, leaving room for improvement in search efficiency. More efficient autonomous material search can be realized by utilizing information on nontarget properties that are dormant in databases. Here, we propose a novel method for autonomous material search using transfer learning and an ensemble neural network. This method can perform an autonomous material search to optimize the target properties while transferring information on nontarget properties. To demonstrate the usefulness of this method, we applied it to search for ternary magnetic alloys with a high Curie temperature while transferring information on magnetic moment and spin polarization. Results indicate that the proposed method improves the efficiency of autonomous material search, particularly in the early stages of the search process.

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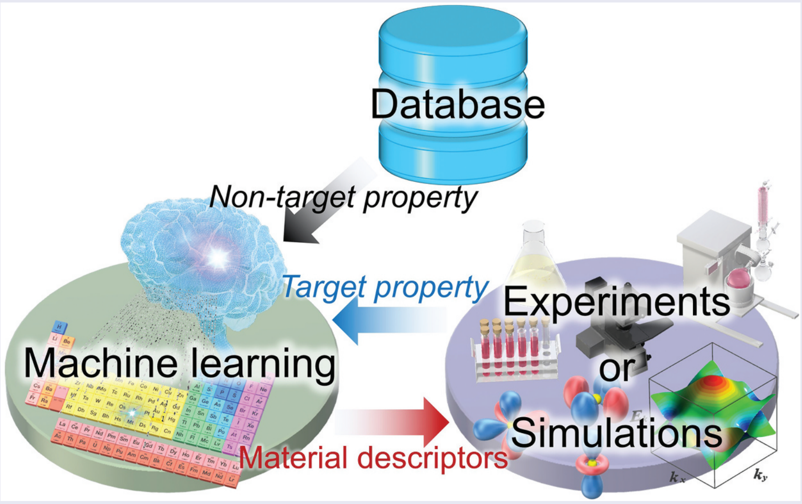

1. Introduction

Recently, the increasing complexity of materials has led to a massive expansion in the material search space. Traditional material search methods involve a laborious process of material synthesis, measurements, and analysis performed by humans. However, with the increasing size of the search space, conventional methods have proven to be inadequate. To address this challenge, autonomous material search methods based on machine learning have been developed. These methods leverage the power of machine learning to identify promising materials without requiring human intervention. One example of this is robotic autonomous material search, which combines material experiments, robotics, and machine learning to perform autonomous material synthesis and measurements [1–9]. Another example is in silico autonomous material search, which combines ab initio calculations and machine learning to efficiently search for materials [10–15]. Using machine learning to identify promising candidate materials and process conditions, these methods can considerably accelerate material discovery.

Typically, conventional autonomous material search methods focus on optimizing a target property using

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Bayesian optimization with data solely based on the target property. Figure 1(a) illustrates this approach, where the autonomous material search loop uses only the data of the target property and its material descriptors (such as composition and crystal structure) to maximize the target property. In the machine learning part of Figure 1(a), a Gaussian regression model is commonly employed: $Y_{TP} = f(X_D)$, where Y_{TP} and X_D represent the target property (objective variable) and material descriptors (explanatory variables), respectively. This model predicts the target property of new materials and quantifies their uncertainties, enabling the determination of material descriptor values for the next material synthesis or material simulation. This autonomous loop of material exploration facilitates the continuous exploration of the material space without requiring human intervention.

However, this conventional approach overlooks the potential benefits of incorporating information on nontarget properties that exist in material databases, limiting the search efficiency. Figure 1(b) shows the process, whereby a machine learning model is constructed by leveraging both target and nontarget property data. Adding nontarget property data can improve the accuracy of the machine learning model and subsequently enable an efficient autonomous material search. However, in the context of autonomous material search, it is impractical to include nontarget property data as explanatory variables in the machine learning model as in $Y_{TP} = f(X_D, X_{NTP})$. Such a model would restrict the search space to those materials with information on the nontarget property X_{NTP}. Thus, another autonomous material search approach that can effectively harness the potential of nontarget properties in existing material databases is necessary.

Herein, we introduce a new approach for autonomous material search, illustrated in Figure 1(b). Our proposed method streamlines the process of optimizing target properties while simultaneously considering nontarget properties, affording an efficient search for materials with desired characteristics. To show the effectiveness of our approach, we apply it to data on the magnetic moment M, spin polarization S, and Curie temperature T_c of ternary magnetic B2 structure materials.

2. Proposed method

Typically, Bayesian optimization with the Gaussian process regression model is the standard approach for autonomous material search. However, incorporating nontarget property information into the Gaussian process regression model can be challenging. Therefore, instead of the Gaussian process regression, we utilized an ensemble neural network (ENN) approach [16]. An ENN employs multiple independently trained neural networks (NNs), producing results superior to those obtained by employing a single NN. For instance, in numerical prediction tasks, taking the average of predicted values from NNs can enhance the prediction accuracy. Additionally, an ENN can approximate the distribution of predicted values, enabling us to quantify uncertainty akin to the Gaussian process. Consequently, the average of predicted values from an ENN serves as the prediction output for autonomous material search, with the variance of predicted values reflecting the degree of uncertainty.

Moreover, the ENN approach offers transfer learning, a technique that enhances model performance by pretraining with diverse data beforehand and retraining the model with the target data [17]. In autonomous material search, transfer learning enables us to leverage nontarget property information. Herein, we transferred the knowledge obtained by pretraining an ENN model with nontarget property data to the ENN model utilized in the autonomous material search.

We employed an ENN comprising 10 NNs, each comprising three hidden layers with 100, 100, and 10 neurons, respectively, from the input to the output

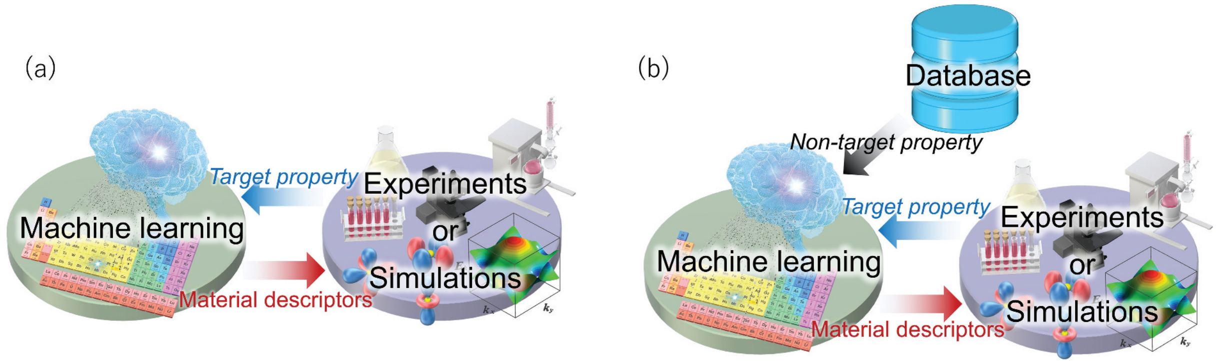

Figure 1. Comparison of conventional and proposed new autonomous material search methods. (a) The conventional autonomous material search method used machine learning and material experiments/simulations. To optimize the target properties, conventional autonomous material search is performed using only the target properties and their material descriptors. (b) The proposed autonomous material search method. Target properties and nontarget properties are utilized in the autonomous material search.

side. For transfer learning, we froze two input-side layers and fine-tuned one output-side layer. During training, we applied early stopping using a validation set comprising 20% randomly selected training data. In other words, parameters were updated through backpropagation using 80% of the data and the remaining 20% determined the optimal parameter selection for the final training results. We utilized the upper confidence bound as the acquisition function, where the predicted values and uncertainty are ensemble average and variance, respectively [18]. We set the exploration coefficient to 3.0. Further details regarding ENN settings and performance are provided in Supplementary Information S1 and S2.

3. Demonstration

To demonstrate the efficiency of ENN-based autonomous search, we applied our methodology to a dataset of ternary magnetic alloys with a B2 structure. The B2 crystal structure is shown in Figure 2(a), comprising two different sites denoted as X and Y in red and blue, respectively. The X and Y sites are populated by elements from the red and blue regions of the periodic table, respectively, as illustrated in Figure 2(b). X sites are populated by one or two elements selected from {Li, Mg, Ti, V, Mn, Fe, Co, Ni, Cu, Ru, Rh, Pd, Ag, Cd, Ir, Pt, and Au}, while Y sites are populated by one or two elements selected from {Li, Be, B, Mg, Al, Si, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, Ge, As, Y, Zr, Nb, Mo, Ru, Ag, In, Sn, Sb, Hf, Ta, W, Pt, and Pb}. One of these sites is disordered and contains two elements, whereas the other site contains one element. Consequently, the ternary alloys are represented as $X^1_{1-x}X^2_xY_1$ and $X_1Y^1_{1-x}Y^2_x$, where x denotes the composition in 0.2 increments. For instance, when two elements (e.g. Fe and Rh) are selected and disordered for the X site, only one element (e.g. Al) is selected for the Y site, the resulting ternary alloys are represented as $Fe_{1-x}Rh_xAl$. This study only considers ternary alloys containing either Fe, Co, or Ni elements and thus comprises a dataset of approximately 17,000 magnetic ternary alloys.

The magnetic moment M, spin polarization S, and Curie temperature T_c were obtained through density functional theory (DFT) calculations based on Green's function. For this purpose, we used the Korringa – Kohn–Rostoker coherent-potential approximation (KKR-CPA) method implemented in the AkaiKKR software [19–24]. Additional information regarding the dataset and DFT calculations can be found in Supplemental Information S3.

For material descriptors, we used molar ratios to represent the combination of elements and their amounts. To provide more detailed material description, we adopted the elemental fingerprints proposed by Hwang et al. [25]. The resulting 123 material descriptors and their corresponding details are listed in Supplemental Information S4.

To simulate a scenario where there are abundant M and S data but no T_c data in the database, we conducted an autonomous material search. Our aim was to maximize the T_c as the target property, while leveraging information on the nontarget properties M and S.

4. Results and discussion

We conducted several autonomous material searches to optimize the T_c. Figure 3(a) shows the results of one such search, which aimed to maximize the T_c using an ENN model without transfer learning (model N). The blue dots indicate the T_c of the material found for each search iteration, whereas the thick black line represents the maximum T_c achieved thus far. As the number of search iterations increased, the training data and accuracy of the ENN model improved, allowing the discovery of materials with a higher T_c.

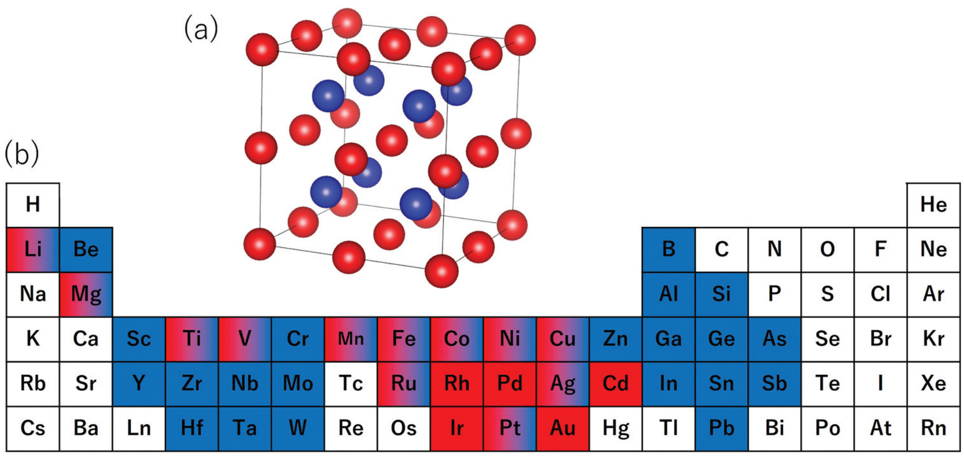

Figure 2. Material data used in this demonstration. (a) The B2 crystal structure, which comprises two different sites denoted as X and Y in red and blue, respectively. (b) A periodic table representing the elements of X and Y sites. One of these sites is disordered and contains two elements, whereas the other site remains fixed.

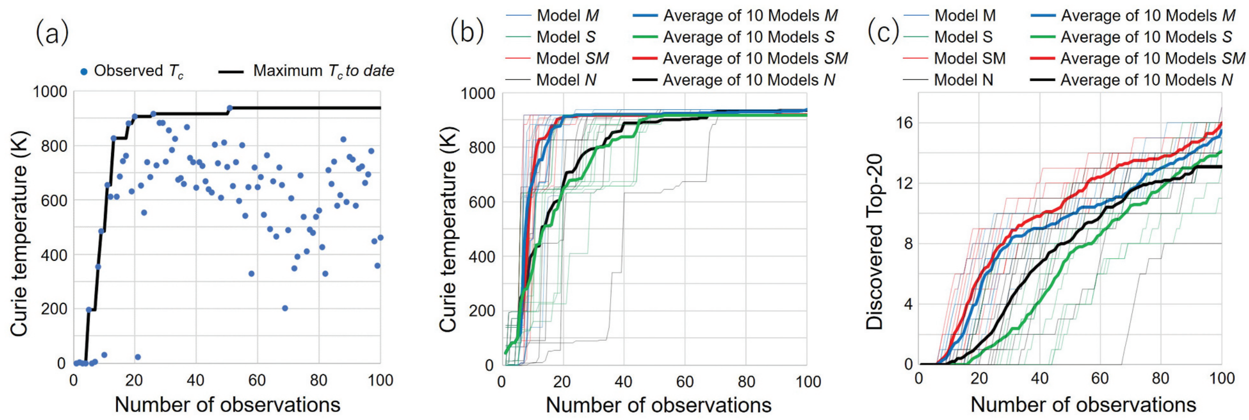

Figure 3. (a) The results of autonomous material search to maximize T_c using an ENN model without transfer learning (model *N*). The blue dots indicate the T_c of the material found for each search iteration, whereas the thick black line shows the maximum T_c achieved till date. (b) The maximum T_c achieved till date in four different autonomous material search cases (models *M*, *S*, *MS*, and *N*). The thick lines show the average of 10 results. (c) The count of the top 20 high T_c materials discovered by the four autonomous material searches.

Figure 3(b) shows the maximum T_c achieved till date in four different autonomous material search cases: ENN with transfer learning from *M* (model *M*), *S* (model *S*), and both *M* and *S* (model *SM*), and no transfer learning (model *N*). We conducted 10 searches each using models *M*, *S*, *SM*, and *N*, and the results are represented as thin blue, green, red, and black lines, respectively. The thick lines indicate the average of these 10 results. Models *M* and *SM* were able to discover materials with a higher T_c more rapidly than model *N*.

In material development, it is often advantageous to propose multiple high-performance materials rather than just a top one. Figure 3(c) shows the number of autonomous material searches versus the count of the top 20 high T_c materials discovered. On average, model *N* identified 4.2 high T_c (Top-20) candidate materials in 30 observations, whereas models *M* and *SM* found an average of 8.8 and 8.9 high T_c candidates in 30 observations. Our findings demonstrate that transfer learning, where nontarget property information is used, can result in efficient autonomous material searches. Additionally, this approach is especially useful when only a limited amount of target property data is available.

However, as shown in Figure 3(b,c), the discovery speed of model *S* was observed to be slower than that of model *N*, indicating that transfer learning is not always an efficient means for autonomous material search. To determine the optimal use of transfer learning in this context, we evaluated the predictive performance of the ENN model after transfer learning. Figure 4 shows the mean absolute error (MAE) as a function of observed data for the four transfer learning types, represented by the blue (*M*), green (*S*), red (*SM*), and black (*N*) lines, respectively.

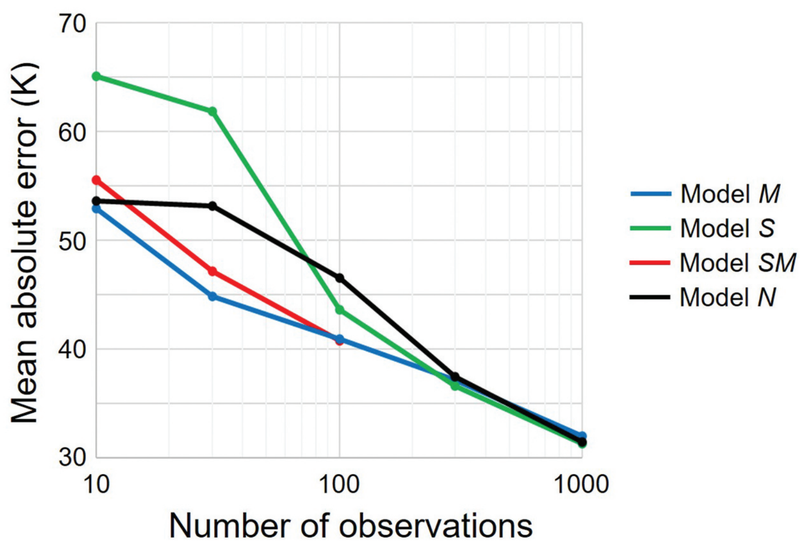

Figure 4. The mean absolute error (MAE) as a function of observed data for the four transfer learning types: models *M*, *S*, *SM*, and *N*. The predictive performance of model *S* is inferior to that of model *N* because of negative transfer.

We found that when the observed data are scarce, the predictive performances of models *M* and *SM* surpass that of model *N*, leading to an efficient autonomous material search, as shown in Figure 3(b,c). However, the predictive performance of model *S* is inferior to that of model *N*, and leveraging the source domain data/knowledge in transfer learning afforded a negative transfer, which decreases learning performance in the target domain [26]. As a result of low predictive performance owing to negative transfer, autonomous material search utilizing model *S* exhibited low efficiency.

To mitigate the potential for negative transfer during the autonomous materials search process, it is crucial to implement a verification step. Principally, there exist two strategies that can be employed for verification.

The first strategy relies on insights derived from materials science. This strategy essentially involves understanding the correlation between the target property and various nontarget properties from a materials science perspective, thereby enabling the selection of an appropriate transfer learning model. For instance, considering Curie temperature's minimal correlation with spin polarization – which reflects the electron states only near the Fermi energy – it becomes clear that model *S* can be excluded from the candidate list of transfer learning models for the autonomous materials search. However, it is important to note that such determinations are not always feasible from a materials science viewpoint.

In situations where the first strategy proves ineffective, we rely on the second strategy, grounded in data science. This involves the generation of multiple transfer learning models using data of various nontarget properties. Their predictive performances are then evaluated using the target property data, with the highest-performing model being selected for use in the autonomous materials search. In this case study, for instance, models *N, S, M*, and *SM* were constructed at each stage of the autonomous materials search. The model demonstrating the highest predictive performance with respect to the target property data (T_c) is selected in each iteration of the autonomous search. For instance, at the stage of the search process where the observed data was 30 as illustrated in Figure 4, model *M* —with the highest predictive performance – is selected. The dynamic selection of transfer learning models at each stage of the search process is anticipated to enhance the overall efficiency of the materials search.

5. Conclusions

We proposed a novel approach for autonomous material search. This method optimizes target properties while leveraging transfer learning to transfer information on nontarget properties. Our results demonstrate the effectiveness of this method in the search for materials with a high T_c while utilizing information on the *M* and *S* of ternary magnetic alloys. The use of transfer learning enabled an efficient autonomous search, particularly in the early stage of the search process. However, we observed instances where negative transfer negatively impacted the search efficiency, highlighting the need for the dynamic selection of models based on their performance. Overall, our approach is widely applicable to diverse properties and material systems and can serve as a foundation for accelerating material development.

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Disclosure statement

No potential conflict of interest was reported by the author(s).

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Data availability statement

The data that support the results reported herein and other findings of this study are available from the corresponding author upon reasonable request.

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