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Composition design for (PrNd–La–Ce)$_2$Fe$_{14}$B melt-spun magnets by machine learning technique*

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Data-driven technique is a powerful and efficient tool for guiding materials design, which could supply as an alternative to trial-and-error experiments. In order to accelerate composition design for low-cost rare-earth permanent magnets, an approach using composition to estimate coercivity (H_{cj}) and maximum magnetic energy product ($(BH)_{max}$) via machine learning has been applied to (PrNd–La–Ce)$_2$Fe$_{14}$B melt-spun magnets. A set of machine learning algorithms are employed to build property prediction models, in which the algorithm of Gradient Boosted Regression Trees is the best for predicting both H_{cj} and $(BH)_{max}$, with high accuracies of $R^2 = 0.88$ and 0.89, respectively. Using the best models, predicted datasets of H_{cj} or $(BH)_{max}$ in high-dimensional composition space can be constructed. Exploring these virtual datasets could provide efficient guidance for materials design, and facilitate the composition optimization of 2:14:1 structure melt-spun magnets. Combined with magnets' cost performance, the candidate cost-effective magnets with targeted properties can also be accurately and rapidly identified. Such data analytics, which involves property prediction and composition design, is of great time-saving and economical significance for the development and application of LaCe-containing melt-spun magnets.

Keywords: permanent magnet, materials design, machine learning, property prediction

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1. Introduction

Due to the excellent magnetic properties, $Nd_2Fe_{14}B$ magnets have been widely applied in sensors, motors, and generators since discovered in 1984.[1,2] With the ever-rising price of Nd element, substituting inexpensive rare-earth metals La and Ce for Nd in melt-spun magnets has recently drawn continuing interests.[3–6] Although the cost of magnets can be reduced, their magnetic properties (coercivity H_{cj} and maximum magnetic energy product $(BH)_{max}$) decrease simultaneously as La or Ce content increases.[7] In fact, the magnets would be unable to satisfy desired application requirement while the replacement of La or Ce exceeding a certain threshold. Thus, optimizing materials composition to balance the trade-off between cost and performance in LaCe-containing melt-spun magnets is crucially important to their further development and application. Traditionally, Nd–Fe–B typed magnets design has been guided by human intuition or trial-and-error experiments. With increasing chemical complexity due to the introduction of La or Ce, the number of potential compositions with targeted properties to explore is too large for the traditional method to be practical. Intuition would cause great composition prediction errors and become unreliable. Experiments, on the other hand, would suffer from lengthy and laborious Edisonian synthesis-test cycles. Hence, an approach that allows us to design cost-effective LaCe-containing magnets in an accurate, fast and economical manner is highly desirable.

Fortunately, the successful application of data-driven techniques in multicomponent materials provides an inspiration for the materials design with such requirements.[8–13] Using machine learning tools, a predictive model connecting composition with property can be constructed by the statistical rules and inferences learnt from massive experimental data. It offers an opportunity to navigate high-dimensional composition space for targeted property, and provides clear guidance for materials design. For example, Austin *et al.*[11] built a model to predict ionic conductivity of $Li_xA_aB_bC_c$ solid lithium-ion conductor sorting out the best 21 compounds with high cost performance from 12831 candidates. Xue *et al.*[12] captured the relationship between composition and thermal hysteresis (ΔT) in $Ni_{50-x-y-z}Ti_{50}Cu_xFe_yPd_z$ shape memory alloy discovering 14 new compounds having $\Delta T < 3.15$ K from a potential space of $\sim 8 \times 10^5$ compositions. However, such efficient data analytics has not been reported on the materials design of permanent magnets. Here, we target this question and focus on developing a predictive data-driven capability to accelerate composition design for (PrNd–La–Ce)$_2$Fe$_{14}$B melt-spun magnets.

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In the present work, four machine learning techniques are attempted to build predictive models by deciphering magnets nonlinear composition-property (H_{cj} or $(BH)_{max}$) relationships. After model evaluation and ranking, the model with the highest accuracy is selected for future property prediction and composition design. Furthermore, we illustrate how to design the low-cost magnets with targeted properties in a composition space with two concerned dimensions: total rare-earth content and Ce content. These researches would serve as a valuable reference for the design of LaCe-containing magnets with good cost-effectiveness.

2. Data-driven method

Figure 1 depicts our overall dataset preparation, model training, model evaluation, and best-model prediction scheme. Firstly, a dataset of 2:14:1 structure melt-spun magnets containing the information of materials composition and magnetic properties is built. Each magnet data is labelled via material descriptors or features X (composition here) and a specific objective y (H_{cj} or $(BH)_{max}$ here). Next, considering that learning algorithms exhibit various advantages on different datasets due to their respective characteristics, four regression algorithms are employed to do data learning in this work: Linear Regression, Decision Trees Regression[14] (DTR), Support Vector Regression with a radial basis function kernel[15] (SVR.rbf), and Gradient Boosted Regression Trees[16] (GBRT). During the model training process, the algorithm parameters, such as the decision tree depth and the minimum number of data required to split a node in DTR, the penalty factor and the kernel function coefficient in SVR.rbf, and the learning rate and the number of boosting stages in GBR, are adjusted to optimize the model performance by the grid-search method.[17] All constructed models are evaluated using k-fold Cross-Validation (CV) with respect to a performance metric of R^2 for predictive accuracy. R^2 represents the variance explained by the model (higher the better), whose maximal value is equal to 1. After comparing their predictive capabilities, the model with the highest R^2 value is defined as the best predictive model. Finally, the best model is used to estimate magnets properties from the inputs of composition. In turn, these virtual properties data can be applied to guide magnets composition design. Machine learning modeling and predictive analytics are implemented in Python 3.6 with the scikit-learn open-source package.[18]

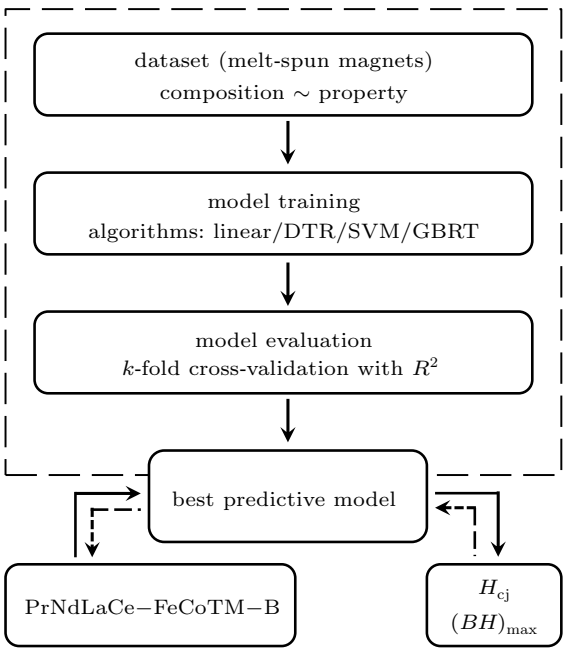

Fig. 1. Schematic diagram of our data-driven approach to property prediction and composition design of 2:14:1 structure melt-spun magnets using machine learning technique.

3. Results and discussion

The dataset of 2:14:1 structure melt-spun magnets collected in this work contains 413 data from the published literatures and the experiments[5,6,19] in our laboratory. There are 204 stoichiometric and rich-rare-earth magnets (single-phase magnet), including 149 La/Ce-containing ones, and 209 lean-rare-earth magnets (composite magnet), among which 40 are La/Ce-replaced ones. The data are preprocessed for consistency using the chemical composition with a form of $(PrNd_{PrNd\_p}La_{La\_p}Ce_{Ce\_p})_{REs}Fe_{Fe}Co_{Co}TM_{TM}B_B$, where PrNd_p + La_p + Ce_p = 1, REs + Fe + Co + TM + B = 100, and TM represents the sum of Zr, Nb, Ga or Ti. The material feature vector X = (REs, PrNd_p, La_p, Ce_p, Fe, Co, TM, B). Figure 2 shows the Pearson correlation coefficients between the magnetic properties (H_{cj} or $(BH)_{max}$) of magnets and each variable in X. Pearson correlation coefficient is a statistic measure of the linear correlation between two variables x_i and y_i. It has a value between +1 and −1, where +1 is total positive linear correlation, 0 is no linear correlation, and −1 is total negative linear correlation.[20] Here the relative magnitude of the coefficient indicates the chemical element's importance to H_{cj} or $(BH)_{max}$. Each element in the composition has its physical basis in terms of their influence on magnets properties, which agrees with the prior knowledge of rare-earth permanent magnets:

(i) The increase of rare-earth content is accompanied by the decrease of iron content, which results in the weakening of intergranular interaction and the decline of saturation magnetization (M_s). Correspondingly, H_{cj} of magnets increases while $(BH)_{max}$ decreases. Therefore, H_{cj} and $(BH)_{max}$ have a positive (or negative) and a negative (or positive) correlation with the sum of rare-earth (or Fe content), respectively.

(ii) Because both magnetocrystalline anisotropy field and M_s of $La_2Fe_{14}B$ and $Ce_2Fe_{14}B$ are lower than those of $Nd_2Fe_{14}B$, the substitution of La or Ce would reduce H_{cj} and

$(BH)_{max}$ of magnets. Both H_{cj} and $(BH)_{max}$ thus show positive correlations with Nd content but negative correlations with La/Ce content.

(iii) Adding a small amount of Co, Zr, Nb, Ga, and Ti in magnets can not only refine grain or optimize grain boundary, that is beneficial to the improvement of H_{cj}, but also slightly increase M_s or enhance the squareness of the demagnetization curve, thereby resulting in the improvement of $(BH)_{max}$. So both H_{cj} and $(BH)_{max}$ are positively correlated with Co or TM content.

(iv) Because rich-B magnets contain much non-magnetic phases, which leads to the decrease of M_s, $(BH)_{max}$ shows a negative correlation with the content of B. However, the positive or negative effect of B on H_{cj} depends on the position of non-magnetic phases in the microstructure of magnets. Those non-magnetic phases may act as pinning points that hinder the domain wall movement increasing H_{cj} or as nucleation points of the reversal magnetic domains reducing H_{cj}. Thus, the statistical correlation between H_{cj} and B content is almost equal to zero.

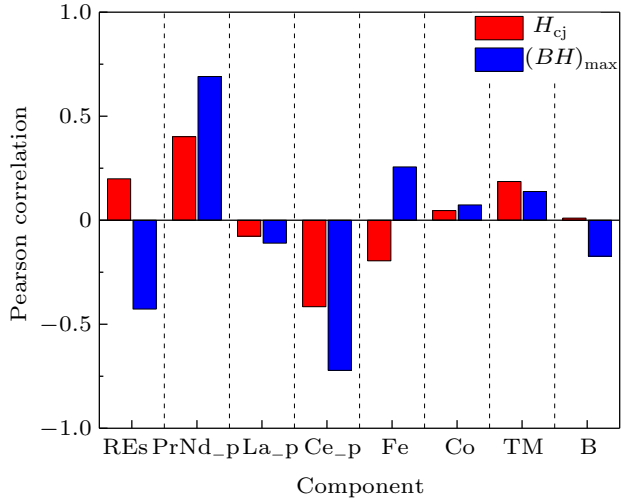

Fig. 2. Pearson correlation coefficients between H_{cj} or $(BH)_{max}$ and all features in $(PrNd–La–Ce)_2(Fe\text{-}Co\text{-}TM)_{14}B$ melt-spun magnets.

Not too high correlation coefficients in Fig. 2 imply that there are complex nonlinear relationships between composition and magnetic properties in 2:14:1 structure melt-spun magnets. To decipher these relationships, an appropriate fitting model f can be learnt from the data to map the features X to one target y, i.e., $y = f(X)$. In turn, it can be used to predict y for unexplored X. Before modeling, dataset is divided into two parts. One part is used to train the model, and the other is used to test quality of the model. Our machine learning algorithms are trained and 5-fold cross-validated using a training set containing 80% of our data. As shown in Fig. 3, R^2-CV of predictive models for H_{cj} based on Linear, DTR, SVR, and GBRT is 0.71, 0.69, 0.82, and 0.88, respectively. Correspondingly, R^2-CV of predictive models for $(BH)_{max}$ is 0.74, 0.70, 0.84, and 0.89, respectively. It can be seen that the models estimating H_{cj} or $(BH)_{max}$ using GBRT all exhibit the highest scores, which are then thinked as the best models. Next, the predicted accuracies of these two models need to be tested to determine whether they are feasible. A simple way to do this is to use an untrained data for evaluation. Figure 4 displays the predicted results from the four models on the test set with the remaining 20% of our data. If the model is perfect, the predicted H_{cj} or $(BH)_{max}$ will be exactly the same as the measured one and all data points will align along the 45° diagonal line. R^2 of H_{cj} and $(BH)_{max}$ in Fig. 4 are close to those on the training set (shown in Fig. 3), respectively. This indicates that all the models have good generalization abilities. Compared with the other three algorithms, the data based on GBRT show the highest convergence along the 45° diagonal line, and mean absolute errors (MAE) of H_{cj} and $(BH)_{max}$ are the smallest, with the value of 1.04 kOe (1 Oe = 79.5775 $A{\cdot}m^{-1}$) and 1.13 MGOe, respectively. It is reasonable to believe that such errors are comparable to that caused by preparation process and absolutely far less than that from human intuition. Thus, they are acceptable to the performance estimation of LaCe-containing melt-spun magnets.

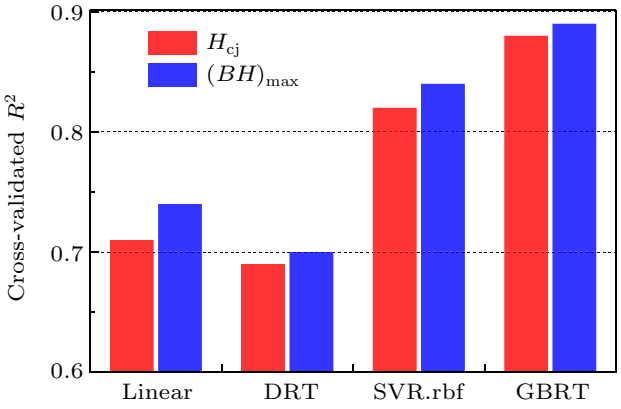

Fig. 3. Performance (R^2) of different predictive models for H_{cj} or $(BH)_{max}$ on the training dataset. For each model, a 5-fold cross-validation is applied.

Now two accurate and robust prediction models (GBRT) for H_{cj} and $(BH)_{max}$ have been constructed. They are capable of building a property prediction dataset in an arbitrary rational composition space with one or more dimensions, such as the variables in X. Figure 5 presents the predicted maps of H_{cj} and $(BH)_{max}$ for $(PrNd_{1-Ce\_p}Ce_{Ce\_P})_{REs}Fe_{93-REs}TM_1B_6$ melt-spun magnets and their corresponding performance-cost ratio contours. (The cost of magnets is calculated merely by rare-earth elements because of their high prices. As of December 2017, the prices of Ce and Nd are approximately 5.6 $/kg and 72.7 $/kg, respectively.) From Fig. 5, we can clearly observe the changes of magnetic properties with the sum of rare-earth as well as the content of Ce. These virtual data in this space help to quickly locate composition regions with targeted

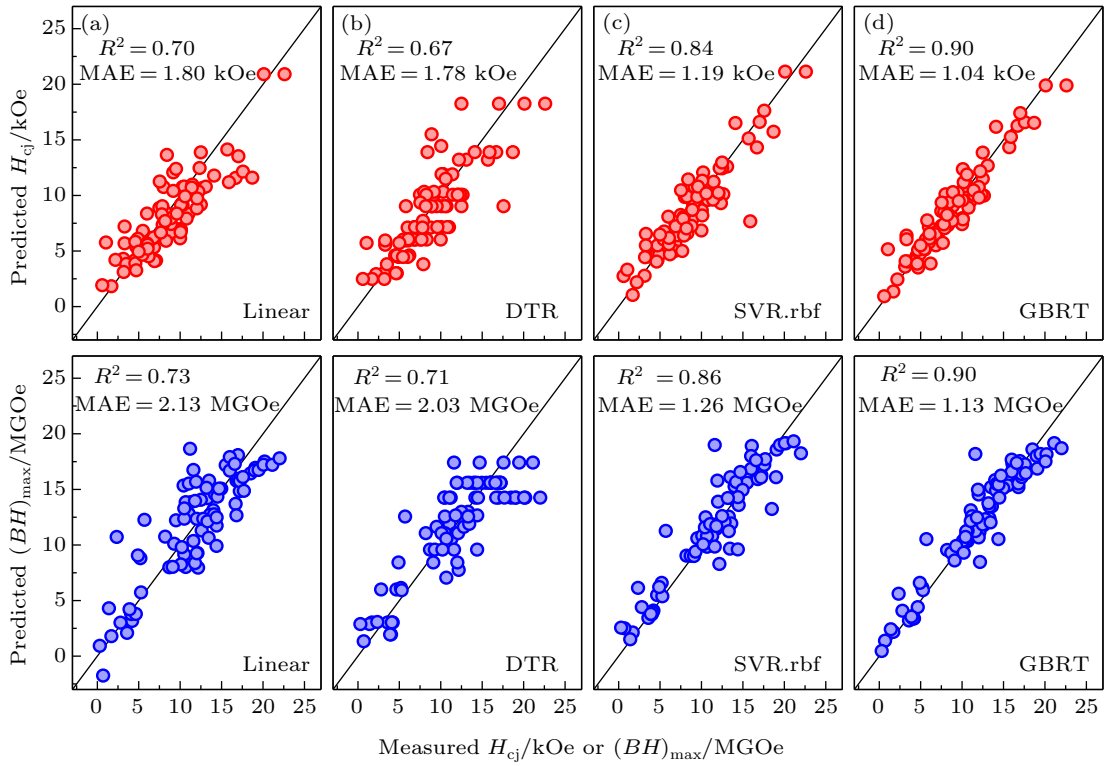

Fig. 4. Predicted scatter plots on the test dataset by the four modeling techniques, (a) linear regression, (b) Decision Trees Regression, (c) Support Vector Regression with a radial basis function kernel, and (d) Gradient Boosted Regression Trees. *X* axis and *Y* axis denote the measured and predicted values, respectively.

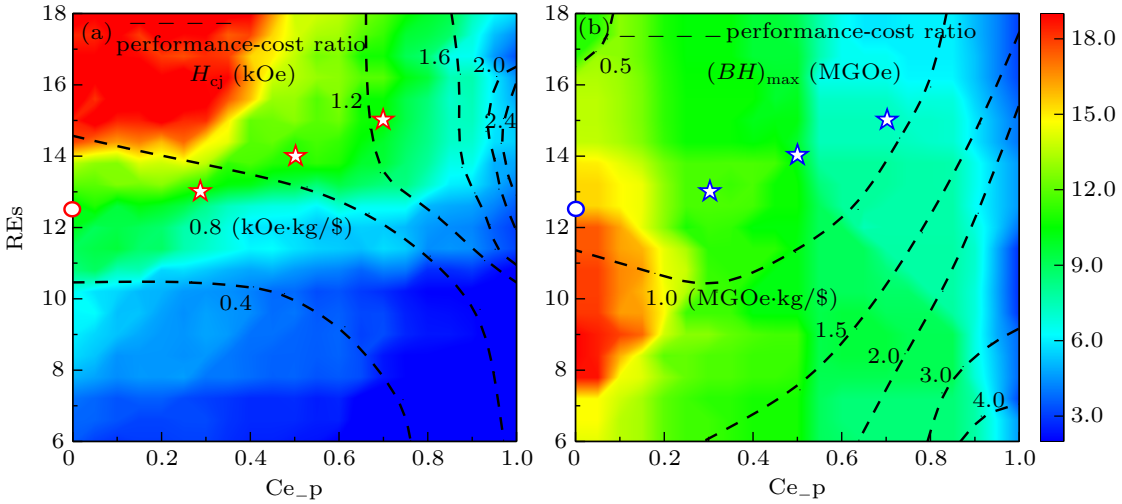

Fig. 5. Predicted maps of (a) H_{cj} and (b) $(BH)_{max}$ using the best models (GBRT) for $(PrNd_{1-Ce\_p}Ce_{Ce\_p})_{REs}Fe_{93-REs}TM_1B_6$ melt-spun magnets, and their corresponding cost performances. Circle represents a pure Nd magnet with H_{cj} about 11.0 kOe, which can be replaced by the economical Ce-containing magnets with the same H_{cj} at the expense of $(BH)_{max}$ (Star).

H_{cj} or $(BH)_{max}$. Considering the cost of materials, inexpensive small composition space with the same properties can also be easily sought out. For example, the melt-spun magnet powders with a stoichiometry of $Nd_{12.5}Fe_{80.5}TM_1B_6$($H_{cj}$ =∼ 11.0 kOe) are often used to prepare bonded magnets. From the economical perspective, this kind of magnets can be replaced by the new Ce-containing magnets with the same H_{cj}. In order to avoid the decrease of H_{cj} after adding Ce element, empirical guides tell us that we can increase total rare-earth content to compensate for the loss of H_{cj} at the expense of $(BH)_{max}$, but it is difficult to accurately determine the composition that is needed. However, the small composition regions of H_{cj} = ∼ 11.0 kOe, e.g., centered on $(Nd_{0.7}Ce_{0.3})_{13}Fe_{80}TM_1B_6$, $(Nd_{0.5}Ce_{0.5})_{14}Fe_{79}TM_1B_6$, or $(Nd_{0.3}Ce_{0.7})_{15}Fe_{78}TM_1B_6$, can be directly located from Fig. 5(a), without requiring experimental guess-and-check in such a two-dimensional space. On the whole, this data analytics can significantly reduce the time and cost of materials design. It will be an efficient tool for promoting the development of 2:14:1 structure melt-spun magnets.

4. Conclusion

In summary, we have demonstrated a machine learning approach that involves property prediction to accelerate composition design of (PrNd–La–Ce)$_2$Fe$_{14}$B melt-spun magnets. The model constructed via the Gradient Boosted Regression Trees algorithm performs a powerful predictive capability for both H_{cj} and $(BH)_{max}$ of magnets. Exploring the virtual composition space by the model, cost-effective magnets can be accurately and quickly identified according to different performance requirements.

Besides the composition of magnets, some microstructural parameters (e.g., grain size or alignment degree) greatly affecting H_{cj} or $(BH)_{max}$ should be added as material features to enhance models' predicted accuracies. But because melt-spun magnets prepared under the optimum conditions reported usually exhibit almost the same microstructure, and the differences in grain size or orientation degree are very small, these material features are neglected while modeling. That is why such high predicted accuracies can be obtained in this work just by the composition of magnets. However, for the magnets using other technologies, like hot-deformed or sintered magnets, they form a variety of microstructure as composition or preparation process changes, and would present more complex "property-composition-structure-process" relationships. In this regard, our follow-up work is focus on the feature engineering and to collect microstructural or process parameters of hot-formed and sintered magnets, and eventually to build high-performance property prediction models of 2:14:1 structure permanent magnets.

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