Machine Learning Technique for Prediction of Magnetocaloric Effect in Rare Earth-based Amorphous Alloys

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Abstract: The magnetic refrigeration technology based on the magnetocaloric effect (MCE) has been considered as a more advantageous technique over present well-used conventional gas compression. However, the composition of a vast number of magnetic materials with large MCE are still remained unknown. On the other hand, data-mining techniques using machine learning are found really efficient in many fields, especially for materials design. So here, due to the distinctive advantages of amorphous alloy over crystalline materials and the intrinsic large MCE of heavy rare earth-based alloys, the system of rare earth-based amorphous alloy was selected to study by this technique. By using a machine learning algorithm called gradient boosting regression tree (GBRT) and putting the external magnetic field into input features together with the composition, two models were successfully built to predict Curie temperature (TC) and magnetic entropy change (ΔSM) with high accuracy. The performance metric coefficient scores of determination (R2) of the two models are 0.96 and 0.91. Finally, because of the error that exists in the ΔSM itself, a new standard was presented to clearly see if the data of ΔSM predicted by us can be accepted. The success of the two models building and their excellent generalization ability suggest that they will be really helpful for our experiments guided and find proper composition for further magnetic refrigeration applications.

1. Introduction

Currently, vapor compression refrigeration technology is the leading refrigeration technology. Although the technology has a wide application, many devices have relatively low Carnot efficiency (often less than 25%), and environmental concerns associated with leakage of the gaseous refrigerant. Increasingly, researchers are exploring alternative cooling technologies that may be more environmentally friendly and energy efficient that can be commercialized in the near future [1-5]. The magnetic refrigeration technology based on the magnetocaloric effect(MCE) has been considered as a more advantageous technique over present well-used conventional gas compression [6,7]. However, the magnetic refrigerators are in the laboratory development stages, and the researchers are still working to search magnetic solids with better MCE performances.

According to the existing knowledge, all the magnetic solids exhibit MCE which is the process of the entropy or the temperature change with respect to the changing of external magnetic field, and the intensity is strongly correlated to their own magnetism and magnetic phase transition [8]. And the key of the development of magnetic refrigeration technology is to find out magnetic refrigeration materials with excellent MCE which is normally correlated with Curie temperature(TC),magnetic entropy change(Δ SM) and some other parameters. So the TC of an excellent magnetic refrigeration material should be in a proper range while the ΔSM should be large enough.

In the exploration and study of magnetic refrigeration material, heavy rare earth-based alloys have always been the focus of research on magnetic refrigeration materials. What's more, amorphous alloy as magnetic refrigeration material has many advantages over crystalline materials. In recent years, magnetic amorphous alloys have attracted more attention due to their large ΔSM over a wide temperature range [9]. On the other hand, the refrigerants are classified as having either a first-order phase transition (FOPT) or second-order phase transition (SOPT). An SOPT magnetic material undergoes a magnetic phase change which has no thermal or magnetic hysteresis, making it suitable for fabricating magnetic refrigeration systems [10,11]. So, the totally disordered structure and second order magnetic transition nature endow amorphous alloys with several advantages over crystalline materials as magnetic refrigerant [12]. Therefore, combining the two systems above, it will be a great breakthrough to the development of magnetic refrigeration technology if the composition of materials with excellent MCE in the system of rare earth-based amorphous alloy can be found. However, predicting the parameters of MCE in the complex system is really tough. Fortunately, data-mining techniques using machine learning can be applied to build models for the prediction of the relationship between physical features with composition-related parameters that can be obtained prior to experiment.

Machine learning is an interdisciplinary that has been widely used in many fields in recent decades and is a field of computer science that provides computer system with the ability to 'learn' and is a branch of artificial intelligence. It is an efficient method to analyze massive and complex data which is perfectly suitable for materials design. To date, few people have made a try to use machine learning studying magnetic refrigeration materials. But materials design by using machine learning has been successfully tried by several research groups. For example, Oliynyk et al [13] made a model that can distinguish Heusler and Heusler alloys by machine learning with existing data. Xue et al [14] used machine learning to model NiTi-based shape memory alloy by self-adaptive method and 36 materials with small thermal hysteresis were predicted by the model from 800000 potential composition combinations at last. Yuan et al [15] used uncertainty of prediction results to design experiments by machine learning algorithm with improved optimization algorithm and a lead-free piezoelectric material with high electric strain was successfully developed by only 3 groups of experiments. And in the MCE field, HoB2 with gigantic MCE was singled out and synthesized through experiments guided by machine learning [16].

Up to now, data-mining techniques using machine learning have not been reported in the materials design for rare earth-based amorphous alloys. Thus in this paper, we choose the system of rare earth-based amorphous alloy to study and successfully built a model by using a machine learning algorithm called gradient boost regression tree (GBRT) and putting the external magnetic field into the input features at the same time to predict TC and ΔSM of rare earth-based amorphous alloys. Finally, due to the error that exists in the ΔSM itself, a new evaluation standard of the model which is much clearer is also presented. The work that successfully predicts the MCE of rare earth-based amorphous alloy will be of great significance to the development of magnetic refrigeration technology.

2. Data

The Materials' data are all collected from the published literatures. Considering that the data are from the paper published, they are usually reliable enough. Our dataset is the system of the rare earth-based amorphous alloys and has 252 pieces of data [17]. As the rare earth based amorphous alloys from the data contain 29 elements in total, we selected all the 29 elements as the input features, in which the values are the ratio of the alloys' chemical composition and it means that the alloy composition doesn't have the element if the value is 0. And the target properties are TC and ΔSM. In our model, we chose the opposite of ΔSM which is (-ΔSM) for all of the data of ΔSM collected by us were negative and this can simplify the calculations in our model. As amorphous ferromagnetic magnetothermal effect materials only have a SOPT, we chose the phase transition temperature of the transition of the materials from ferromagnetic state into paramagnetic state as TC. And since the values of external applied magnetic field in the data collected are distributed in the range of 1 T-9 T and the fact that most magnetothermal materials' ΔSM will change obviously with the change of the external magnetic field applied, one can jump to a conclusion that the external magnetic field may be strongly correlated with TC and ΔSM. Due to the correlation above, if the magnetic field is used as an input feature together with the chemical composition to predict TC and (-ΔSM), the accuracy of the model must be really high. Therefore, we added the external magnetic field into the input features with the 29 elements together aiming to optimize the model so that it can be put into practical application to find the composition of rare earth-based amorphous alloys with outstanding magnetocaloric effect.

3. Method

The process of the method using machine learning to study the MCE materials is shown in Figure 1. The whole process mainly contains four parts, which are dataset's collecting and splitting, model training, model evaluation and model prediction. The dataset's collection has been discussed above, which is for the system of rare earth-based amorphous alloys. After collecting the data, they should be split up into two parts, one for training the model which is called the training set and one for evaluating the model after being trained which is called the test set. Normally, the whole data's 80% is selected for the training set and the other 20% for the test set. To make sure that the model's training effect and generalization ability will be better, the process of the selection must be random.

When all of the two datasets have been prepared, then the model should be trained and evaluated. The first step is to train the model by using the training set. As for the algorithm of the model, the task we faced in supervised learning of traditional machine learning is a regression problem and the algorithms that can handle this task mainly contain linear regression, decision tree regression, support vector regression, and some ensemble methods. The algorithm we used here is GBRT, which belongs to the ensemble methods. The GBRT algorithm produces a prediction models, which is an ensemble of regression tree models. The algorithm trains weak regression tree models once to make a weak prediction each time. Although the prediction errors are large at first, the algorithm will iterate training new weak regression trees to make the prediction better. Each new tree helps to correct errors made by the previously trained trees. Boosting is based on the idea of whether a weak model can be modified to become better. There are two important parameters in the GBRT algorithm: one is the number of estimators, which means the number of weak regression tree models; the other is the learning rate, which means the speed of improvement during iterating training weak regression trees. The GBRT algorithm is considered to be one of the best methods in machine learning. It can fit data where the relationship between features and target properties is complex well and perform robustly when facing outliers.

After building the GBRT model, the best parameters of the model should be searched for by the grid-search method [18]. The performance metric is R2 for the model prediction of training set after using k-fold cross-validation. R2 represents the variance explained by the model (the higher, the better), and its maximal value is equal to 1.

Once finishing the model training on the training set, the model's predictive performances should be evaluated on the test set. The metrics are R2 and MAE (mean absolute error: the lower, the better). And although the equation of ΔSM has been defined, the numerical integration involved in evaluating the total entropy functions may result in the accumulation of errors that can reach 20-30% of the calculated ΔSM values [19]. So when predicting the test set, if the error of the values of the predicted (-ΔSM) from the values of the (-ΔSM) from the dataset collected is less than 20-30%, we can say that the predicted values can be accepted by us.

Finally, the model can be used to estimate target properties for new materials via inputs of chemical composition and external magnetic field, which can serve as a quick guide to composition design and help find proper materials.

In this paper, all of the models' setup and analysis are implemented in Python 3.8 with the scikit-learn open-source package.

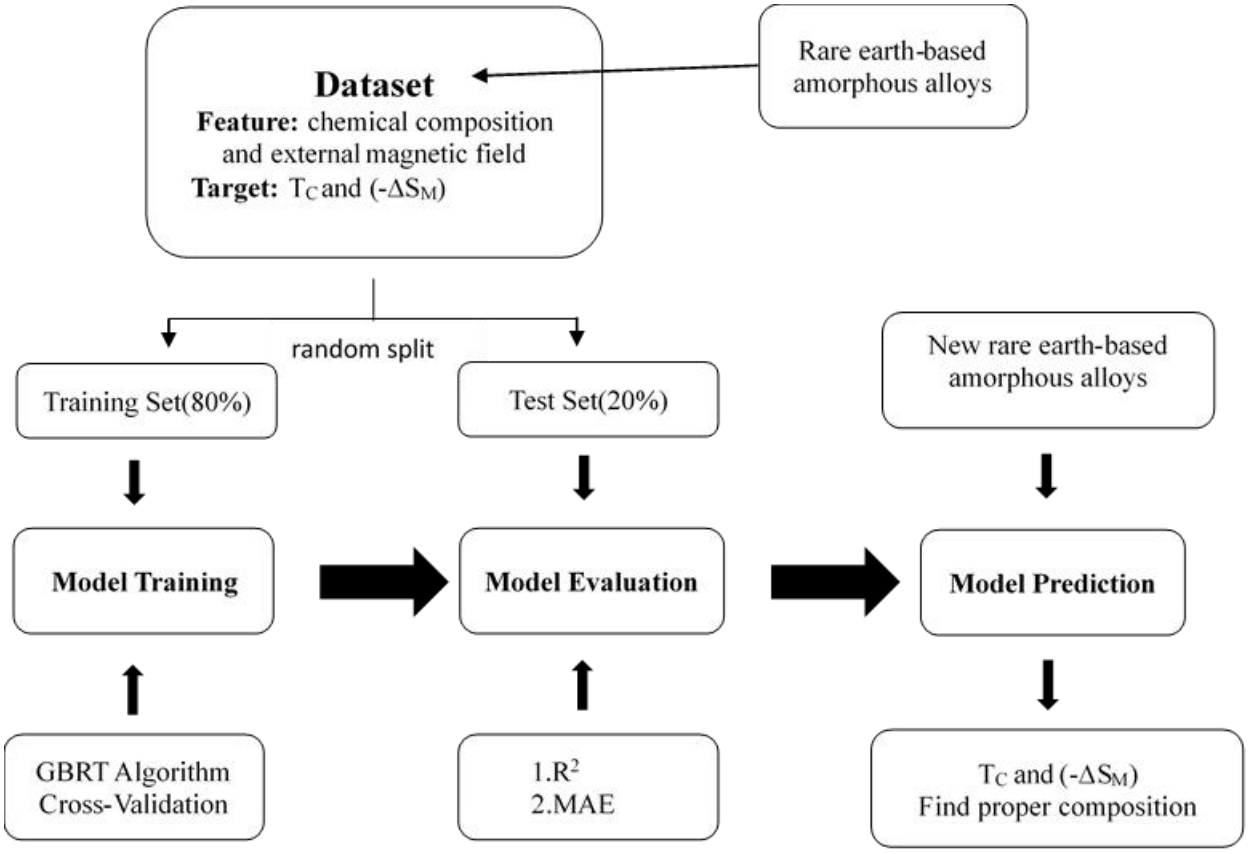


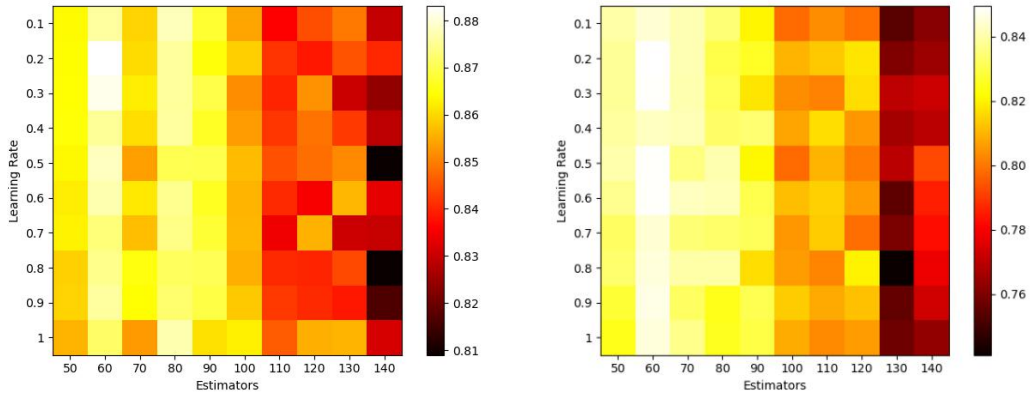
Figure 1: The flow chart of the process of the method using machine learning to study the rare earth based amorphous alloy materials.

4. Result and Discussion

The number of the data points collected by us to train the TC and (-ΔSM) models are 252 in total.

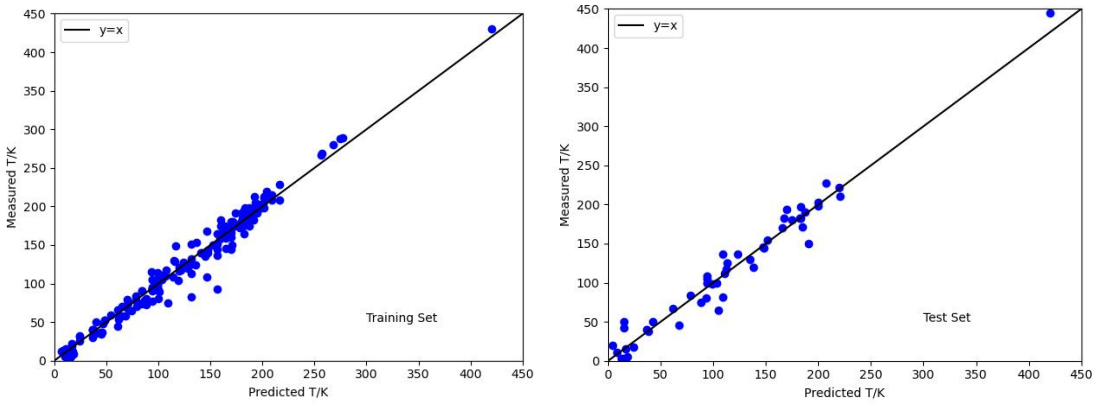
We randomly split the dataset between the training set (80%) and the test set (20%) and used the training set to train the model we built. Due to the relatively small data, fivefold cross-validation is

used for training the model. And we use grid-search for finding the best values of parameters (number of estimators and learning rate) of the model which make up the best R2 for the model as the best prediction model. During the procedure, we chose learning rate from 0.1 – 1 and number of estimators from 50 – 140, respectively, for all models. The results are depicted in Figure2 (a) and (b). We trained 100 combinations of two parameters model and found that the best combination for both of TC and (-ΔSM) is the learning rate of 0.2 with the number of estimators of 60.



(a) (b)

Figure 2: (a) and (b) Performances of TC and (-ΔSM) prediction models trained with different parameter combinations. The best parameter combination is marked with red star.



(a) (b)

Figure 3: Predicted scatter plots on TC dataset. (a) Training set and (b) test set.

After the best model is trained, it should be evaluated that if it is accurate enough. The common way is to use the trained model to predict the test set and evaluate how the model perform in the test set that it has never seen before. The metrics we used to evaluate are R2 and MAE. The results are depicted in Figure 3 for TC and Figure 4 for (-ΔSM). The X axis is the predicted results from the model and the Y axis is the measured data from our dataset. The perfect model is the one that the predicted results are all the same with the measured data and all data points will align along the 45° diagonal line. What's more, in Figure 4 (c) and (d), the red line is the data which are 20% error from the measured ones while the green line is the data which are 30% error. If the data points are located between the two green lines, we can say that those data of the values of the predicted (-Δ SM) can be accepted by us while if most of the data points are located between the two red lines, we can say that the accuracy of our model is really high.

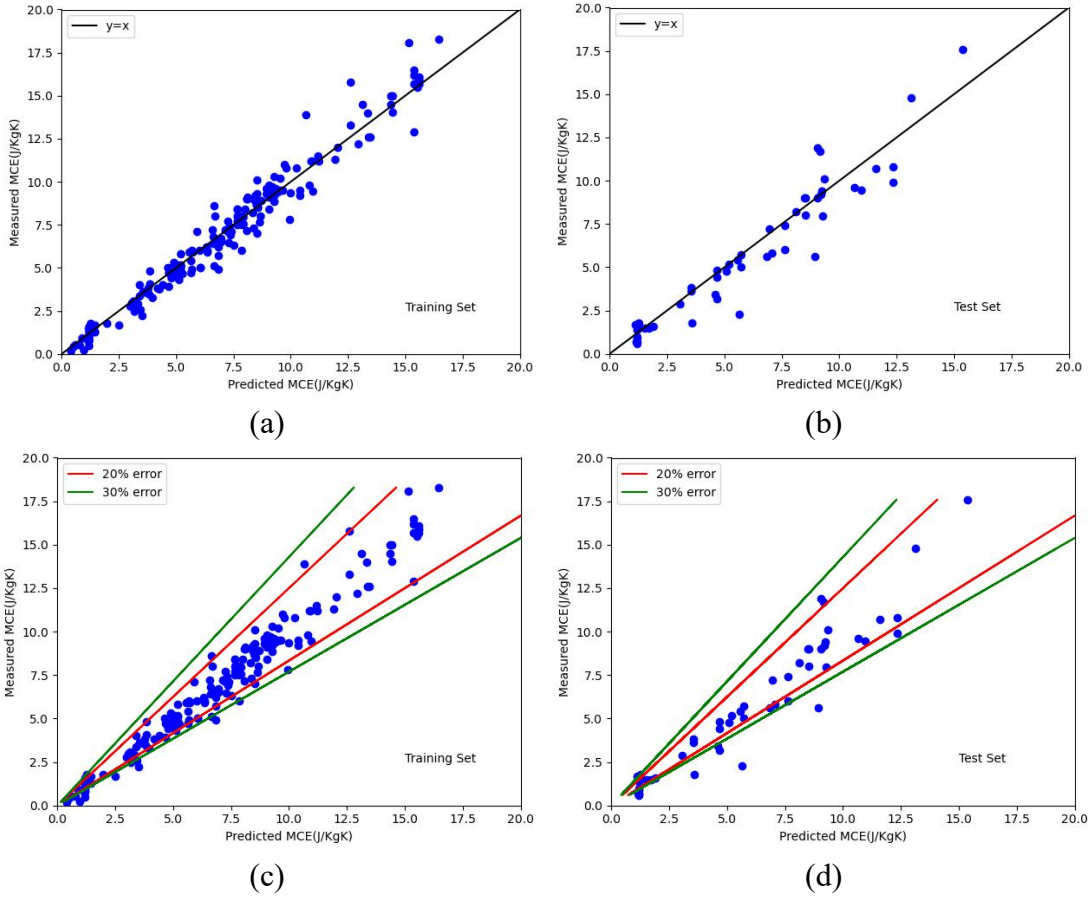


Figure 4: Predicted scatter plots on (-SM) dataset. (a) and (b) Training set and test set for the metrics R2 and MAE. (c) and (d) Training set and test set for the error line to see if the predicted MCE can be accepted.

The results of the prediction are shown in Figure 3 for TC and in Figure 4 for (-ΔSM). In Figure 3 (a), the R2 is 0.97 and the MAE is 8.75 K for the training set which shows that the model had trained perfectly on the training set. And the result of the evaluation is shown in Figure 3 (b) whose R2 is 0.96 and MAE is 12.54 K for the test set. The result indicates that the model has good generalization ability for untrained data. Although the result of the training set is slightly higher than that of the test set which means it is a little bit overfit (a phenomenon that the model prediction corresponds too closely or exactly for the training set and may result in failing to fit additional data reliably), in principle it is unavoidable due to the relatively small dataset. Overall, the model for TC has a great generalization ability.

As for the results for (-ΔSM) in Figure 4 (a) and (b), the R2 is 0.97 and the MAE is 0.53 J/Kg • K for the training set while the R2 is 0.91 and the MAE is 0.84 J/Kg • K for the test set, which also implies a little overfit that is unavoidable and good generalization ability. What's more, as we can see in (c) and (d) of Figure 4, only 5 data points are out of the two green lines for both the training set and the test set which means that all of the data predicted by us in addition to the 10 data in the two datasets can be accepted by us. On the other hand, most data points that are obviously between the two red lines can also show the high accuracy of the prediction of our model. One can find that when using the error lines to evaluate, the model performs nearly the same on the training set and the test set, which implies a powerful generalization ability of the model in another way.

Both of the two models we have built have good predictive performance. Only paying attention to the two metrics of R2 and MAE, the model of the prediction of TC performs obviously better than that of (-ΔSM). Since the dataset of the two models are all the same, the mainly reason of the difference of the performance must be the target magnetic properties the models predict.

The Curie temperature, TC, is an intrinsic magnetic property for materials that is more strongly correlated with the chemical composition. However, ΔSM is a calculated value with accumulation errors [19] which is more sensitive to the detailed measurement methods and experiments. And due to the unavoidability of the error, we used four error lines to evaluate the model. In this method, nearly all of the values of the (-ΔSM) are accepted which means the model can predict the value of (-ΔSM) in high accuracy as well.

5. Conclusions

The MCE has been predicted in other systems, such as La(Fe,Si/Al)13-based materials [20]. But all of those systems are constrained in a formalized crystal structure. Our work has proved that the MCE of the materials can also be well predicted in the system of a disordered structure which are the amorphous alloys. What's more, after putting the external magnetic field into the features together with the chemical composition and using a new method proposed by us to evaluate the model which was built to predict the value of (-ΔSM), the accuracy of the prediction in our two models are higher than all of those models in other systems.

In addition, due to the unique superiority of the rare earth based amorphous alloys discussed above, the finding that the MCE of the rare earth based amorphous alloys can be accurately predicted by our method will be a breakthrough in the development of the MCE materials. When the number of the data becomes large enough, the composition of the rare earth based amorphous alloys with superior MCE can certainly found in the future.

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