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A STUDY ON THE OPTIMIZATION OF METALLOID CONTENTS OF Fe-Si-B-C BASED AMORPHOUS SOFT MAGNETIC MATERIALS USING ARTIFICIAL INTELLIGENCE METHOD

The soft magnetic properties of Fe-based amorphous alloys can be controlled by their compositions through alloy design. Experimental data on these alloys show some discrepancy, however, with predicted values. For further improvement of the soft magnetic properties, machine learning processes such as random forest regression, k-nearest neighbors regression and support vector regression can be helpful to optimize the composition. In this study, the random forest regression method was used to find the optimum compositions of Fe-Si-B-C alloys. As a result, the lowest coercivity was observed in $Fe_{80.5}Si_{3.63}B_{13.54}C_{2.33}$ at.% and the highest saturation magnetization was obtained $Fe_{81.83}Si_{3.63}B_{12.63}C_{1.91}$ at.% with R^2 values of 0.74 and 0.878, respectively.

Keywords: Fe-based amorphous; Soft magnetic properties; Artificial intelligence; Machine learning; Random forest regression

1. Introduction

Fe-Si electrical steels have been widely used in electrical machinery due to their excellent soft magnetic properties. However, the operation frequency of these steels is generally limited to under 400 Hz due to high eddy current loss [1-3]. In order to increase the operation frequency without loss of soft magnetic properties, Fe-based amorphous and nano-crystalline alloys have been developed and widely used in high frequency applications using a rapid solidification process at a high cooling rate (>10^4 K/s). To obtain an amorphous structure, various metalloids (i.e. boron, silicon), non-metallic elements (i.e. carbon, phosphorus) and transition metals (i.e. copper, niobium) are incorporated in Fe-based amorphous alloys [4,5]. In addition, the amorphous structure is known to enhance the soft magnetic properties such as the permeability and coercivity [6,7] but also reduce the saturation magnetization [4,6-8]. In response, the nano-crystalline structure has been widely used to increase the saturation magnetization, however, deteriorates the soft magnetic properties.

Among Fe-based nano-crystalline alloys, the Fe-Si-B-Nb-Cu alloy has been the most widely used in field [8-11]. This alloy shows a coercivity of 25 A/m or less but a saturation magnetization value of 150 emu/g which corresponds with magnetic flux density of 1.2 T. Due to this low flux density, there is strong demand to develop a new soft magnetic material with a saturation magnetization up to 170 emu/g and a coercivity of less than 30 A/m. This indicates that the amounts of metalloids should be controlled in the range of 15~20 at.% [12]. Previous studies revealed that soft magnetic properties in Fe-based amorphous alloys are very sensitively dependent on the amounts and ratios of metalloids and transition metals [4,8]. However, their effects on the soft magnetic properties have not been clarified. The machine learning process uses a large amount of experimental data to predict results based on the results derived from the training. The problem of the effectiveness of metalloid elements on soft magnetic properties will be solved by trained algorithm processes.

In this study, we investigated the correlation between compositions and soft magnetic properties through machine learning methods. The saturation magnetization and coercivity of Fe-Si-B-C amorphous alloys was predicted through random forest regression (RFR), k-nearest neighbors (k-NN) and support vector regression (SVR) and the results were compared to experimental data.

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2. Experimental

2.1. Sample preparation

The mother alloy of $Fe_{bal.(}Si_aB_bC_c)$ ($16 \leq a + b + c \leq 21$ at.%) alloys were prepared by an arc melting process. Fe (99.90 wt.%), Si (99.90 wt.%), Fe-B (Fe : 83.87 wt. %, B : 16.13 wt. %) and Fe-C (Fe : 95.7 wt.%, C : 4.3 wt.%) powders were used to fabricate the mother alloy. The contents of metalloids and non-metallic elements were controlled in the range of 1 at. % ≤ Si ≤ 6 at.%, 10 at.% ≤ B ≤ 15 at.% and 0.5 at.% ≤ C ≤ 3 at.%. The ribbons were solidified by a single melt-spinning apparatus under Ar atmosphere. The thickness of the ribbons was measured as approximately 25 μm. The soft magnetic properties of the Fe-Si-B-C amorphous alloys were evaluated using a vibrating sample magnetometer (VSM) (Lakeshore, 7410). The coercivity (Hc) and the saturation magnetization (Ms) was evaluated by an applied magnetic field of 125 G and 5000 G, respectively.

2.2. Data collection

Datasets were collected from the soft magnetic properties of 31 different Fe-Si-B-C amorphous alloy samples in the experimental process. The datasets contained the saturation magnetization and coercivity according to various metalloid contents. Table 1 shows the compositional ranges of the Fe-Si-B-C samples.

TABLE 1

The compositional ranges of Fe-Si-B-C (at.%)

Element	Minimum	Maximum	Average	Deviation
Si (at.%)	1	6	4.49	1.80
B (at.%)	10	15	13.13	1.42
C (at.%)	0.5	3	1.83	0.77

2.3. Machine learning training

The correlation between the magnetic properties of Fe-Si-B-C amorphous alloys and metalloid contents was evaluated using machine learning algorithms, specifically RFR, k-NN, and SVR. RFR was employed to train the target values using decision trees and estimate the average values [13,14]. RFR was trained 1 to 100 decision trees. k-NN meanwhile was predicted to the average value of the nearest data used to the k nearest value, with k values ranging from 1 to 22. SVR was shown to be affected by factors C, ϒ, and ε. The set of C was 1, 10, 100, and 1000. The ϒ values were 1, 0.1, and 0.001. We used ε sets of 0.0001, 0.0005, 0.001, 0.005, 0.01, 0.05, and 0.1 [15]. We used a radial basis kernel with a constant value of 0.1. The dataset for three algorithms, such as RFR, k-NN, and SVR, was divided into 70% for training with 30% for testing, and the predicted R^2 values, which identify the accuracy of predicting magnetic properties related to metalloid contents was calculated.

$$R^2 = \left(\frac{\sum\left[(xi - \bar{x})(xi - \bar{x})\right]}{\sqrt{\left[(xi - \bar{x})\right]^2 * \left[(xi - \bar{x})\right]^2}} \right)^2 \tag{1}$$

Eq. (1) shows R^2 the values. Algorithm assessments were performed by scikit-learn Version 0.23.1 module in Python Version 3.7, USA.

3. Results and discussion

3.1. Magnetic properties of experimental data

The magnetic properties of Fe-Si-B-C ribbons fabricated by the melt spun process are presented in Fig. 1. Fig. 1(a) shows the changes in the magnetic properties by controlling the Fe, B + C, and Si contents. The highest coercivity was observed at a Si content of 6 to 7 at.% and a B + C content of 12 at.% and less. However, the coercivity was shown to be effectively decreased when the B + C content was over 14 at.% even at the same Si range. Fig. 1(b) shows the effect of C content. As shown in the figure, the coercivity increased as the C contents exceeded 2 at.%. Despite that the content of C was only 1 to 2 at.%, the coercivity increased when the B + Si contents was 16 at.% or more. Fig. 1(c) shows the B effect. With increasing the B content, the coercivity decreased to 14 at.%. The lowest coercivity was found in the composition range of 5 to 6 at.% Si + C and 13.5 at.% of B. However, when the B content was 12.5 at.% or more, the coercivity did not always decrease. Figs. 1(d)~(e) show that the saturation magnetization is not linearly dependent on the Fe content. As shown in the figures, high Fe content does not mean high saturation magnetization values. These data clearly show that there should be an optimized chemical composition of Fe- based amorphous material.

3.2. Magnetic properties of machine learning results

The Pearson correlation coefficient values 'r' was calculated by a correlation analysis of metalloid elements to predict the sensitivity of soft magnetic properties according to metalloid elements and the results are shown in Figs. 2(a) and (b). Figs. 2(a) and (b) show the effects of the elements on the saturation magnetization and the coercivity, respectively. The correlation coefficient can be negative or positive correlation values. In general, the range of 0.7 to 1.0 means a perfect positive relationship, the range of 0.3 to 0.7 is a high positive relationship, and the range of 0.1 to 0.3 is a low positive relationship. The tendency of the negative values is the same as that of the positive values, for example, the range of –0.7 to –1.0 is a perfectly negative relationship [16], and so on. Fe is shown to have a high positive relationship with saturation magnetization. This means that the saturation magnetization is roughly linearly dependent on the Fe content. However, since Si has a high negative relationship

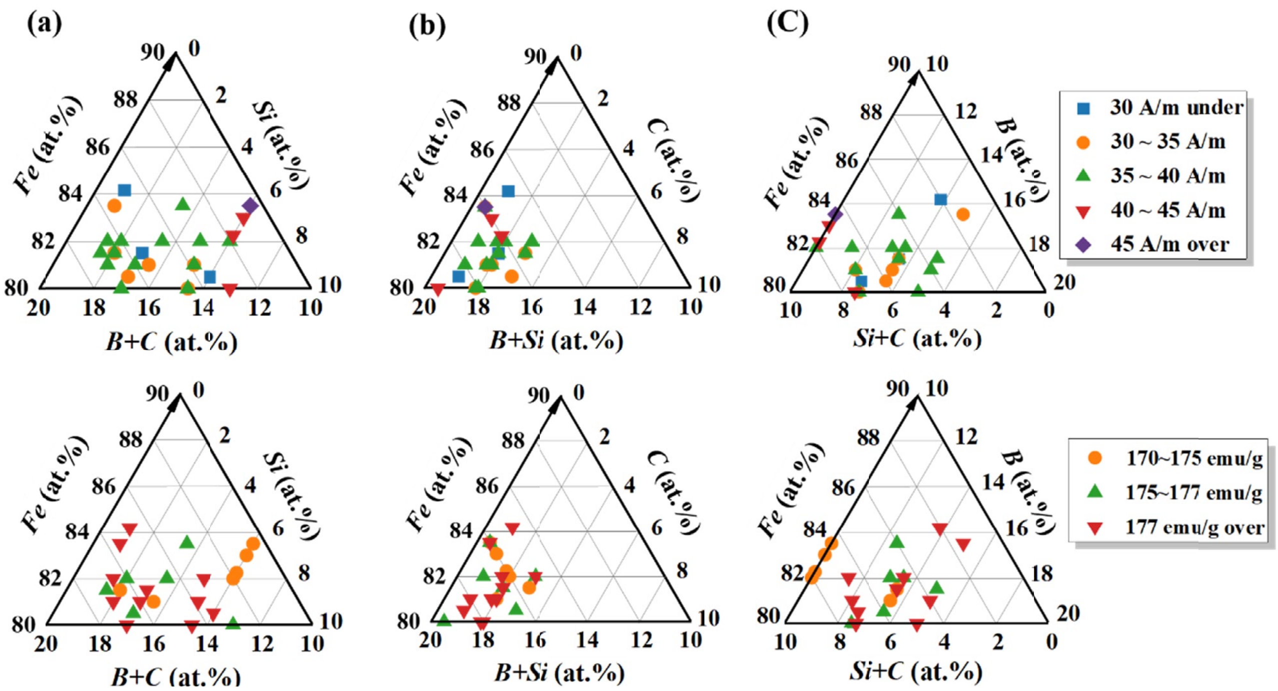

Fig. 1. Ternary diagram of coercivity and saturation magnetization of amorphous Fe-Si-B-C alloys at room temperature. (a) shows the Si effects on soft magnetic properties compared with B + C ratio, (b) shows B effects on soft magnetic properties compared with Si + C ratio, (c) shows C effects on soft magnetic properties compared with Si + B ratio

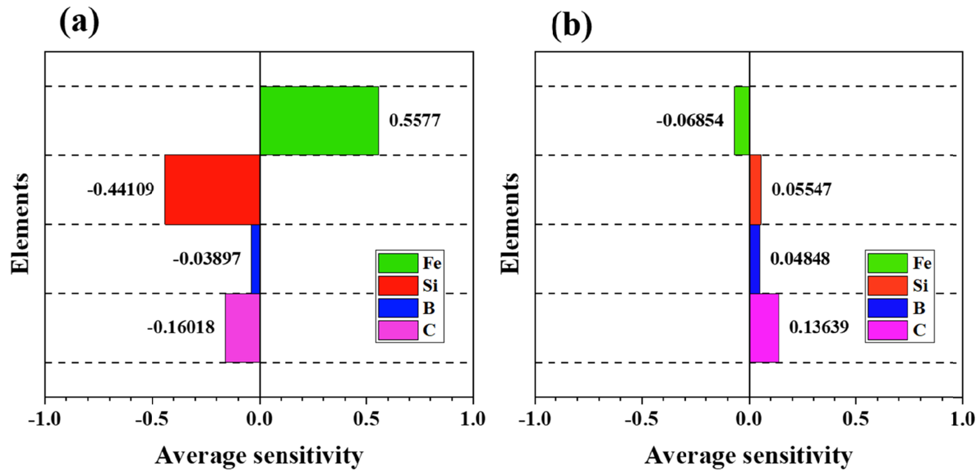

Fig. 2. Prediction of Pearson correlation coefficient values by a correlation analysis for sensitivity of metalloid elements. (a) shows average sensitivity for saturation magnetization, (b) shows average sensitivity for coercivity

with the saturation magnetization, it should cause a decrease of the saturation magnetization as the Si content increases. However, elements such as B and C show a weak relationship, indicating that the saturation magnetization is strongly affected by their contents.

Fig. 3 shows the accuracy of R^2 using various machine learning algorithms: SVR, RFR, and k-NN. The RFR method exhibits the highest R^2 values of 0.74 for coercivity and 0.878 for saturation magnetization, as presented in Fig. 3(a). It is generally held that the data are reliable if the R^2 value is above 0.6 and accurate if the R^2 values are above 0.9 [13].

This means that the computational value of 0.74 is not sufficiently accurate to determine the metalloid content ranges to obtain the desired soft magnetic properties. Previous literature, RFR and k-NN were used to estimate R^2, which represents the accuracy of soft magnetic properties. The computational R^2 values for saturation magnetization used RFR algorithm was 0.86 and R^2 values for coercivity was 0.76 [13]. The R^2 values calculated by the experimental results are similar to or higher than the data from previous study.

Fig. 3(b) shows the accuracy of the coercivity of the RFR. The R^2 values for the training and test sets were 0.802 and 0.625, respectively. Fig. 3(c) shows the accuracy of the saturation magnetization of the RFR and the R^2 values for the training and test sets were 0.937 and 0.649, respectively. The R^2 values of the test set were lower than those of the training set due to a limited

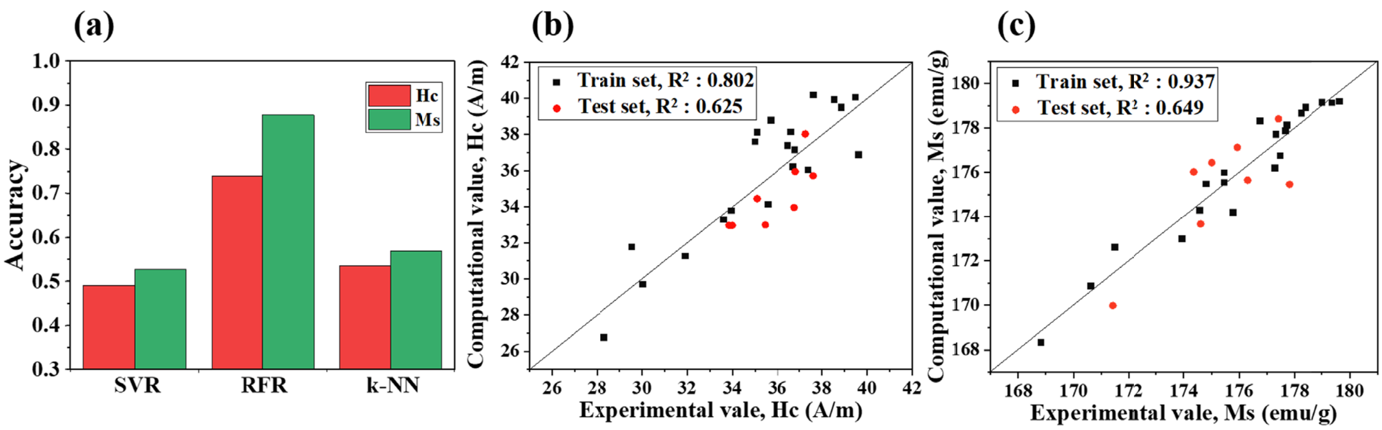

Fig. 3. Results of the accuracy by the machine learning algorithm process and R^2 values with RFR algorithm; (a) R^2 accuracy of magnetic properties for coercivity and saturation magnetization with three machine learning algorithms, SVR, RFR, and k-NN program, (b) Comparison of coercivity between computational values calculated by RFR algorithm and experimental results; black square point is dataset for train process and red circle is dataset of test results, (c) Comparison of saturation magnetization between computational values calculated by the RFR algorithm and experimental results; black square points are dataset for the training process and red circles are the dataset of test results

amount of data onto calculation. Thus, more data collection for Fe-Si-B-C alloys with various metalloid contents would increase the accuracy of the prediction of their soft magnetic properties.

Fig. 4(a) shows the effects of Si element on the coercivity and saturation magnetization by the computational results of the RFR algorithm for Fe-Si-B-C alloys with fixed B and C content. Fig. 4(b) and (c) show the effects of B and C calculated in the same manner as done for the Si effect, respectively. Fig. 4(a) shows that the lowest coercivity was obtained at a Si content of 3.2 at.% and that the coercivity increased but the saturation magnetization decreased as the Si content increases. As presented in Fig. 4(b), the minimum coercivity was obtained at a B content of 14.2 at.%. Fig. 4(c) shows the effect of C on the soft magnetic properties. The tendency of coercivity was similar to that of saturation magnetization and as the C content increased, the coercivity and saturation magnetization decreased. The metalloid contents to obtain minimum coercivity was evaluated by the RFR method with optimized Si, B, and C contents. The weighted values of each element were equal. For example, the chemical composition providing the lowest coercivity with various Si amounts and fixed B, and C was $Fe_{81.6}Si_{3.2}B_{13.2}C_2$ at.%. Additionally, the lowest coercivity composition among various B contents with fixed Si, and C was $Fe_{79.95}Si_{3.85}B_{14.2}C_2$ at.%. The optimized content of elements affecting soft magnetic properties was calculated as an average value. Consequently, the lowest coercivity was observed in $Fe_{80.5}Si_{3.63}B_{13.54}C_{2.33}$ at.% and the highest saturation magnetization was obtained in $Fe_{81.83}Si_{3.63}B_{12.63}C_{1.91}$ at.%, as predicted by the trained RFR algorithm.

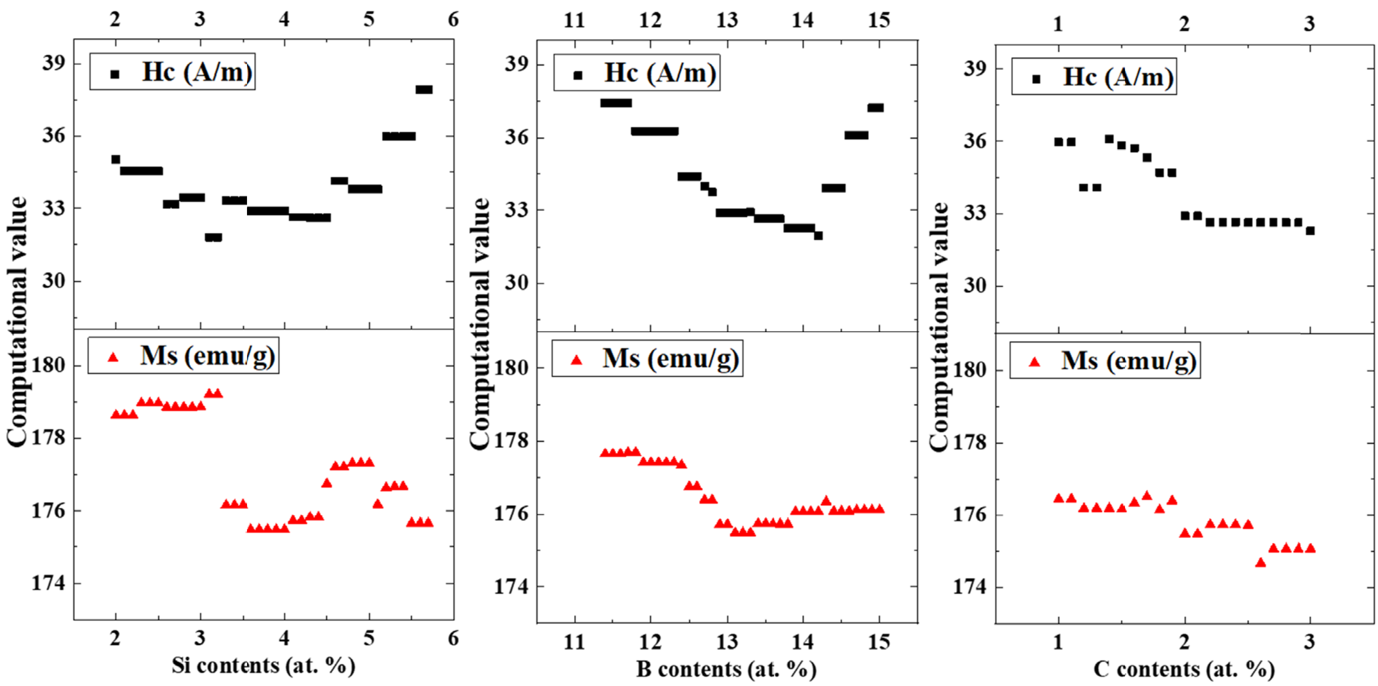

Fig. 4. The soft magnetic properties with changes in metalloid contents; (a) Si effect – B of 13.2 at.% and C of 2 at.% were fixed, (b) B effect – Si of 3.85 at.% and C of 2 at.% were fixed, (c) C effect – B of 13.2 at.% and Si of 3.85 at.% were fixed

4. Conclusions

In this study, the soft magnetic properties obtained from experimental data of Fe-Si-B-C amorphous ribbon were evaluated to reduce the coercivity and increase the saturation magnetization by controlling the metalloid composition. In the experimental data, the lowest coercivity was obtained in the composition of 6~7 at.% Si, and above 14 at.% B+C content and the saturation magnetization was uncertain of metalloids effects. The RFR machine learning algorithm was used to predict the effects of metalloid contents on the soft magnetic properties. When Si content of 3.2 at.%, B content of 14.2 at.%, and C content of 3 at.% were added, the coercivity decreased to 31 A/m in the Fe-Si-B-C amorphous soft magnetic composition of calculating data. The saturation magnetization of an alloy used calculating data onto 3.2 at.% Si, 11.5 at.% B, and 1.7 at.% C reached 177.5 emu/g.

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