Inverse Design of Fe-Based Bulk Metallic Glasses Using Machine Learning

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Abstract: Fe-based bulk metallic glasses (BMGs) are a unique class of materials that are attracting attention in a wide variety of applications owing to their physical properties. Several studies have investigated and designed the relationships between alloy composition and thermal properties of BMGs using an artificial neural network (ANN). The limitation of the wide-scale use of these models is that the required composition is yet to be found despite numerous case studies. To address this issue, we trained an ANN to design Fe-based BMGs that predict the thermal properties. Models were trained using only the composition of the alloy as input and were created from a database of more than 150 experimental data of Fe-based BMGs from relevant literature. We adopted these ANN models to design BMGs with thermal properties to satisfy the intended purpose using particle swarm optimization. A melt spinner was employed to fabricate the designed alloys. X-ray diffraction and differential thermal analysis tests were used to evaluate the specimens.

Keywords: Fe-based bulk metallic glasses; materials design; machine learning; artificial neural network; inverse design

1. Introduction

Fe-based BMGs have vast industrial application potential in electronics, magnetics, and aerospace industries owing to their high mechanical strength, heat resistance, strong corrosion resistance, and excellent soft magnetic properties [1–4]. The heat resistance of the Fe-based BMGs is determined by the crystallization temperature and glass transition temperature. When the temperature surrounding BMGs exceeds the crystallization temperature, BMGs transform into a crystal structure. This causes a decline in heat resistance, mechanical properties, and corrosion resistance. Thus, it is important to predict the thermal properties of Fe-based BMGs. The thermal properties of Fe-based BMGs depend on their alloy composition. This is key for regulating the composition ratio to design Fe-based BMGs. To understand and control the effect of composition on thermal properties, diverse research has been conducted [5–19].

Classical alloy modelling methods are generally based on a “trial-and-error” process that requires considerable time, effort, and costs. In the case of ANN, one of the latest statistical modelling methods, it is possible to predict the properties of various alloys with less time and cost through existing data. Owing to these advantages, researchers have applied ANN to predict the properties of metallic glasses [1,20–34]. Keong et al. predicted the crystallization temperature of Ni-P based amorphous alloys [20]. Cai et al.
predicted the reduced glass transition temperature of Zr-Al-Ni-Cu BMGs [21]. The alloy design using ANN could predict and design novel alloys without experimentation and cost. However, finding specific composition pairs and target values using combinations of input parameters remains an issue.

One promising scheme for optimizing tools for material discovery is the metaheuristic method. The metaheuristic method is an approximate search method for solving complex optimization problems [35]. Metaheuristic methods include genetic algorithm (GA), particle swarm optimization (PSO), and ant colony optimization. To design new materials, several studies have adopted the metaheuristic method [36–43]. Anijdan et al. used a GA model with an ANN to design an Al–Si casting alloy [36]. Shojaeeefard et al. reported the welding process between AA7075 and AA5083 using PSO with ANN [37]. Ming et al. proposed a modified Mg–Li–Al alloy using PSO with an ANN [38]. These previous studies prove the possibility of discovering new Fe-based BMGs according to a similar modelling approach.

The goal of the present study is to propose the inverse design method using both ANN model and PSO model for the alloy design of Fe-based BMGs. We gathered a dataset containing the composition and thermal properties of Fe-based BMGs from the literature to build the ANN model. A few alloy compositions were proposed using PSO modeling and their thermal properties were experimentally validated. This study was performed to show one of the possible numerical methods to determine the Fe-based BMG composition, not to find a certain composition with target properties.

2. Overall Process to Design New Alloys

Figure 1 shows the four main steps to design the Fe-based BMGs in the present study. The first step was to collect and refine the database of Fe-based BMGs for generating the ANN. The second step was to learn and select the ANN for predicting the thermal properties of Fe-based BMGs. The next step was to invert the design of the ANN using a metaheuristic method to suggest the composition of Fe-based BMGs. The final step is to fabricate the Fe-based BMGs samples for experimental verification. The details of each step are described in the following sections.

![Figure 1. Process of Fe-based BMGs design.](image-url)

2.1. Database Collection

The datasets for the Fe-based BMGs were gathered from the usable data in the published literature [44,45]. Datasets were constructed with 16 compositions (Fe, Cr, Mn, Mo, ...
B, C, Ni, Co, W, P, Si, Y, Zr, Al, Nb, and Hf) and thermal properties, including crystallization temperature ($T_x$) and glass transition temperature ($T_g$). The ranges of the composition and thermal properties are listed in Table 1.

Table 1. Range of the composition and thermal properties for the dataset used in this study.

| Variable | Minimum | Maximum | Average | Deviation |
|----------|---------|---------|---------|-----------|
| Cr (at.%) | 0       | 16      | 2.7     | 5.0       |
| Mn (at.%) | 0       | 19.5    | 1.2     | 4.6       |
| Mo (at.%) | 0       | 16      | 4.7     | 5.4       |
| B (at.%)  | 0       | 24      | 12.3    | 7.7       |
| C (at.%)  | 0       | 18      | 5.0     | 6.0       |
| Ni (at.%) | 0       | 36      | 0.7     | 3.9       |
| Co (at.%) | 0       | 36      | 3.9     | 7.4       |
| W (at.%)  | 0       | 4       | 0.2     | 0.5       |
| P (at.%)  | 0       | 13      | 3.0     | 4.6       |
| Si (at.%) | 0       | 9.9     | 1.4     | 2.3       |
| Y (at.%)  | 0       | 6       | 1.0     | 1.7       |
| Zr (at.%) | 0       | 10      | 1.6     | 3.4       |
| Al (at.%) | 0       | 4       | 0.2     | 0.6       |
| Nb (at.%) | 0       | 8       | 1.2     | 1.9       |
| Hf (at.%) | 0       | 5       | 0.2     | 1.0       |
| $T_x$ (°C) | 455     | 715     | 550.0   | 55.6      |
| $T_g$ (°C) | 374     | 645     | 550.8   | 68.0      |

2.2. ANN Model Learning

ANN learning is the process of seeking the optimal weight factors to represent the best performance between input and output data. Hyper-parameters, including the number of layers, activation function, optimizer, and dataset separation, are the main factors in ANN model learning. In this study, we changed the number of layers from 1 to 2 and the neurons from 1 to 100. The datasets were separated into 70%, 15%, and 15% for training, validation, and testing, respectively. The rectified linear unit (ReLU) function was applied to the hidden layers, while the linear transfer function (purelin) was employed to the output layer. Ridge regression and early stopping were used to suppress overfitting. Two ANN models were trained for prospect $T_x$ and $T_g$, respectively. The schematic structure of the ANN model is illustrated in Figure 2. The ANN model structure with the number of layers and neurons was determined using the coefficient of determination ($R^2$) value.

Figure 2. Schematic structure of the ANN model.
2.3. Inverse Design

Although the ANN model can predict the thermal properties of Fe-based BMGs, a long calculation time is required because of the diversity and complexity of the combination of composition factors. The inverse design using metaheuristic algorithms offers a key to rapidly finding a composition in the expansive space of the mixtures of composition. In this study, PSO was adopted as a metaheuristic algorithm for the inverse design of Fe-based BMGs. The target \( T_x \) was limited from 550 to 600 °C, while the target \( T_g \) ranged from 520 to 570 °C.

2.4. Experimental Verification

Fe powder with a purity of 99.9% and average particle size (APS) of <75 µm, Cr powder with a purity of 99% and APS of <140 µm, Mo powder with a purity of 99.95% and APS of 3–7 µm, B powder with a purity of 99% and APS of 1–2 µm, graphite powder with a purity of 99.6% and APS of <20 µm, Ni powder with a purity of 99.9% and APS of 3–7 µm, and P powder with purity 98.9% and APS of <140 µm were mechanically alloyed using a high-energy ball mill (a Pulverisette-5 planetary mill) at 250 rpm for 24 h. Tungsten carbide balls with a diameter of 10 mm were used for milling in a sealed cylindrical SKD-11 tool steel jar with a ball-to-powder ratio of 30:1 under an argon atmosphere. Arc melting was selected to make a bulk sample for the melt spinner. The milled powder alloys were packed into a cylindrical steel die with an inner diameter of 200 mm, an outer diameter of 600 mm, and a height of 800 mm. The milled powder was compressed to 10 MPa. The green compacts were arc melted under an argon atmosphere. Fe-based BMG ribbons were produced using a melt spinner with a 4000 rpm copper rotating wheel and argon atmosphere. Differential thermal analysis (DTA) was carried out using a thermal analyzer (TG-DTA, TG-8121, Rigaku, Tokyo, Japan) with a heating rate of 0.64 °C/s. The phase state of the specimens was qualitatively analyzed using X-ray diffraction (XRD, MAX-2500, Rigaku, Tokyo, Japan) and selected area electron diffraction (SAED, JEM-ARM200F, JEOL, Tokyo, Japan).

3. Results and Discussion

The two-layered ANN model was successfully learned with extremely reliable \( R^2 \) values, which were 0.9662/0.9391 (train/test) for \( T_x \), and 0.9473/0.9453 (train/test) for \( T_g \), respectively. The optimal structures of the ANN models were 7-64-58-1 and 7-53-50-1 for \( T_x \) and \( T_g \), respectively. We proposed four new chemical compositions of Fe-based BMGs to satisfy the restricted ranges of the thermal properties. The suggested compositions are summarized in Table 2.

Table 2. Fe-based BMG alloys proposed by inverse design.

| Variable     | AP1  | AP2  | AP3  | AP4  |
|--------------|------|------|------|------|
| Cr (at.%)    | 0.6  | 0.2  | 0.3  | 0    |
| Mo (at.%)    | 2.6  | 1.4  | 1.6  | 8.6  |
| B (at.%)     | 20.2 | 20.1 | 22.1 | 25.6 |
| C (at.%)     | 12.8 | 9.9  | 10.2 | 9.8  |
| Mn (at.%)    | 0    | 12.1 | 0    | 0    |
| Ni (at.%)    | 0    | 0    | 1.0  | 0    |
| P (at.%)     | 0    | 0    | 0    | 5.4  |
| Fe (at.%)    | Bal. | Bal. | Bal. | Bal. |
| \( T_{x,\text{cal}} \) (°C) | 568.5| 571.5| 572.3| 624.1|
| \( T_{g,\text{cal}} \) (°C) | 564.0| 530.2| 557.6| 610.9|
| \( T_{x,\text{exp}} \) (°C) | 587.4| 590.6| 605.6| 589.7|
| \( T_{g,\text{exp}} \) (°C) | 548.3| 529.7| 568.8| 553.3|

Figure 3a shows the XRD patterns of the designed alloy samples, whereas Figure 3b–e shows the DTA results to verify the \( T_x \) and \( T_g \) values. Figure 4 shows SAED results that have metallic glasses phase formation. In the XRD results, a small amount of crystal
structure was detected. However, the metallic glass phase formation was confirmed through SAED results showing a metallic glass selected area electron diffraction pattern without lattice fringes. The measured results are compared in Table 2.

![Figure 3](image-url)

**Figure 3.** (a) XRD and (b–e) DTA results of the designed Fe-based BMG samples.

![Figure 4](image-url)

**Figure 4.** (a–d) Selected area electron diffraction of the designed Fe-based BMG samples.
In contrast, Figure 5 compares the experimental thermal properties with the predicted values to confirm the accuracy of the inverse design approach in the present study. Black circles are the literature data used to train and test the ANN. Red asterisk symbols indicate the designed Fe-based BMGs. The inverse design approach with the pair of ANN and PSO methods could provide a path for finding new Fe-based BMG alloy compositions with desirable thermal properties.

**Figure 5.** Predicted (a) $T_x$ and (b) $T_g$ values using the ANN model compared with the experimental data. Red asterisk symbols indicate the values of the four designed Fe-based BMG samples by the inverse design method.

4. Conclusions

In this study, we presented the ANN design and metaheuristic method for Fe-based BMGs and validated these predictions using powder metallurgy and melt spinning. The ANN models were organized using a dataset composed of several hundred Fe-based BMGs experiments obtained from the relevant literature. These data were used to train ANN models to predict the crystallization temperature and glass transition temperature of BMGs based on their composition. Then, PSO was adopted for obtaining the alloy composition, which has an apposite crystallization temperature and glass transition temperature. Suggested alloys were validated experimentally, with results showing that their thermal properties fall within the chosen constraints. This ANN model and PSO method can facilitate the search for novel Fe-based BMGs with appropriate thermal properties despite the challenge of finding an alloy in numerous alloy combinations using ANN.

**Author Contributions:** Conceptualization, M.-H.L., and S.-J.L.; methodology, J.J., and N.S.; investigation, J.J., and S.B.S.; writing—original draft preparation, J.J.; writing—review and editing, S.B.S.; supervision, H.-J.K. and S.-J.L.; project administration, H.-K.L. and H.-J.K.; funding acquisition, H.-K.L. and S.-J.L. All authors have read and agreed to the published version of the manuscript.

**Funding:** This research was supported by the Ministry of Trade, Industry and Energy (MOTIE) and the Korea Institute for Advancement of Technology (KLIAT) through the International Cooperative R&D program (P006837). Also, this work was supported by a Korea Institute for Advancement of Technology grant, funded by the Korea Government (MOTIE) (P0002019), as part of the Competency Development Program for Industry Specialists.

**Institutional Review Board Statement:** Not applicable.

**Informed Consent Statement:** Not applicable.

**Data Availability Statement:** The data presented in this study are available on request from the corresponding author.

**Conflicts of Interest:** The authors declare no conflict of interest.
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