Using Experimental Data

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A multi-dimensional random number generation algorithm was used to distribute chemical concentrations of each of the alloying elements in the candidate alloys as uniformly as possible while maintaining the prescribed bounds on the minimum and maximum allowable values for the concentration of each of the alloying elements. The generated candidate alloy compositions were then examined for phase equilibria and associated magnetic properties using a thermodynamic database in the desired temperature range. These initial candidate alloys were manufactured, synthesized and tested for desired properties. Then, the experimentally obtained values of the properties were fitted with a multi-dimensional response surface. The desired properties were treated as objectives and were extremized simultaneously by utilizing a multi-objective optimization algorithm that optimized the concentrations of each of the alloying elements. This task was also performed by another conceptually different response surface and optimization algorithm for the purpose of double-checking the results. A few of the best predicted Pareto optimal alloy compositions were then manufactured, synthesized and tested to evaluate their macroscopic properties. Several of these Pareto optimized alloys outperformed most of the candidate alloys on most of the objectives. This proves the efficacy of the combined meta-modeling and experimental approach in design optimization of the alloys. A sensitivity analysis of each of the alloying elements was also performed to determine which of the alloying elements contributes the least to the desired macroscopic properties of the alloy. These elements can then be replaced with other candidate alloying elements such as not-so-rare earth elements.

1. Introduction

Rare earth element (REE) based magnets have a very high magnetic energy density \((BH)_{\text{max}}\). This means that it is possible to synthesize smaller magnets while maintaining the superior magnetic properties. These magnets also have higher coercivity \((H_C)\), making it difficult to demagnetize under external magnetic fields. Neodymium magnets are the strongest available magnets in this family. However, Nd-Fe-B (Neodymium-Iron-Boron) performs the best up to 150-degree centigrade. From 150 ºC to 350 ºC, Sm-Co (Samarium-Cobalt) magnets are used. These magnets usually need a protective coating in order to prevent corrosion. REE-based magnetic materials are essential in electric cars, in wind turbine electric generators, and any high-efficiency electric devices requiring magnetic fields. Thus, REEs are strategic materials determining which national economies will survive and prosper in the post-combustion-engine era. The problem is that deposits of most of the rare earth elements used for synthesizing these magnets are located in China. Due to depleting resources and stringent trade rules from the suppliers, it is important to look at other options to synthesize these magnets.

Alnico magnets, [1] are permanent magnetic alloys based on the Fe-Co-Ni-Al system without REEs. Alnico magnets have very high \(B_r\) values, comparable to REE magnets. Alnico magnets have lower \(H_C\) values and can be demagnetized in the presence of an external magnetic field. However, low \(H_C\) also means that these magnets can be easily magnetized to saturation. A high \(B_r\) and low \(H_C\) value help to cast this material in complex shapes while magnetizing it in the production heat treatment stages. Alnico magnets offer excellent corrosion resistance and high-temperature stability and are the only magnets that can be used up to 800 ºC. The excellent high-temperature stability combined with a comparatively higher temperature limits have been successfully exploited by researchers in the past. These properties make it a perfect choice for military and automotive sensor applications. Hence, any improvement in the existing properties of Alnico alloys will help in covering the gap between the Alnico and REE based magnets. This will help in addressing a few important energy conversion applications.
In the present research work, a novel approach is presented of computational tools in design and multi-objective optimization of permanent magnetic Alnico type alloys. The proposed research combines a number of numerical design optimization algorithms with several concepts from artificial intelligence and experimentally evaluated desired properties of an affordable set of candidate alloys. These alloys were further screened by various statistical tools in order to determine any specific trend in the data. This information will be helpful to the research community in developing a material knowledgebase for new alloys for targeted properties.

2. Background

At present, researchers around the globe are working on designing magnetic alloys that will be able to cover the gap between the properties achieved by Alnico magnets and the rare-earth magnets, basically by adding small amount of those rare-earth elements that are less critical in the sense of supply [2]. This will help in addressing a few important energy conversion applications. Sellmyer et al. [3] and Balamurugan [4] worked on a few rare-earth free alloys and the properties were found to be in the vicinity of Alnico alloys. Zhou et al. [5] re-manufactured a few commercial Alnico alloys to demonstrate the scope of improvement in this field. However, the difference between the theoretically calculated and the measured properties were quite large for (BH)max and Hc. Hence, random experimentation may be misleading in terms of improvement in alloy properties while being both expensive and time-consuming.

Designing of new alloys or even improving the properties of existing alloys is a challenging task mainly due to experimental database. In order to develop a reliable knowledge base [6] for design of new alloys, one needs to focus on determining various correlations (composition-property, property-property, composition-composition) from the available databases (simulated and experimental). This information can be coupled with the theoretical knowledge (atomistic and continuum based theories) to develop the knowledge base. Integrated Computational Materials Engineering (ICME) approach [7] along with the materials genome initiative [8] highlighted the importance and growing application of computational tools in of new alloys. It aims at reducing a new alloy development cycle from currently 10 years to two years or even less [8]. In recent years, various data-driven techniques combined with evolutionary approaches [9] have been successfully implemented in alloy design [10, 11, 12, 13, 14] and also in improving thermodynamic databases such as “ThermoCalc” [15] for alloy development. Data mining approaches like principal component analysis [PCA] and partial least square[PLS] regression have been successfully used in designing Magnesium alloys [16], as well as determining properties of Lutetium[17]. These applications demonstrate the efficacy of application of computational tools for materials design.

Alnico magnets were discovered in 1931 by Mishima in Japan [1]. Initially, these magnets were based on Fe-Co-Ni-Al system. Magnetic properties in these magnets are attributed to the presence of a two-phase system of BCC (Body Centered Cubic) α1 and α2. It was later observed that separation of α1 and α2 is due to a metallurgical phenomenon popularly known as “Spinodal” decomposition. α1 is Fe-Co rich ferromagnetic phase while α2 is Ni-Al rich phase. These phases are stable up to 850-degree centigrade (Curie temperature is about 860°C). Above 850°C, FCC (Face centered cubic) phase begins to appear. An FCC γ phase was observed in a few compositions. This phase is quite detrimental for magnetic properties. In later years, attempts were made to stabilize the magnetic α1 and α2 phases while at the same time eliminate or reduce the amount of FCC γ phase. These attempts include, modification of heat treatments and addition of other elements to enhance various magnetic properties of these materials. Since, 1980, there has been limited research on alnico magnets mainly due to the discovery of powerful REE-based magnets. The Recent rise in prices of rare earth elements led to the search of rare-earth free magnets. In recent years, there has been a significant amount of work in this field. Due to their proven high-temperature stability and related properties, Alnico magnets are a popular choice for research.

Currently, Alnico alloys contain 8+ elements. In our work, we used 8 alloying elements. These elements include Iron (Fe), Cobalt(Co), Nickel(Ni), Aluminum(Al), Titanium(Ti), Hafnium(Hf), Copper(Cu) and Niobium(Nb). Variable of these elements have been tabulated in Table 1. It is important to know the role of these alloying elements initially for the manufacture of alloys for targeted properties. Later, this information can be utilized to select meta-model for a certain property. This is done with a purpose to develop a knowledge base for discovery of new materials while improving properties of existing materials.
Research over the years suggests that Coercivity and Remanence are conflicting. That is one has to sacrifice on one of these properties to improve the other property. Additionally, magnetic energy density is dependent on $H_c$ and $B_r$. Therefore, in order to increase $(BH)_{max}$ one needs to optimize $H_c$ and $Br$. Role of various alloying elements and its effect on $H_c$ and $Br$ has been addressed in the following text:

1. Cobalt: It is a $γ$ stabilizer. Hence, a anneal is needed to homogenize it to a single $α$ phase. Cobalt increases coercivity and Curie temperature.
2. Nickel: It is also a $γ$ stabilizer. Hence, a anneal is needed to homogenize it to a single $α$ phase. Nickel increases $H_c$ (Less than Cobalt). But, it does this at the expense of $B_r$.
3. Aluminum: It is a $α$ stabilizer. Hence, it will help in reducing the solutionization temperature. It affects $H_c$ positively.
4. Copper: It is also a $α$ stabilizer. Hence, it will help in reducing the temperature. It affects $H_c$ positively and increases it. Additionally, it also increases $B_r$. In Alnico 8 and 9, Cu precipitates out of the $α_2$phases into particles and increases the magnetic separation between $α_1$ phases. It results in an increase in $H_c$. In Alnico5-7, Cu remains in $α_2$phases and it leads to an increase in $H_c$ while a decrease in Curie temperature.
5. Titanium: It is also a $α$ stabilizer. It is one of the most reactive elements, hence it reacts with impurities such as S and N and precipitates out or purifies the magnet. It also eliminates Carbon. Carbon is a strong $γ$ stabilizer and hence needs to be eliminated at any cost. Titanium helps in grain refining and assists columnar grain growth. It is to be noted that, due to columnar grain growth, of grains are aligned perpendicular to the chill plate. Large shape anisotropy can be achieved if spinodal decomposition occurs in this direction. Titanium increases $H_c$ while it decreases $B_r$.
6. Niobium: It is also a $α$ stabilizer. It helps in neutralizing the affects of Carbon. Like Titanium, Nb too assists in columnar grain growth. Nb increases $H_c$, while it decreases $B_r$. But decrease in Br due to Nb is less than that observed due to Ti.
7. Hafnium: it is not a $γ$ stabilizer. It is used for enhancing high-temperature properties. Hf usually precipitates at the grain boundary and helps in improving creep properties. Recent studies related to Co-Hf magnets motivated us to use Hf in this work.

### 3. Current Research

In this work, we used a set of computational tools to develop a novel approach for design and optimization of high temperature, highintensity magnetic alloys.

The steps involved in the proposed approach can be listed as follows:

1. Initial dataset:From the open literature, we defined the variable bounds of 8 alloying elements in the alloy. We used one of the best-known quasi-random number generators, Sobol’s algorithm [10, 11, 12], to generate chemical concentrations for each of the 80 candidate alloys (see Table 1 and Table 2). The initial set of alloys were screened on the basis of limited knowledge of phase equilibrium and magnetic property from a commercial thermodynamic database,”Factsage”[18].
2. Manufacture and testing: The alloys were manufactured and tested for various properties of interest as shown in Table 3.
3. Response surface generation: From the available data, response surfaces were developed for the measured and calculated properties listed in Table 3. We used a commercial optimization package,”modeFRONTIER”[19] for this purpose. The response surfaces were tested on various accuracy measures and the most accurate one was chosen for further study. Various approaches to develop response surfaces are listed as follows: Radial basis functions (RBF), Kriging, Anisotropic Kriging and Evolutionary Design.
4. Multi-objective optimization: The most accurate response surfaces were used to extremize the various properties as per the objectives specified in Table 3. It was observed that most of the optimization tasks yielded alloys with similar chemical composition for a set of objectives. Hence, several optimization runs were performed to get a diverse pool of results. The optimization algorithms used in modeFRONTIER are listed as follows:Non-dominated Sorting Genetic Algorithm II (NSGA2), Multi-Objective Particle Swarm Optimization (MOPSO), Multi Objective Simulated Annealing (MOSA) and FAST (FAST optimizer uses response surface models (meta-models) to speed up the optimization process using search algorithms like NSGA2, MOPSO, MOSA).
5. This work was independently carried out at three different place using:
   1. Commercial optimization package, “Indirect Optimization based on Self-Organization algorithm (IOSO)”.
   2. Hybrid response surface by Professor M.J. Colaco.
   3. Surrogate model selection algorithm developed by Dr. Souma Chowdhury.
Predictions from both the optimization packages were merged and a set of alloys were selected for further manufacture and testing.

6. The work is performed in cycles. That is, Step 2-5 will be repeated until the improvements of multiple macroscopic properties of such magnetic alloys become negligible.

7. Sensitivity analysis: Various statistical tools were used to determine composition-property relations. This is done in order to find the most and the least influential alloying element.

This work is aimed at developing a knowledgebase that will help the research community in designing new alloys.

In data-driven material science, knowledge discovery [6,20] for designing new materials requires:

1. Data: In this work, our database is a combination of experimentally verified data predicted by a well-known random number generator and data from Pareto-optimized predictions as discussed above.
2. Correlations: Here, we used various linear and nonlinear correlation, clustering, and principal component analysis tool to discover various trends in the dataset.
3. Theory: The above information can be coupled with theoretical knowledge to motivate the experimentalist to move forward with the manufacture of new alloys.

Table 1: Concentration bounds AlNiCo type alloys

<table>
<thead>
<tr>
<th>Alloys number</th>
<th>Variable bounds (Wt %)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1 – 85</td>
</tr>
<tr>
<td>1 Cobalt (Co)</td>
<td>24 – 40</td>
</tr>
<tr>
<td>2 Nickel (Ni)</td>
<td>13 – 15</td>
</tr>
<tr>
<td>3 Aluminum (Al)</td>
<td>7 – 9</td>
</tr>
<tr>
<td>4 Titanium (Ti)</td>
<td>0.1 – 8</td>
</tr>
<tr>
<td>5 Hafnium (Hf)</td>
<td>0.1 – 8</td>
</tr>
<tr>
<td>6 Copper (Cu)</td>
<td>0 – 6</td>
</tr>
<tr>
<td>7 Niobium (Nb)</td>
<td>0 – 2</td>
</tr>
<tr>
<td>8 Iron (Fe)</td>
<td>Balance to 100</td>
</tr>
</tbody>
</table>

Table 2: Cycle and alloy number

<table>
<thead>
<tr>
<th>Cycle no.</th>
<th>Alloys Designed</th>
<th>Best alloy</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1-80</td>
<td>#30</td>
</tr>
<tr>
<td>2</td>
<td>81-85</td>
<td>#84</td>
</tr>
<tr>
<td>3</td>
<td>86-90</td>
<td>#86</td>
</tr>
<tr>
<td>4</td>
<td>91-110</td>
<td>#95</td>
</tr>
<tr>
<td>5</td>
<td>111-120</td>
<td>#117</td>
</tr>
<tr>
<td>6</td>
<td>120-138</td>
<td>#124</td>
</tr>
<tr>
<td>7</td>
<td>139-143</td>
<td>#139</td>
</tr>
<tr>
<td>8</td>
<td>144-150</td>
<td>#150</td>
</tr>
<tr>
<td>9</td>
<td>150-160</td>
<td>#157</td>
</tr>
<tr>
<td>10</td>
<td>160-165</td>
<td>#162</td>
</tr>
<tr>
<td>11</td>
<td>166-173</td>
<td>#169</td>
</tr>
</tbody>
</table>

Table 3: Quantities to be simultaneously extremized using multi-objective optimization

<table>
<thead>
<tr>
<th>Properties</th>
<th>Units</th>
<th>Objective</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 Magnetic energy density ((BH)_{max})</td>
<td>kg m^{-1} s^{-2}</td>
<td>Maximize</td>
</tr>
<tr>
<td>2 Magnetic coercivity (H_{c})</td>
<td>Oersted</td>
<td>Maximize</td>
</tr>
<tr>
<td>3 Magnetic remanence (B_{r})</td>
<td>Tesla</td>
<td>Maximize</td>
</tr>
<tr>
<td>4 Saturation magnetization (M_{s})</td>
<td>Emu/g</td>
<td>Maximize</td>
</tr>
</tbody>
</table>
5. Remanence magnetization ($M_r$) | Emu/g | Maximize
6. ($BH_{max}/mass$) | m$^{-1}$ s$^{-2}$ | Maximize
7. Magnetic permeability ($\mu$) | kg m A$^{-2}$ s$^{-2}$ | Maximize
8. Cost of raw material (cost) | $$/kg | Minimize
9. Intrinsic coercive field ($jH_c$) | Am$^{-1}$ | Maximize
10. Density($\rho$) | Kg m$^{-3}$ | Minimize

4. Results

At present, we have worked through 11 cycles of design and optimization followed by experimental validation. Table 2 lists the alloys manufactured in each of the cycles and the best alloy in each cycle.

Work done in all the cycles are described as follows:

Cycle # 1 (Alloy 1-80):
Candidate alloy compositions were predicted by Sobol's algorithm and the initial set of 80 elements were chosen for manufacture and testing. Measured properties were not according to our expectation but then also we proceeded further for design and optimization in hope of improved results.

Cycle # 2 (Alloy 81-85):
One of the predicted alloys (alloy # 84) outperformed the initial set of alloys as well as the other predicted alloys. This demonstrates the efficacy of the current approach in using computational tools in materials design. Hence, we moved forward in hope of further improvements. The variable bounds were modified and the new bounds are listed in Table 1.

Cycle # 3(Alloy 86-90):
Alloy # 86 was the best candidate in this set. But it was observed that the measured properties of the new set (alloy 86-90) were in the vicinity of the previous pool of alloys. This can be attributed to non-uniform distribution of alloying elements in the variable space. This eventually led to the development of inaccurate response surfaces and subsequently inaccurate predictions. Since, there was no significant improvement; we predicted the next set of alloys by Sobol's algorithm. Hence, the next set of alloys was predicted in order to provide response surfaces with more support points in the variable space.

Cycle # 4 (Alloy 91-110):
Alloy # 95 was the best performer in this group. Our approach to provide more support points for the response surfaces proved to be helpful in improvement of properties. Alloy # 95 had an $H_c$ of 980 Oe (against 750 Oe for the previous best alloy # 84). This improvement motivated us to proceed towards design and optimization task.

Cycle # 5 (Alloy 111-120):
Alloy # 117 is the best alloy in this dataset in terms of ($BH_{max}\)$. There was a significant improvement in the properties of the new alloys. Alloy # 111 and 114 had a $H_c$ of 1050 Oe while alloy # 117 reported 1000 Oe (against 980 Oe for the previous best alloy # 95). This improvement motivated us to proceed towards design and optimization task.

Cycle # 6 (Alloy 121-138):
Alloy # 124 was the best performer in this group. There was a significant improvement in both ($BH_{max}\) and $H_c$. Hence, we moved forward towards design and optimization part.

Cycle # 7 (Alloy 139-143):
Alloy # 139 was the best performer in this group. But, its properties were in the vicinity of alloy # 124. Since, there was no significant improvement in the desired properties; we halted the design and optimization process.
This was done in order to minimize waste of resources and funding. The need was to perform a sensitivity analysis of the variables and associated properties.

**Cycle # 8-11 (Alloy 144-173):** In these cycles, variable bounds were relaxed by 5% while the methodology remains the same.

**Cycle # 8 (Alloy 144-150):** modeFRONTIER

**Cycle # 9 (Alloy 151-160):** Surrogate model selection algorithm (SM)

**Cycle # 10 (Alloy 161-165):** modeFRONTIER

**Cycle # 11 (Alloy 166-173):** Hybrid response surface and modeFRONTIER

Figure 1, 2 and 3 shows the comparison between various approaches for a set of properties.

![Figure 1: Scatter plot: Magnetic energy density vs Magnetic coercivity](image-url)
Figure 2: Scatter plot: Magnetic energy density vs Magnetic remanence, comparison of solutions by various approaches

Figure 3: Scatter plot: Magnetic Coercivity vs Magnetic remanence, comparison of solutions by various approaches

It can be observed that the alloys predicted by meta-modeling and optimization dominates the ones predicted by Sobol’s algorithm. Though, it must be observed that the achieved properties are not at par with those available commercially. Hence, we need to do sensitivity analysis of the response surfaces as well as look for patterns in the dataset.
5. Sensitivity analysis

It was done in order to determine the composition-property relationship and to find various trends and patterns within the dataset. Initially, Pearson’s linear correlation method was used. But the coefficients were too low. It was expected as the dataset is quite noisy. A few other methods used to address this issue are discussed below.

Single Variable Response:

It is a methodology often applied for qualitative analysis of the training results obtained from Evolutionary Neural Network and Bi-Objective Genetic Programming [21]. In SVR, a trend of variation is created by generating values between 0 and 1 on a time scale. The trend line is irregular, that is there are regions of constant values, sharp increases, and sharp decreases in the line. This has been referred to as an input signal in the following text. For SVR testing, the input signal (a trend of variation) was used for one of the variables while the other variables were kept constant at an average value. The various responses were tabulated in Table 4 for each of the models. For the responses, the following terminologies were used:

Direct: This means that the model output increases by increasing the value of an input signal and decreases on decreasing the value.

Inverse: This means that a particular variable will affect the model output in an opposite manner.

Nil: This means that the model was unable to find any correlation between that particular variable and the model output.

Mixed: This means that the model has a different response for a different set of data of any particular variable.

<table>
<thead>
<tr>
<th>Sl. No.</th>
<th>Properties</th>
<th>Variable response</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Fe</td>
</tr>
<tr>
<td>1</td>
<td>Magnetic energy density ((BH)max)</td>
<td>Nil</td>
</tr>
<tr>
<td>2</td>
<td>Magnetic coercivity (Hc)</td>
<td>Mixed</td>
</tr>
<tr>
<td>3</td>
<td>Magnetic remanence (Br)</td>
<td>Mixed</td>
</tr>
<tr>
<td>4</td>
<td>Saturation magnetization (Ms)</td>
<td>Direct</td>
</tr>
<tr>
<td>5</td>
<td>Remanence magnetization (Mr)</td>
<td>Nil</td>
</tr>
<tr>
<td>6</td>
<td>((BH)_{\text{max}}/\text{mass})</td>
<td>Nil</td>
</tr>
<tr>
<td>7</td>
<td>Magnetic permeability (m)</td>
<td>Mixed</td>
</tr>
<tr>
<td>8</td>
<td>Cost of raw material (cost)</td>
<td>Inverse</td>
</tr>
<tr>
<td>9</td>
<td>Intrinsic coercive field (jHc)</td>
<td>Mixed</td>
</tr>
<tr>
<td>10</td>
<td>Density((\rho))</td>
<td>Mixed</td>
</tr>
</tbody>
</table>

Since, the dataset is quite noisy, we were left with a lot of mixed responses. A few important findings can be listed as follows:

1. Copper shows a direct response for \(H_c\) and \(B_r\), thus response surface predictions are at par with available literature. This has been discussed earlier in the text.
2. Hafnium shows a direct response for \(H_c\) and \(B_r\). Hf has not been previously used in Alnico alloys. Hence, further data-analysis is required before reaching a final conclusion.
Nickel shows response with \((BH)_{\text{max}}\). Hence, metamodeling can prove to be an asset for developing alloys in future as well as in predicting the properties of alloys with a new composition.

### Principal component analysis (PCA)

Principal component analysis was performed in order to determine correlations between variables and various properties by reducing the dimensionality of the dataset without losing much information. Here, each principal component (PC) is a linear combination of all the original descriptors (variables and properties). The first principal component (PC1) accounts for maximum variance in the dataset, followed by PC2 and so on. In all of these cases, PC1, PC2, and PC3 were able to capture most of the variance of the dataset.

To properly analyze the dataset for knowledge discovery, we divided the dataset into 4 types and did the PCA analysis accordingly.

Dataset was divided as follows:

1. Experimental: Alloy #1-80
2. Optimization: Alloy # 81-173
3. Data categorized on the basis of Multi-Criterion Decision Making (MCDM): 40 alloys were selected.
4. Whole dataset: Alloy # 1-173.

We used a statistical toolbox, "IBM SPSS" for this work.

1. Experimental: Alloy #1-80: These are the initial set of compositions predicted by Sobol’s algorithm. Hence, we did not perform PCA on the elements. Various properties were analyzed and it is reported below. Scree plots were plotted in order to determine the number of effective principal components required to represent the whole dataset. It was found that 2 PC’s are able to extract most of the information from the dataset. Figure 4 shows the scree plot for the properties while Figure 5 shows the position of various properties in the PC space.
It can be seen that $H_c$ and $jH_c$ coincide at the same spot. It makes sense as one is the inverse of the other. Similarly, $Mr$ and $Br$ can form a cluster and also $m$ and Density can be taken as another cluster. This means that properties that form a cluster are dependent on each other. Analysis of other datasets will further clarify these findings.

1. Optimization: Alloy # 81-173
In this data, we went for PC analysis for the elements. From scree plot in Figure 6, it was found that 3 PC’s are able to extract most of the information from the dataset. Figure 6 shows the scree plot for the elements while Figure 7 shows the position of various elements in the PC space.
Figure 6: Scree plot for PCA analysis: 3 PCA components were chosen.

Figure 7: Orientation of various elements in the PC space.

It can be seen that Cu and Hf seem to be part of a cluster. This means that there may exist Cu-Hf rich precipitates in the alloy. Since, Hf precipitates at the grain boundaries. Also from SVR analysis, both Cu and Hf showed a direct response for Hc and Br. Hence, this must be analyzed further before moving for microstructure analysis.

Additionally, Ni and Al too seem to be part of a cluster. This is quite evident in Alnico alloys.

From scree plot in Figure 8, it was found that 3 PC’s are able to extract most of the information from the dataset. Figure 8 shows the scree plot for the elements while Figure 9 shows the position of various properties in the PC space.
In figure 9, Hc and jHc are again coinciding. While, it can be seen that Br, Mr and (BH)max seems to be part of a cluster. Hence, these properties may be dependent on each other.

1. Data categorized on the basis of Multi-Criterion Decision Making (MCDM): 40 alloys were selected. Due to software limitations, we focused on optimizing (BH)max, Hc and Br only. While, we left the other properties of interest though they are quite important for the magnet. Hence, in this part, we selected 40 alloys on the basis of objective defined in Table 2. We used Multi-Criterion Decision Making methodology to select these alloys.
From figure 10, we can see that 3 PC’s were chosen. Figure 11 shows the orientation of various elements on the PC space.

Figure 11 supports our finding that is an occurrence of Cu-Hf cluster as well as Ni-Al cluster. To further clarify it, we will proceed towards analyzing the whole dataset.
Figure 12 shows scree plot for various properties while figure 13 shows the orientation of these properties in the PC space.

1. Whole dataset: Alloy # 1-173.

In this analysis, we used the complete dataset available with us. Figure 14 shows the plot for various elements. It can be seen that 3 PC’s are required to extract substantial information from the dataset. Figure 15 shows the orientation of various elements in the PC space.
In this set, we can see that Cu-Hf seems to be part of the cluster. Here, in PC1vsPC2, we can see that Ti can also be considered to be part of this cluster. Ni-Al too forms a cluster. Hence, we have sufficient information from the above analysis to move forward towards microstructure analysis. Figure 16 shows the scree plot for various properties while Figure 17 shows the orientation of various elements in the PC space.
PC analysis has been successfully used for materials discovery. Hence, one can test a new composition with the one available from a database to get an information regarding the property of interest. Hence, we did cluster analysis on the PC of our dataset. Here we used the whole dataset and marked the top 10 alloys on the basis of (BH)max values. It can be seen from Figure 18, that these superior alloys are clustered in a very small region while a majority of the PC space is covered by comparatively inferior alloys. Hence, if a certain composition is in the vicinity of these top 10 alloys, then they can be given a chance during the selection of alloys for experimental validation.
Figure 18 consists of all 173 alloys. Hence, it is a bit difficult to visualize. In this case, we used the dataset selected by MCDM and did PC analysis on it. Thereafter, we did cluster analysis on the dataset. Figure 19 shows the orientation of various alloys on the PC space.

7. Discussions

Figure 1, 2 and 3 shows the scatter plots of Magnetic energy density vs Magnetic coercivity and Magnetic remanence. Top 10 alloys are marked on the figure. Alloys were ranked on the basis of (BH)$_{max}$ values in Figure 1, 2 and 3 and Table 2. So far, the best alloy is alloy # 124 (predicted by MAIDROOC). Figure 1, 2 and 3
demonstrates the efficacy of the present approach as the Pareto-optimized alloys (modeFRONTIER and IOSO) dominates the initial 80 candidate alloys as well as most of those predicted by Sobol’s algorithm in later stages. The present alloy development time was about 1 year (less than currently 10 years). At the same time, in order to improve from the initial set of 80 alloys to the current best, timeframe is about 5 months. Hence, our approach was able to successfully recover from the initial flaws which would have been impossible by random experimentation. One peculiar finding in this work is that, we were able to achieve increase in \( H_c \) without compromising on \( B_r \).

In SVR, only Nickel shows some weak response for \((BH)_{\text{max}}\). Hence, there is scope for improvement in the response surface. Cu shows direct correlation with \( H_c \) and \( Br \) which can be confirmed from the literature. Additionally, Hf also shows positive correlation with \( H_c \) and \( Br \).

PCA analysis proved to be helpful in reducing the dimensionality of the dataset for visualization. Pca analysis points towards occurrence of Cu-Hf rich phase and Ni-Al rich phase. Ni-Al rich phase is known but Cu-Hf phase needed further investigation.

Above findings are quite helpful in development of knowledge base for development of new materials. At the same time, it has the potential to same time and money otherwise invested for random experimentation.

Effect of Hf and Nb rich phase needs to be further investigated in the following works.

At present, ab-initio based calculations as well as Calphad approach are effective for limited systems(3-4 elements) and cannot handle 8 elements [14]. Use of statistical tools will be helpful in determining the most influential alloying elements. This will be helpful in theoretical validation of the above findings. Additionally, one can work on finding the most stable phases needed for enhanced performance of these alloys by focussing on the most influential elements.

Hence, this work will be helpful in developing a knowledge base for the research community for development of hard magnetic alloys as well as provide a guideline for designing new alloys for desired properties.

Acknowledgement:

Authors would like to express their gratitude to Prof. Carlo Poloni, founder and president of ESTECO, for providing modeFRONTIER software free of charge for this project. This work was partially funded by the US Air Force Office of Scientific Research under grant FA9550-12-1-0440 monitored by Dr. Ali Sayir. The views and conclusions contained herein are those of the authors and should not be interpreted as necessarily representing the official policies or endorsements, either expressed or implied, of the US Air Force Office of Scientific Research or the U.S. Government. The U.S. Government is authorized to reproduce and distribute reprints for government purposes notwithstanding any copyright notation thereon.

8. References

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