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MULTI-OBJECTIVE DESIGN AND OPTIMIZATION OF HARD MAGNETIC ALLOYS FREE OF RARE EARTHS

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Abstract

This work demonstrates a novel approach to design and optimization of rare-earth free magnetic materials for targeted properties by effectively using various computational and statistical tools. From the open literature, we defined the alloying elements and bounds of their concentrations to develop a new system of Alnico alloys. Initial compositions of candidate alloys were generated using a quasi-random sequence generation algorithm. Response surface methodology approach was used to develop surrogate models to efficiently link alloy chemistry with desired macroscopic properties for these multi-component systems. The most accurate meta-models were used for multi-objective optimization of desired properties by utilizing various evolutionary approaches. Various statistical tools and pattern recognition techniques were used to determine patterns and correlations within the created dataset. Pareto-optimized candidate alloys were experimentally validated and used to improve the accuracy of the response surface generation used by the multi-objective optimizer to find the next generation of Pareto-optimal alloys. Results over the cycles show significant experimentally verified improvement in the properties of these alloys.

Keywords: magnetic materials,rare-earth elements, response surfaces, meta-models, multi-objective optimization, Pareto-optimized predictions

Introduction

For magnetic materials, it is important to address three basic bulk properties of interest: magnetic remanence (Br), magnetic coercivity (Hc) and magnetic energy density ($(BH)_{max}$). Hcis the ability of a magnet to withstand strong external magnetic field without demagnetizing. Br corresponds to the amount of magnetic flux density left in the magnet without demagnetizing. A high $(BH)_{max}$ means that one can synthesize smaller magnets while maintaining superior magnetic properties. Hence, one has to deal with conflicting objectives in order to address the problem of designing new magnetic materials.

Magnetism is a result of alignment of magnetic dipoles in a particular direction. This phenomenon is observed predominantly in two group of elements: 3d elements (Cr, Mn, Fe, Co, Ni) and 4f elements (Ce, Nd, Sm, Eu, Gd, Th, Dy, Ho, Er, Tm). 4f elements mentioned here belong to Lanthanides or are also known as Rare-Earth Elements (REE). REE based magnets have very high $(BH)_{max}$, Br and Hc. However, Nd-Fe-B magnets perform the best below 150 ºC. From 150 ºC to 350 ºC, Sm-Co magnets are used, but they are susceptible to corrosion. REE-based magnetic materials are currently essential in electric cars, in wind turbine electric

generators, and any high-efficiency electric devices requiring magnetic fields. 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 presence of an external magnetic field. However, low H_c also means that these magnets can be easily magnetized to saturation. A high Br 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 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 trends in the data. This information will be helpful to the research community in developing a material knowledgebase for manufacture of new alloys for targeted properties.

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].Sellmayer et al. [3, 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, differences between the theoretically calculated and the measured properties were quite large for $(BH)_{max}$ and H_c.

In order to develop a reliable knowledge base [6] for design of new alloys, one needs to focus on determining various correlations from the available limited size databases. 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 design of new alloys. It aims at reducing a new alloy development cycle from currently 10 years to two years or 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], and determining properties of Lutetium[17].

Alnico magnets were initially 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) $\alpha_1$and α_2. It was later observed that separation of $\alpha_1$and $\alpha_2$is due to a metallurgical phenomenon popularly known as "spinodal" decomposition where $\alpha_1$is Fe-Co rich ferromagnetic phase, while $\alpha_2$is Ni-Al rich phase. These phases are stable up to 850°C which is almost its Curie temperature. Above 850, Face Centered Cubic (FCC) phase begins to appear. This phase is quite detrimental for magnetic properties. In later years, modification of heat treatments and addition of other elements were made to stabilize the magnetic $\alpha_1$and $\alpha_2$phases, while eliminating or

reducing the amount of FCC γ phase. The recent rise in prices of rare earth elements led to a search for REEfree magnets. Currently, Alnico alloys contain 8+ elements. In our work, we used 8 alloying elements: Iron (Fe), Cobalt(Co), Nickel(Ni), Aluminum(Al), Titanium(Ti), Hafnium(Hf), Copper(Cu) and Niobium(Nb). It is important to know the role of these alloying elements initially for improving targeted properties of the alloys. Later, this information can be utilized to select meta-model for a certain property. Desired properties are listed in Table 1.

Table 1: Quantities to be simultaneously extremized using multi-objective optimization

	Properties	**Units**	**Objective**
1	Magnetic energy density ($(BH)_{max}$)	kg m^{-1} s^{-2}	Maximize
2	Magnetic coercivity (H_c)	Oersted	Maximize
3	Magnetic remanence (B_r)	Tesla	Maximize
4	Saturation magnetization (M_s)	Emu/g	Maximize
5	Remanence magnetization (M_r)	Emu/g	Maximize

Current Research on Combined Experimental/Computational Design of Alloys

In this work, we used computational tools to develop a novel approach for design optimization of high temperature, highintensity magnetic alloys.The steps involved in this work were:

1. Initial dataset:From the open literature, we defined the concentration 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 initial candidate alloys (see Table 2 and Table 3). The initial set of alloys was screened on the basis of limited knowledge of phase equilibrium and magnetic properties from a commercial thermodynamic database, Factsage[18].
2. Manufacture and testing: These80 alloys were manufactured and tested for various properties of interest as shown in Table 1.
3. Response surface generation: Response surfaces were developed from the experimentally evaluated 80 initial candidate alloys [19]. The response surfaces were tested using various accuracy measures and the most accurate one was chosen for further study.
4. Multi-objective optimization: The most accurate response surfaces were used to extremize the various properties (Table 1). Most of the optimization tasks yielded alloys with similar chemical compositions for a set of objectives. Thus, several optimization runs were performed to get a diverse pool of results. The optimization algorithms used in modeFRONTIER[19] were: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 to accelerate the optimization process using search algorithms such as NSGA2, MOPSO, MOSA).
5. This work was also carried out at three different locations using commercial optimization package IOSO developed by I.N. Egorov, a hybrid response surface generation developed by M.J. Colaco, and surrogate model selection algorithm developed by S. Chowdhury.

Predictions from both optimization packages (IOSO and modeFRONTIER) were merged and a set of Pareto-optimized alloys was selected for further manufacture and testing.

6. The work was performed in cycles. That is, Steps 2-5 were repeated until the improvements of multiple macroscopic properties of such magnetic alloys became negligible.

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

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

1. Data: In this work, our database was a combination of experimentally evaluated data predicted by a well-known random number generator and data from Pareto-optimized alloys.
2. Correlations: Here, we used various linear and nonlinear correlations, clustering, and principal component analysis tools to discover various trends in the dataset.
3. Theory: The above information can be coupled with theoretical knowledge to assist alloy designers.

Table 2: Concentration bounds of AlNiCo type alloys

	Alloying elements	Concentrtion bounds (Wt %)	
		Alloys' numbers	
		1 – 85	**86 – 173**
1	Cobalt	24 – 40	24 – 38
2	Nickel	13 – 15	13 – 15
3	Aluminum	7 – 9	7 – 12
4	Titanium	0.1 – 8	4 – 11
5	Hafnium	0.1 – 8	0.1 – 3
6	Copper	0 – 6	0 – 3
7	Niobium	0 – 2	0 – 1
8	Iron	Balance to 100	

Table 3: Design cycle - alloy numbers

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

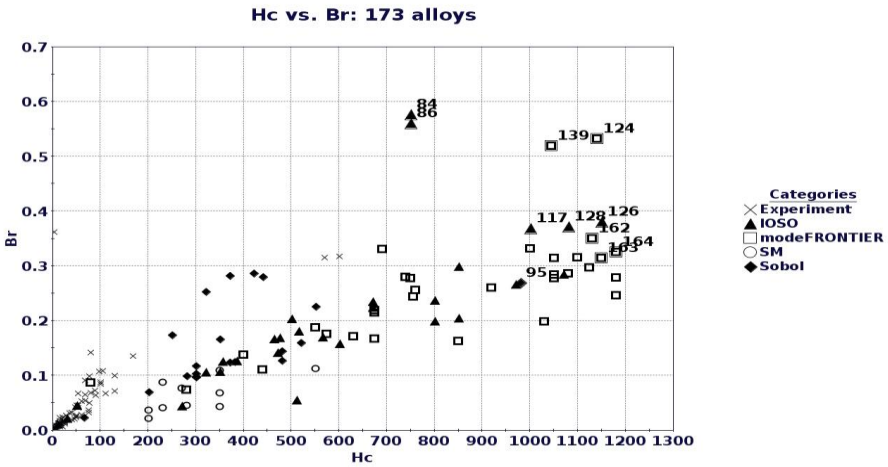

Figure 1: Scatter plot of Br vs. Hc showing significant performance improvements over the initial set of candidate alloys achieved using the design optimization of alloys.

At present, we have worked through 11 cycles of design and optimization followed by experimental validation. Tables 2 and 3 list alloys manufactured in each of the cycles and the best alloy in each cycle. Figures 1 and 2 show the interactions between $(BH)_{max}$,Hc and Br. Candidate alloys have been marked with different legends in order to demonstrate the efficacy of

design and optimization. Top 10 alloys (ranked on the basis of $(BH)_{max}$) have been marked in both figures.

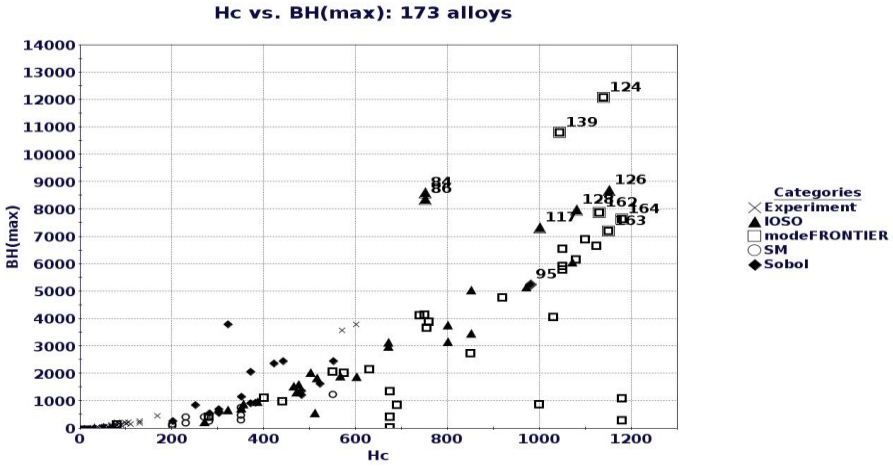

Figure 2: Scatter plot of $(BH)_{max}$ vs. Hcshowing significant performance improvements over the initial set of candidate alloys achieved using the design optimization of alloys.

Sensitivity analysiswas also performed in order to determine the relationships and to find various trends and patterns within the dataset. Meta-models were selected on the basis of several accuracy measures. In this work, we are focusing on one of the methods used to determine the response of various elements on desired properties of interest.

Single Variable Response:

In SVR testing, an 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 terminology was used:

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

Inv: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.

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

Table 4: Single variable response for various properties

Sl. No.	Properties	Variable response							
		Fe	Co	Ni	Al	Ti	Hf	Cu	Nb
1	Magnetic energy density ((BH)max)	Nil	Nil	Mix	Nil	Nil	Nil	Nil	Nil
2	Magnetic coercivity (Hc)	Mix	Mix	Mix	Inv	Mix	Dir	Dir	Mix
3	Magnetic remanence (Br)	Mix	Mix	Mix	Inv	Mix	Dir	Dir	Inv
4	Saturation magnetization (Ms)	Dir	Inv	Dir	Mix	Inv	Dir	Mix	Mix
5	Remanence magnetization (Mr)	Nil	Nil	Nil	Nil	Nil	Nil	Nil	Nil

Since, the dataset is quite noisy, we were left with a lot of mixed responses. 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. Hence, we can move forward to another important aspect in magnetism: microstructures.

Microstructure analysis

From the above statistical analysis, we moved towards experimental validation of one of the findings.. Figure 3 shows the optical micrograph of alloy # 95.

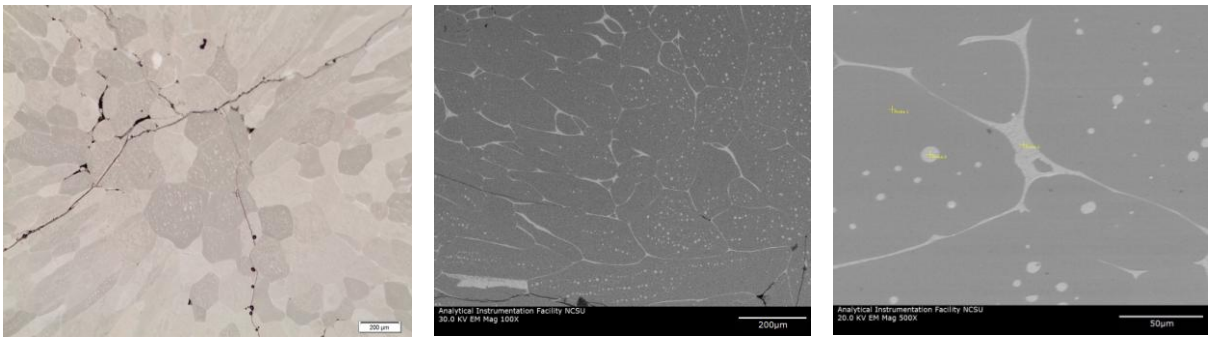

Figure 3: Alloy # 95 - Optical micrograph showing white spots (left), BSE image (middle)and SEM micrograph (right), showing white spots and spots to be analyzed by EDS.

Figure 3 shows a number of white spots. In Alnico 8 and 9 it is a well-known fact that Ti and Cu form precipitate that helps in refining the alloy from impurity. At the same time, Cu precipitates help in separation of magnetic phases and help in increase ofcoercivity and remanence to some extent. Figure 3 also shows the electron micrograph of alloy # 95. White spots are still visible. Backscattered electron (BSE) images were used for composition mapping. Table 5 shows the result from BSE images. Composition is homogeneous and close to the nominal composition.

Since the nominal composition is at par with the composition from BSE imaging, we moved one step further. We chose three spots: grain, grain boundary, and the white spot. Thereafter, we performed Electron Dispersive Spectroscopy (EDS) analysis of these spots for further information. Figure 3 shows the SEM micrograph and the spots that were analyzed. Table 6 shows the results from EDS analysis.

Table 5: Composition mapping by BSE imaging.

Name	Fe	Co	Ni	Al	Ti	Hf	Cu	Nb	C	O
Nominal composition	32.36	36.86	13.54	7.20	4.11	2.07	2.94	0.93	0	0
Whole image	32.09	35.64	11.99	8.76	5.21	0.04	2.56	1.32	1.54	0.86

EDS analysis confirms precipitation of Hf at the grain boundary and on white precipitates. It can also be seen that the concentration of Cu and Ti is slightly higher on GB and white spot than grain. Another interesting finding is Nb rich phase at GB and white precipitates.

Table 6: Results from EDS analysis for alloy # 95

Name	Fe	Co	Ni	Al	Ti	Hf	Cu	Nb	C	O
Nominal composition	32.359	36.857	13.545	7.200	4.116	2.068	2.938	0.931	0	0
Point 1	32.5	35.45	14.32	9.67	4.3	0	3.32	0.44	0	0
Point 2	23.88	34.79	14.99	6.85	4.78	7.95	3.77	2.96	0.03	0
Point 3	25.78	34.49	14.43	8.88	4.92	5.89	3.42	2.17	0.03	0

Discussions

Top 10 alloys are marked in Figures 1 and 2 based on their (BH) values. So far, the best alloy is alloy # 124 (predicted by MAIDROC). Figure 1 and 2 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 the initial set of 80 alloys to the current best, the 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 an increase in Hc without compromising on Br.

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

Microstructure analysis confirms the formation of precipitates. BSE images show homogeneous composition, but white spots are still needed some clarification. EDS analysis of white spots and GB confirmed presence on Cu-Hf precipitates. Hence, SVR analysis proved to be helpful in determining the formation of precipitates.Above findings are quite helpful in the development of knowledge base for design of new materials. At the same time, it has the potential to save time and money otherwise invested for random experimentation.

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