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Combined machine learning and CALPHAD approach for discovering processing-structure relationships in soft magnetic alloys

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ABSTRACT

FINEMET alloys have desirable soft magnetic properties due to the presence of Fe_3Si nanocrystals with specific size and volume fraction. To guide future design of these alloys, we investigate relationships between select processing parameters (composition, temperature, annealing time) and structural parameters (mean radius and volume fraction) of the Fe_3Si domains. We present a combined CALPHAD and machine learning approach leading to well-calibrated metamodels able to predict structural parameters quickly and accurately for any desired inputs. To generate data, we have used a known precipitation model to perform annealing simulations at several temperatures, for varying Fe and Si concentrations. Thereafter, we used the data to develop metamodels for mean radius and volume fraction via the *k*-Nearest Neighbour algorithm. The metamodels reproduce closely the results from the precipitation model over the entire annealing timescale. Our analysis via parallel coordinate charts shows the effect of composition, temperature, and annealing time, and helps identify combinations thereof that lead to the desired mean radius and volume fraction for nanocrystals. This work contributes to understanding the linkages between processing parameters and microstructural characteristics responsible for achieving targeted properties, and illustrates ways to reduce the time from alloy discovery to deployment.

1. Introduction

FINEMET alloys belong to a class of soft magnetic alloys based on the Fe-Si-Nb-B-Cu system [1]. In comparison with other soft magnets, FINEMET alloys possess high saturation magnetization [1] and high permeability [2–5], low core loss [1–3,5], low magnetostriction [1–3,5,6], excellent temperature characteristics, small aging effects, and excellent high frequency characteristics [1–3,5]. As a result, FINEMET alloys have been successfully used in a number of applications including choke coils [1,2,7–9], mobile phones [2], noise reduction devices [2], computer hard disks [2], and transformers [1–3,8,9]. Superior soft magnetic properties are attributed to the nanocrystalline α''-(Fe, Si) phase (Fe_3Si with D03 structure) in the size range of 10−15 nm diameter (radius 5−7.5 nm) and 0.7 volume fraction [1–6,8–16]. Since its discovery, researchers have investigated FINEMET alloys to improve upon multiple soft magnetic properties by performing experiments followed by characterization using advanced diagnostic tools [2,4,5,8,10–12,14–16].

In materials design, understanding the various processing-structure-property (PSP) linkages plays an important role in designing advanced materials. In particular, correlations between microstructure and desired properties [17–20], are essential for the deployment of new materials into service. In addition, composition variations and processing parameters (e.g., heat treatment schedule) play an integral role in modeling the microstructure(s) responsible for achieving desired properties, where optimizing processing parameters along with composition remains a challenging task [21]. As an alternative to costly experimentation, the CALPHAD approach allows for investigating the effect of composition variations and heat treatment on the size distribution and volume fraction of the phase(s) that are responsible for optimal or desired properties; indeed, it has been used for studying soft magnets containing amorphous phases [22–24] using the commercial software Thermocalc [25]. Recent studies indicate that simulations based on CALPHAD [26,27] are in need of efficiency improvements if they are to be used for optimization of the composition and heat treatment schedule. To address this challenge, it is important to develop models that can both replicate maximum information available from prior studies and, in addition, demonstrate effectiveness in optimizing the processing protocol. This effectiveness should not come, for example, from repeating the same calculations at different compositions,

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but rather from learning the results obtained in several selected cases in order to predict the behaviour at other compositions.

Machine learning approaches have been used to help reduce the time required in the alloy design process [28–38]. Supervised machine learning approaches such as artificial neural networks [37–40], *k*-Nearest Neighbour algorithm (*k*-NN) [41,38], genetic programming [37,38,40,42], kriging [43,44], and unsupervised approaches such as Principal Component Analysis (PCA) [30,31,35], Hierarchical Clustering Analysis (HCA) [29,31,34], and Self Organizing Maps (SOM) [28] have been previously used in various areas of materials science and can also be helpful in this case. From an implementation point of view, there exist several open-source software packages to develop response surfaces or metamodels using several different concepts from artificial intelligence. A machine learning model based on results from the CALPHAD approach will serve as an important rapid screening tool before performing experiments; it also helps predict outcomes in case of uncertainties in the composition of the material or in furnace temperature during annealing.

In this article, we present a combined CALPHAD-machine learning approach for optimizing composition along with processing parameters for FINEMET alloys by developing metamodels (response surfaces, or surrogate models) for the simulated crystallization of Fe_3Si domains. We have acquired data for mean radius and volume fraction of Fe_3Si nanocrystals through a recently developed precipitation model [45] in Thermocalc [25], capable of simulating the nucleation and growth of Fe_3Si nanocrystals from an amorphous phase. Thereafter, we have used a *k*-NN algorithm to generate computationally inexpensive metamodels to replace exhaustive Thermocalc modeling without any significant loss of accuracy. This way, we are able to demonstrate the efficacy of our combined CALPHAD-machine learning approach by predicting compositions and processing parameters that would lead to achieving the desired mean radius and volume fraction of Fe_3Si nanocrystals. The developed metamodels capture the established nucleation and growth evolution [46,47] within the CALPHAD approach, for the entire annealing timescale and for compositions and parameters that were not included in the training set. Another important observation is that the metamodels can predict outcomes in a fraction of the time taken by simulations performed in Thermocalc [48]. Lastly, we propose Parallel Coordinates Charts (PCC) [49] for comprehensive visualization of the relationships between processing parameters and optimized quantities, and for rapid identification of the parameters that lead to crystallization of Fe_3Si nanocrystals in the desired size range and volume fraction. Our proposed approach helps reduce the alloy development time since it can serve as a tool for rapid screening of the multi-dimensional parameter space before performing experiments. As such, this combined machine learning and CALPHAD approach illustrates a case of addressing the challenge of simultaneously determining the effect of composition variation and processing parameters [17–21] on the microstructure of FINEMET, and can also be extended to other nanocrystalline alloys.

2. Methods

Fig. 1 shows the schematic flowchart of the process we followed in order to develop our combined CALPHAD-machine learning approach for optimization of nanocrystal size and volume fraction. This approach is enabled by a nucleation and growth model (precipitation model) in Thermocalc [50], recently parameterized for FINEMET [45]. We used this model to generate data for mean radius and volume fraction of Fe_3Si nanocrystals grown upon annealing the amorphous material, data which serves as a training set for developing metamodels. Analysis of the results created by the metamodels reveals correlations between the input parameters (composition, temperature, and time) and the optimized quantities. The three aspects of this work (Fig. 1) are described in some detail below.

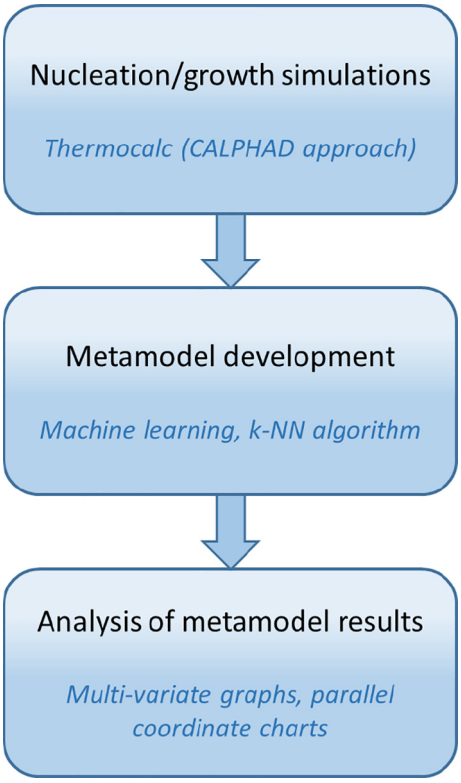

Fig. 1. Flowchart of steps followed in this work for a class of FINEMET alloy with composition $Fe_{72.89}Si_{16.21}B_{6.9}Nb_3Cu_1$

2.1. Generating data for developing a metamodel

To generate mean radius and volume fraction data [55], we have used the TC-PRISMA [50] module in Thermocalc, which relies on thermodynamic (TCFE8) [48] and mobility [51] databases. TC-PRISMA [50] uses the Kampmann-Wagner Numerical (KWN) method [46,47] for simulating nucleation and growth of precipitates during annealing. The KWN method is an extension of the Langer-Schwartz approach [52] and its modified form [53]. To use the precipitation model, several input quantities in TC-PRISMA [50] were previously parameterized [45] so that the precipitation model simulates specifically the nucleation and growth of Fe_3Si nanocrystals during annealing. The FINEMET base composition is $Fe_{82.35}Si_{9.21}B_{1.51}Nb_{5.64}Cu_{1.29}$ in weight %, or $Fe_{72.89}Si_{16.21}B_{6.90}Nb_3Cu_1$ in atomic %; we will refer only to the latter in the remainder of the article. Simulations of precipitation were performed for new compositions $Fe_{72.89+x}Si_{16.21-x}B_{6.90}Nb_3Cu_1$ generated by varying the content of Fe and Si by x ($-3 \leqslant x \leqslant 3$). Isothermal annealing was carried out at a set of temperatures between 490 °C and 550 °C in (increments of 10 °C) to for up to 2 h holding time. We obtained a significant amount of Thermocalc data for mean radius and volume fraction of Fe_3Si nanocrystals [55], which serves as training set for the machine learning stage of the workflow (Fig. 1).

2.2. k-Nearest Neighbour Algorithm

The mean radius and volume fraction were generated in order to be used for creating response surfaces or metamodels to help in the design of future nanocrystalline FINEMET alloys. We use the *k*-NN algorithm [41] as implemented in the software modeFRONTIER [54] to construct the metamodels. This algorithms stores all the available information and predicts a new output (in this case, mean radius and volume fraction) based on a measure of similarity (distance function) of the new input with the stored cases. Specifically, to predict the new target/output that corresponds to a new input, the straightforward approach is