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Research Paper

Prediction of Heusler Alloys with Giant Magnetocaloric Effect using Machine-Learning

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Abstract
Heusler alloys are intermetallic that offer a unique and broad array of properties. These properties are both scientifically intriguing and valuable for a variety of beneficial practical applications. One of these applications is magnetic cooling, taking advantage of the giant magnetocaloric effect (GMCE) in some Heusler alloys. Since the late 1990s, numerous scientific papers were published, attempting to harness Heusler alloys for green refrigeration. Manufacturing the alloys by additive manufacturing further offers control and enables tuning of their properties by controlling their microstructure. Although the scientific literature contains extensive information on these alloys' chemistry and performance, it is the massive volume of scientific papers that makes it difficult, if not impossible, to keep up to date with relevant discoveries. To enable predicting the composition of excellent performing giant magnetocaloric Heusler alloys, manufactured by laser powder bed fusion (LPBF), we employed artificial intelligence, specifically unsupervised learning in the current work. We trained an unsupervised learning model using word embedding and the Word2vec algorithm on different data sets in the literature to extract hidden knowledge, relations, and interactions based on words that appear in similar contexts in the text while often having similar meanings. Properties inherent to giant magnetocaloric materials were addressed in the model. The outcome was the prediction of Heusler alloys, manufactured by LPBF, with an excellent giant magnetocaloric effect.

Keywords
Giant Magnetoresistance, Heusler Alloys, LPBF, Machine Learning, Word2Vec

1. Introduction

The intermetallic Heusler alloys, first discovered by Friedrich Heusler over 100 years ago [1], have demonstrated a wide range of unique and highly desirable properties. Some of this unique combination of properties may include ferromagnetism [2] (sometimes even when the alloy is made of non-magnetic elements [3]), antiferromagnetism [4], ferrimagnetism [5], half-metallicity [6], giant anomalous Hall effect [7], and superconductivity [8]. Ternary Heusler alloys can be full Heusler (X_2YZ) ~~and~~ or half Heusler (XYZ), where X and Y are transition metals and Z is in the p-block of the periodic table. Their unique combination of properties and their versatility make them suitable for a wide range of applications. Some of these applications include solar energy and thermoelectric conversion [9,10], applications in spintronics [11,12], as superconductors [13,14], topological insulators [15,16], and giant magnetocaloric effect materials [17,18]. Their properties can be further tuned by non-stoichiometry and chemical substitution with a 4th element [19]. The structure of a full Heusler alloy is shown in Figure 1.

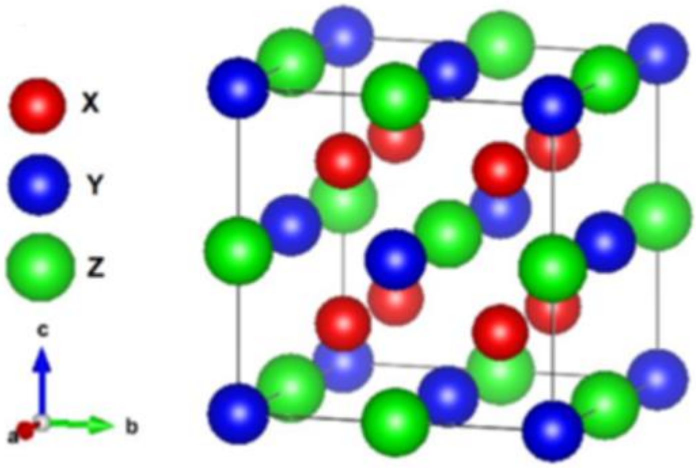

Figure 1. Crystal structures for austenite in $Ni_{50}Mn_{25}Ga_{25}$ Heusler alloy

The discovery, around the turn of the Century, of elements and alloys that demonstrate the 'giant' magnetocaloric effect (GMCE) [20] has generated an interest in magnetic refrigeration technology as a favorable candidate to replace conventional vapor-compression refrigeration. Magnetic refrigeration offers environmentally friendly, substantially higher cooling efficiency, and compactness. Numerous studies have followed ever since, and the quest for better-performing magnetocaloric materials continues. Heusler alloys are some of the most promising GMCE materials, due to their magnetostructural coupling [21]. There is a wealth of scientific information in the literature about materials that demonstrate a giant magnetocaloric effect. These materials were developed based on scientific principles, combined with experimental trial and error. The development of excellent magnetocaloric Heusler alloys must be built on thorough existing knowledge of successful alloys. However, the enormous volume of scientific papers that are produced each year and the rate of its growth make it difficult, if not impossible, to keep up to date with relevant scientific discoveries. This massive amount of information also makes it complicated to find implicit and hidden connections, relationships, and dependencies within the information that may guide the direction of future research or lead to valuable new insights. To overcome these hurdles, we employed artificial intelligence, specifically unsupervised learning, in our current work on the discovery of Heusler alloys, manufactured by LPBF and demonstrating a giant magnetocaloric effect.

2. Method

Guided by the approach of Tshitoyan et. al. in "Unsupervised word embeddings capture latent knowledge from materials science literature" [22], we used word embedding for our implementation and trained unsupervised word-embedding models on different data sets in materials science to extract hidden knowledge, relations, and interactions based on words that appear in similar contexts in the text while often having similar meanings. Properties inherent to giant magnetocaloric materials are addressed in the model.

Word embedding is one of the most important versatile examples of unsupervised learning methods. It is a representation of words obtained from a large unlabeled corpus, by embedding both semantic and syntactic meanings. It is commonly used in tasks of modern natural language processing (NLP). Individual words are represented as real-valued vectors, often comprising tens or hundreds of dimensions, in a predefined vector space. Each word is mapped to one vector, then a neural network is used to produce the vectors. In the project, we used the Word2Vec word embedding method (see Figure 2), which is based on a shallow neural network with two layers that takes a large corpus of text as its input. It produces a vector space, typically of several hundred dimensions, with remarkable linear relationships called analogies. These allow for math operations such as vec("king") - vec("man") + vec("woman") ≈ vec("queen"). It comprises two techniques, CBOW (continuous bag of words) and Skip-gram.

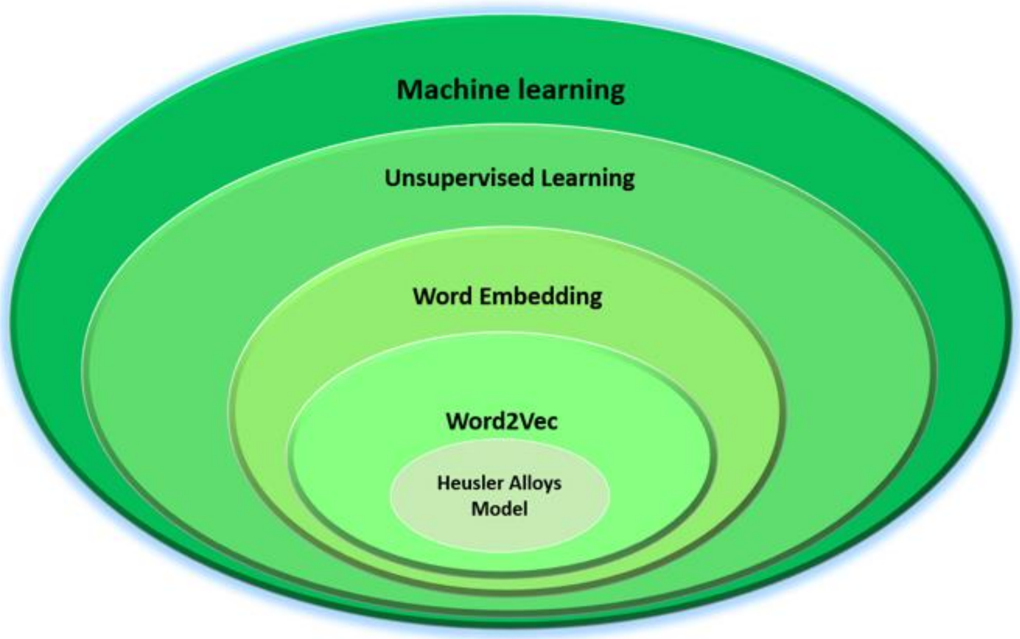

Figure 2. Word2Vec was used to discover Heusler alloys

Through Science Direct application programming interfaces (APIs) (https://dev.elsevier.com/), interactive APIs, and some Python (3.6) code [23], we performed the text mining process. The first step has consisted of data cleaning. Starting from the uncleaned data, words longer than 20 letters were deleted. Then all uppercase characters in the corpus were converted to lowercase. Finally, stop words and punctuation were deleted. The next stage was training the model.

2.1 Word2Vec Training

We used a combination of the Word2Vec implementation in genism [24], and the open-source code (with a few modifications) used by Vahe Tshitoyan et al. [22], to train our model. We considered only vocabulary that occurs more than five times because words that appear less than five times in a billion-word corpus are likely typos and garbage; also, there is not enough knowledge they can provide. The vector size, or size of the embedding, in our model, was 200. We used Skip-gram in the Google code [25] because it provides better results (at the cost of speed). Subsequent analysis was based on similarities and analogies.

3. Results and discussion

The research focus was on Heusler alloys demonstrating Giant Magnetocaloric Effect (GMCE) as observed in some of the Ni-Mn-X-based Heusler alloys. The alloys must be able to be manufactured by LPBF to allow for good control of structure and properties. In the quest to predict the best alloys, we considered only alloys that demonstrate a merged magneto-structural phase transition, as that is when the GMCE will be largest [26,27]. The magnetic entropy change is another property to watch for and was considered in the work.

Ternary Ni-Mn-Ga is one of the best-known magnetocaloric materials [28]. Other Ni-Mn-based Heusler alloys have shown good promise as well [29]. In the quest to discover other magnetocaloric alloys, we used main words, such as giant magnetocaloric effect, Ni-Mn-based Heusler alloys, magnetic entropy change, and magneto structural transition as seeds to our model to find new candidate alloys. We queried our model to find the most similar words for these keywords. Because this resulted in many different alloys, properties, and parameters, we tightened the search by making the keywords more specific. We studied some intersections between the word similarities, searching for alloys that occur simultaneously within the similar words for both “giant magnetocaloric effect” and “Ni-Mn-based Heusler alloys.” As shown in Figure 3, mnfepge_compounds, ni-mn-in-co, fe49rh51, nicomnsn, mncoge, ni50mn35in15_heusler, nimnsn, ni43mn46sn11, lafesi13based, mnas1-xsbx, gd5ge2si2, mnassb, tb5si2ge2, erco2, mnnige, mnfe2psi, gd5si2ge2, nazn13type, mnfep1xasx, ni-co-mn-in, mnfepsi, ni-co-mn-sb, and nimnz are in the intersection of similarities considered. This introduces them as predicted candidates.

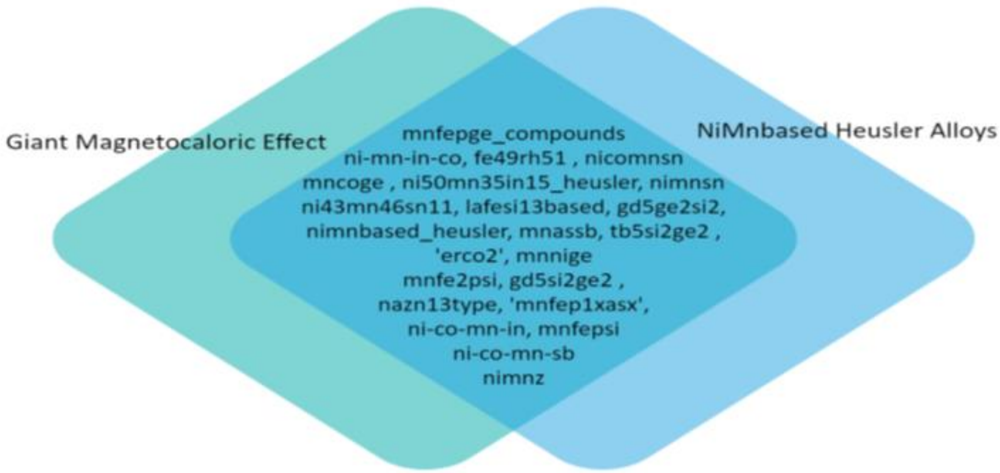

Figure 3. The intersection of the similarities for both “Giant Magnetocaloric Effect” and “Ni-Mn-Based Heusler Alloys”

Other intersections between the similarities that were obtained, such as the intersection of the similar words between “magnetic entropy change” and “nimnbased heusler alloys” are presented in Figure. 4a, and the intersection of the similar words between “magnetic entropy change” and “giant magnetocaloric effect” in Figure. 4b.

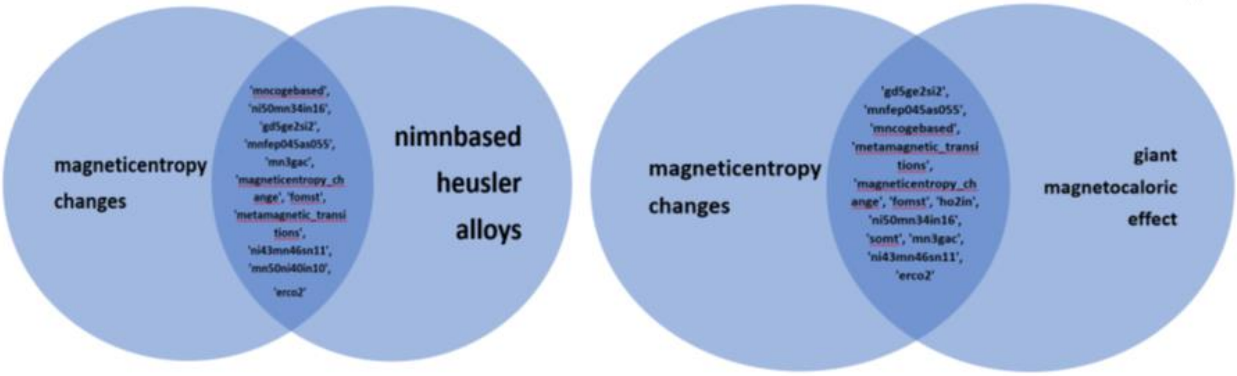

Figure 4. Intersections of similarities (a) Magnetic Entropy Change and Ni-Mn-Based Heusler Alloys; (b) Magnetic Entropy Change and Giant Magnetocaloric Effect

To find the top candidate alloys, the intersection between the most dominant factors in the GMCE was obtained (Figure 5). Because mnfep045as055, erco2, ni43mn46sn11, mncogebased, gd5ge2si2, mn3gac, and ni50mn34in16 are the alloys that occur within the similar words for Ni-Mn-based Heusler alloys, magnetic entropy changes, GMCE, and magnetostructural transition, they could be considered as top candidates. Alloy $MnFeP_{0.45}As_{0.55}$, as compared to well-known magnetic alloys, such as the gadolinium alloy, is thought to be a promising magnetic material with improved magnetic properties [30]. $ErCo_2$ shows a large magnetocaloric effect, suggesting a high potential for a working substance of magnetic refrigeration at 30-50 K [31]. In addition, ni43mn46sn11 and ni50mn34in16 are alloyed with behavior that allows the magnetic-field-driven transition from martensitic phase with low magnetization to austenite with high magnetization, which results in a GMCE property [32]. MnCoGe-based alloys have a strong interplay between structure and magnetism, which results in the exhibited GMCE [21]. It further emerged that gd5ge2si2 is a very good alloy for application as an active regenerator material in room temperature magnetic refrigerators because of the GMCE with a transition temperature at around 276 K [33]. All these alloys have not been produced by LPBF processes (as of the date of writing). Thus, they may be good candidate materials to be made by LPBF, with good GMCE.

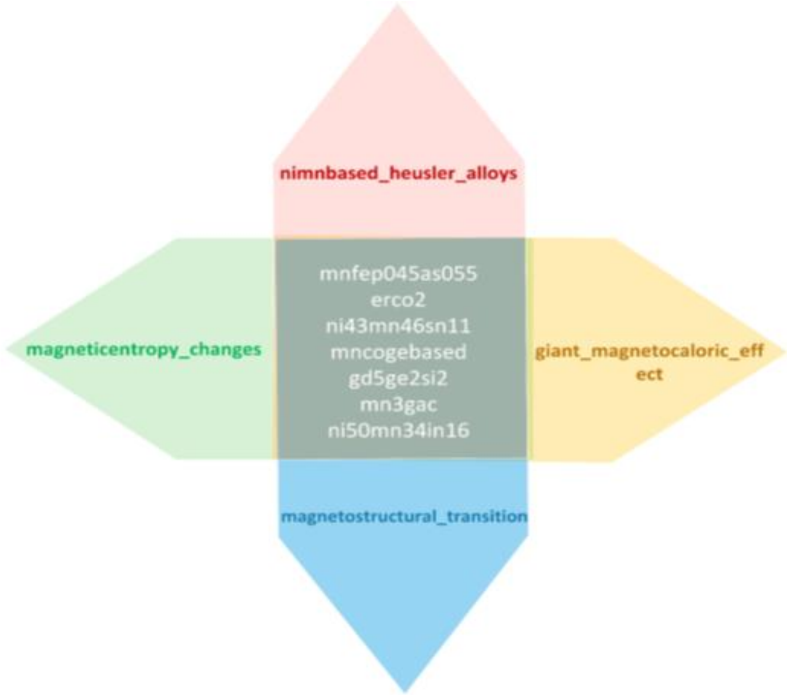

Figure 5. The intersection between Ni-Mn-Based Heusler alloys, magnetic entropy changes, GMCE, and magnetostructural transition

4. Conclusions

In this work, we trained several unsupervised learning models using word embeddings and the Word2Vec algorithm with information from additive manufacturing literature focusing on LPBF processes, GMCE, and Ni-Mn-based Heusler alloys. We trained an unsupervised word embedding model (`AMW2V'), using Word2Vec primarily focused on Ni-Mn-based alloys, selective laser melting (SLM), selective laser sintering (SLS), and direct metal laser sintering (DMLS), in the literature from 2012 to 2022 from Science Direct at Elsevier, to search for candidate alloys with GMCE to be produced with LPBF processes. AMW2V predicted several candidates, such as mnfep045as055, erco2, ni43mn46sn11, mncogebased, gd5ge2si2, mn3gac, and ni50mn34in16, according to their similarities with the given Ni-Mn-based Heusler alloys exhibiting GMCE properties based on magnetic entropy changes and magnetostructural transition. The research supports a direction of producing candidate alloys using LPBF with optimum process parameter combinations, including laser power, scanning speed, hatch spacing, and layer thickness, allowing manufacturers to accurately produce parts with complex shapes and intricate features from the candidate alloys.

6. Acknowledgment

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