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研究課題名（和文）Query-and-Learn Machine Learning framework to model the stability mechanism of REFe12 magnets

研究課題名（英文）Query-and-Learn Machine Learning framework to model the stability mechanism of REFe12 magnets

研究代表者

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研究成果の概要（和文）：REFe磁石を研究するための機械学習フレームワークを構築することを目指して、以下の3つの主な目標を達成した：(1)ThMn12型磁石と様々な置換元素を含むSmFe12の量子計算を用いて、材料発見のためのデータクエリの量子計算コストを最適化するアクティブラーニングベースのフレームワークを開発した。(2)Sm-Fe族の結晶構造とその安定性の関係を探るため、多目的結晶構造探索を最適化する進化的アルゴリズムベースのフレームワークを開発した。(3)物理的特性に基づく材料の類似性尺度を開発し、この方法ではデンプスター・シェーファー証拠理論に基づいた類似性測定に不確実性が組み込まれた。

研究成果の学術的意義や社会的意義
This work contributes to the research of rare-earth iron-based magnetic materials by (1) finding optimal materials composition of high-performance magnet discovery and (2) introducing Machine Learning frameworks with standardized materials discovery process and results interpretation.

研究成果の概要（英文）：This research aims to build a query-and-learn Machine Learning frameworks to investigate the stability mechanism of REFe magnets with RE as rare-earth element. There are three main goals were achieved. Firstly, this research developed an active learning-based framework to optimize the quantum data querying cost in materials discovery. Quantum calculations for SmFe12 with ThMn12-type magnets with various substitution elements were systematically investigated. Secondly, this research developed an evolutionary algorithm-based framework to optimize multi-objective crystal structure search. The crystal structure-stability relationship of multiple Sm-Fe families was investigated. Lastly, this research developed an evidence-based similarity measure for materials regarding physical property. The method incorporates measurement uncertainty into the similarity measure under the Dempster-Shafer evidence theory

研究分野：Materials Informatics

キーワード：materials discovery　structure prediction　rare-earths magnets　Dempster-Shafer theory　Sm-Fe magnets

## 1. Background at the beginning of the research

**Research and development of strongest magnets:** Nowadays, there is a vast of utilities requiring high-performance magnet components such as automotive vehicles or electronic devices. While the global needs for magnetic materials have been increasing for years [1], only a few rare-earth-based materials are widely accepted due to requirements about balancing between thermal, stability, and magnetic properties. The most widely accepted material is the Nd-Fe-B magnets, which was developed in the middle 80s [2]. In searching for the next generation of permanent magnets, the iron-rich ternary compounds $REFe_{12}$ (RE: rare-earth elements) with the original tetragonal $ThMn_{12}$ structure discovered in the late 80's [3–5] have been considered a potential structure family. However, the binary $REFe_{12}$ is *not stable* and stabilizing elements such as Ti, V, Cr, Mo, W or Ga must be alloyed for obtaining the $ThMn_{12}$-type phase [6-7]. In 2017, $Sm(Fe_{0.8}Co_{0.2})_{12}$ thin films has been reported [8] with excellent hard magnetic properties with high spontaneous magnetization at 1.78T (1.90T with partial substitution of Sm with Zr reported in 2019 [9]), anisotropy field at 12T and high Curie temperature at 859K. In Figure 1, comparing with existed materials, the $Sm(Fe_{0.8}Co_{0.2})_{12}$ thin film shows out-performance and then become promising as permanent magnet material, if it can be stabilized in a bulk form [8]. Therefore, in finding new magnets of the $REFe_{12}$ system, *modeling mechanism to stabilize structures* in this family is considered as the most important task.

**Research of modeling stability mechanism:** There is a large number of researches that focus intensively to capture the structure-stability mechanism of $REFe_{12}$-substituted structures by observing correlations such as preferential substitution sites [8, 10-13], thermal sub-phases decomposition [14], 3D atom maps and composition profile [15], semantic diagram of structural relations between $REFe_5$-$REFe_{12}$-$RE_2Fe_{17}$ [12]. Besides that, substitution induces other magnetic properties that were carefully investigated, such as spin-wave dispersion with Co substitution [16]; V substitution induces a rapid change of Curie temperature [17], crystal electric field effect with Ti substitution [18]. These observed correlations lead researchers to have insights about the key-driving mechanisms of structural stability. Higher efficiency policies in designing new candidates are then formulated based on these observations. We assume that by introducing a more concisely viewpoint of structure–stability correlations, e.g., a classification map of structure deformation between optimal substituted structures and the original $ThMn_{12}$ structure, more *productive substitution policies* to accelerate the success rate of finding bulk stable $REFe_{12}$-substituted structure will be reliable.

## 2. Purpose of research

This research aims to (1) *model stability mechanism* of REFe12-substituted structures and (2) utilize extracted correlations to accelerate *finding optimal stability* REFe12-substituted structures by proposing a *query-and-learn* process that comprises *first-principle calculation* and *Interpretable Machine learning* methods.

## 3. Research method

(1) We proposed a query-and-learn active learning approach combined with first-principles calculations to rapidly search for potentially stable crystal structure via elemental substitution, to clarify their stabilization mechanism, and integrate this approach to $SmFe_{12}$-based compounds with $ThMn_{12}$ structure, which exhibits prominent magnetic properties. Using the optimized structures and formation energies obtained from the first-principles calculations after each active learning cycle, we construct an embedded two-dimensional space to rationally visualize the set of all the calculated and not-yet-calculated structures for monitoring the progress of the search. by quantitatively measuring the change in the structures before and after optimization using OFM descriptors, the correlations between the coordination number of substitution sites and the resulting formation energy are revealed. The negative formation energy family $SmFe_{12-\alpha-\beta}[Al/Ga]_\alpha Y_\beta$ structures show a common trend of increasing coordination number at substituted sites, whereas structures with positive formation energy show a corresponding decreasing trend [18].

(2) We proposed a machine learning-aided genetic algorithm structure generation to investigate the space of possible geometrical arrangement given a chemical composition formula. In parallel with structure generation inherited using the USPEX program, a pool of structures is created for every population using the sub-symmetry perturbation method. A framework using embedded orbital field matrix representation as structure fingerprint and Gaussian process as a predictor has been applied to ranking the most potential stability structures [19].

(3) We proposed an application of the Dempster–Shafer theory to develop an evidential

regression-based similarity measurement method, which can rationally transform data into evidence. It then combines such evidence to conclude the similarities between materials, considering their physical properties [20].

**4. Research result**

There are three objectives were achieved:

(1) Developing an active learning-based framework to optimize the quantum calculation cost of querying data in materials discovery. Quantum calculations for $SmFe_{12}$ with $ThMn_{12}$-type magnets with various substitution elements were systematically investigated. Our machine learning model with an embedding representation attained a prediction error for the formation energy of $1.25 \times 10^{-2}$ (eV/atom) and required only one-sixth of the training data compared to other learning methods. Moreover, the time required to recall most potentially stable structures was nearly four times faster than the random search. The formation energy landscape visualized using the embedding representation revealed that the substitutions of Al and Ga have the highest potential to stabilize the $SmFe_{12}$ structure. In particular, $SmFe_9[Al/Ga]_2Ti$ showed the highest stability among the investigated structures [18].

(2) Developing an evolutionary algorithm-based framework to optimize multi-objective crystal structure search. The crystal structure-stability relationship of multiple Sm-Fe families was investigated. The original structure $SmFe_{12}$ with the well-known tetragonal *I4/mmm* symmetry is investi- gated with a parabolic dependence between formation energy and its magnetization by continuous distortions of the unit-cell lattice parameter and individual sites. Notably, a $SmFe_{12}$ structure with *I4/mmm* symmetry is found with 7.5% increasing magnetization while keeping the similar formation energy with the most stable structures in this family. With $SmFe_{11}CoN$ family, structures with N interstitial position in the center of Sm and Fe octahedron show outperform all other structures in both ability of stabilization and remaining high magnetization of the original structure [19].

(3) Developing an Evidence-based similarity measure for materials regarding physical property. The method incorporates measurement uncertainty into the similarity measure under the Dempster-Shafer evidence theory. We used two material datasets, including 3*d* transition metal–4*f* rare-earth binary and quaternary high-entropy alloys with target properties, Curie temperature, and magnetization. Based on the information obtained on the similarities between the materials, a clustering technique is applied to learn the cluster structures of the materials that facilitate the interpretation of the mechanism. The unsupervised learning experiments demonstrate that the obtained similarities are applicable to detect anomalies and appropriately identify groups of materials whose properties correlate differently with their compositions. Furthermore, significant improvements in the accuracies of the predictions for the Curie temperature and magnetization of the quaternary alloys are obtained by introducing the similarities, with the reduction in mean absolute errors of 36% and 18%, respectively [20].

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５．主な発表論文等

〔雑誌論文〕　計5件（うち査読付論文　5件／うち国際共著　2件／うちオープンアクセス　5件）

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|---|---|
| １．著者名<br>Nguyen Duong-Nguyen、Kino Hiori、Miyake Takashi、Dam Hieu-Chi | ４．巻<br>48 |
| ２．論文標題<br>Explainable active learning in investigating structure stability of SmFe12-α-βXαYβ structures X, Y {Mo, Zn, Co, Cu, Ti, Al, Ga} | ５．発行年<br>2022年 |
| ３．雑誌名<br>MRS Bulletin | ６．最初と最後の頁<br>31～44 |
| 掲載論文のDOI（デジタルオブジェクト識別子）<br>10.1557/s43577-022-00372-9 | 査読の有無<br>有 |
| オープンアクセス<br>オープンアクセスとしている（また、その予定である） | 国際共著<br>－ |

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| １．著者名<br>Ha Minh-Quyet、Nguyen Duong-Nguyen、Nguyen Viet-Cuong、Kino Hiori、Ando Yasunobu、Miyake Takashi、Deneux Thierry、Huynh Van-Nam、Dam Hieu-Chi | ４．巻<br>133 |
| ２．論文標題<br>Evidence-based data mining method to reveal similarities between materials based on physical mechanisms | ５．発行年<br>2023年 |
| ３．雑誌名<br>Journal of Applied Physics | ６．最初と最後の頁<br>053904～053904 |
| 掲載論文のDOI（デジタルオブジェクト識別子）<br>10.1063/5.0134999 | 査読の有無<br>有 |
| オープンアクセス<br>オープンアクセスとしている（また、その予定である） | 国際共著<br>－ |

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| １．著者名<br>Nguyen Duong-Nguyen、Dam Hieu-Chi | ４．巻<br>133 |
| ２．論文標題<br>Machine learning-aided Genetic algorithm in investigating the structure property relationship of SmFe12-based structures | ５．発行年<br>2023年 |
| ３．雑誌名<br>Journal of Applied Physics | ６．最初と最後の頁<br>063902～063902 |
| 掲載論文のDOI（デジタルオブジェクト識別子）<br>10.1063/5.0134821 | 査読の有無<br>有 |
| オープンアクセス<br>オープンアクセスとしている（また、その予定である） | 国際共著<br>－ |

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| １．著者名<br>Uematsu Hideshi, Ishiguro Nozomu, Abe Masaki, Takazawa Shuntaro, Kang Jungmin, Hosono Eiji, Nguyen Nguyen Duong, Dam Hieu Chi, Okubo Masashi, Takahashi Yukio | ４．巻<br>12 |
| ２．論文標題<br>Visualization of Structural Heterogeneities in Particles of Lithium Nickel Manganese Oxide Cathode Materials by Ptychographic X-ray Absorption Fine Structure | ５．発行年<br>2021年 |
| ３．雑誌名<br>The Journal of Physical Chemistry Letters | ６．最初と最後の頁<br>5781～5788 |
| 掲載論文のDOI（デジタルオブジェクト識別子）<br>10.1021/acs.jpclett.1c01445 | 査読の有無<br>有 |
| オープンアクセス<br>オープンアクセスとしている（また、その予定である） | 国際共著<br>該当する |

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| 1．著者名<br>Ha Minh-Quyet, Nguyen Duong-Nguyen, Nguyen Viet-Cuong, Nagata Takahiro, Chikyow Toyohiro, Kino Hiori、Miyake Takashi, Denoeux Thierry, Huynh Van-Nam, Dam Hieu-Chi | 4．巻<br>1 |
| 2．論文標題<br>Evidence-based recommender system for high-entropy alloys | 5．発行年<br>2021年 |
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| 掲載論文のDOI（デジタルオブジェクト識別子）<br>10.1038/s43588-021-00097-w | 査読の有無<br>有 |
| オープンアクセス<br>オープンアクセスとしている（また、その予定である） | 国際共著<br>該当する |

〔学会発表〕　計7件（うち招待講演　0件／うち国際学会　7件）

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| 1．発表者名<br>Duong-Nguyen NGUYEN, Hieu-Chi DAM |
| 2．発表標題<br>Query-and-learn Based Active Learning Framework for Screening New SmFe12 Structures |
| 3．学会等名<br>11th International Conference on Materials for Advanced Technologies（国際学会） |
| 4．発表年<br>2023年 |

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| 1．発表者名<br>Minh-Quyet HA, Duong-Nguyen NGUYEN, Hieu-Chi DAM |
| 2．発表標題<br>An Application of Evidence Theory for Revealing Similarity Between Materials Based on Physical Mechanisms |
| 3．学会等名<br>11th International Conference on Materials for Advanced Technologies（国際学会） |
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| 1．発表者名<br>NGUYEN Duong Nguyen, DAM Hieu Chi |
| 2．発表標題<br>Explainable active learning to investigate the structure-stability of SmFe12-a-BX a Y structures X, Y = {Mo, n, Co, Cu, Ti, Al, Ga} |
| 3．学会等名<br>Materials Research Meeting 2021（国際学会） |
| 4．発表年<br>2021年 |

１．発表者名
NGUYEN Duong Nguyen, DAM Hieu Chi

２．発表標題
Active learning in discovery SmFe12−x−yAxBy magnets A, B as Mo, Zn, Co, Cu, Ti, Al, Ga

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４．発表年
2021年

〔図書〕　計0件

〔産業財産権〕

〔その他〕

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６．研究組織

| | 氏名<br>（ローマ字氏名）<br>（研究者番号） | 所属研究機関・部局・職<br>（機関番号） | 備考 |
|---|---|---|---|

７．科研費を使用して開催した国際研究集会

〔国際研究集会〕　計0件

８．本研究に関連して実施した国際共同研究の実施状況

| 共同研究相手国 | 相手方研究機関 |
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