

論文 / 著書情報  
Article / Book Information

|                   |   |
|-------------------|---|
| 題目(和文)            | 3d磁性遷移金属ヘースハイエントロヒー合金薄膜の相形成と磁気特性  |
| Title(English)    | Phase formation and magnetic properties of 3d transition metal based high entropy alloy films   |
| 著者(和文)            | Li Boya   |
| Author(English)   | Boya Li   |
| 出典(和文)            | 学位:博士(学術),<br>学位授与機関:東京工業大学,<br>報告番号:甲第12925号,<br>授与年月日:2024年9月20日,<br>学位の種別:課程博士,<br>審査員:史 蹟,春本 高志,林 幸,村石 信二,LEI XIAOWEN   |
| Citation(English) | Degree:Doctor (Academic),<br>Conferring organization: Tokyo Institute of Technology,<br>Report number:甲第12925号,<br>Conferred date:2024/9/20,<br>Degree Type:Course doctor,<br>Examiner:,,,, |
| 学位種別(和文)          | 博士論文  |
| Category(English) | Doctoral Thesis   |
| 種別(和文)            | 要約  |
| Type(English)     | Outline   |

(博士課程)

Doctoral Program

# 論文要約

THESIS OUTLINE

系・コース： 材料  
Department of, Graduate major in 材料

系  
コース

学生氏名： Li Boya  
Student's Name

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| 論文題目<br>Thesis Title | Phase formation and magnetic properties of 3d transition metal based high entropy alloy films |
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要約

Thesis Outline

The organizations and contents of the thesis are summarized as following seven chapters:

**Chapter 1 Introduction:** The characteristics of high entropy alloys (HEAs) are introduced, and the advantages and challenges of high entropy alloy films (HEAFs) used as soft magnetic materials are discussed. HEAFs containing magnetic elements (Fe, Co, and Ni) exhibit good ferromagnetism. Due to the high entropy effect produced by the addition of non-magnetic elements, FeCoNi-based HEAFs are expected to show high resistivity, excellent chemical/thermal stability, and mechanical properties, considered as soft magnetic material with excellent comprehensive physical properties. On the other hand, it is pointed out that the development of magnetic HEAFs still faces many challenges, such as precise control of film composition, improving magnetic properties and understanding the influence of non-magnetic elements on the magnetic properties of HEAFs. Therefore, the main objectives of this study are to establish a preparation method that can accurately control the composition of the film and clarify the influence of non-magnetic elements on the microstructure, magnetic and electric properties of HEAFs.

**Chapter 2 Preparation and characterization of FeCoNi-based HEAFs:** We study the conditions for preparing FeCoNi-based HEAFs from pure metal targets by magnetron sputtering method. The HEAFs prepared under optimized conditions have uniform composition distribution and excellent soft magnetic properties. The basic characterization techniques on the chemical composition, microstructures, magnetic properties, and electrical properties are introduced. It also explains how to calculate the mixing enthalpy, mixing entropy, valence electron concentration, and atomic radius mismatch, which relate to microstructure of HEAFs.

**Chapter 3 Microstructure and magnetic properties of Pt-containing FeCoNiCu<sub>x</sub>Pt<sub>x</sub> high entropy alloy films:** HEAFs are prepared from multilayer precursors using a two-step method involving magnetron sputtering deposition and vacuum annealing. By adjusting the thickness of each layer of the multilayer precursor, the composition of the film can be precisely controlled. Through vacuum annealing at elevated temperatures, complete mixing of the multilayer

component can be realized to form uniform high entropy alloy films. By changing their concentrations, the effects of Pt and Cu on microstructure and magnetic properties of  $\text{FeCoNiCu}_x\text{Pt}_x$  HEAFs have been systematically investigated. With the concentration of Pt and Cu increase, the saturation magnetization ( $M_S$ ) tends to decrease while the coercivity ( $H_C$ ) increases. The results show that  $\text{FeCoNiCu}_{0.2}\text{Pt}_{0.2}$  HEAFs exhibit excellent soft ferromagnetic behavior ( $M_S = 802 \text{ emu/cm}^3$ ,  $H_C = 52 \text{ Oe}$ ) and strong in-plane magnetic anisotropy, which are suitable for application as the core material of high frequency thin film inductors. Also, the effect of Pt on total magnetization have been studied by DFT calculation. The mechanisms of the interrelationships between composition, microstructure and electronic structure have been investigated.

**Chapter 4 Effect of Al and Mn on the magnetic properties of  $\text{FeCoNiCu}_x\text{M}_x$  (M = Al/Mn) high entropy alloy films:**

The two-step method same as in the previous chapter are used to prepare  $\text{FeCoNiCu}_x\text{M}_x$  (M = Al, Mn) HEAFs. And the effects of Al and Mn on the microstructure and magnetic properties were reported. As the concentrations of Al and Mn increase, the  $M_S$  of HEAFs decreases. DFT calculations show that Al atoms in the film have no magnetic moment, while Mn atoms have magnetic moments in the opposite direction to magnetic elements. So that, the presence of Al atoms reduces  $M_S$  due to the dilution effect on magnetic element atoms, while the presence of Mn atoms reduces  $M_S$  due to the antiferromagnetically couple with the magnetic element atoms.

**Chapter 5 Effect of Si on microstructure and magnetic properties of  $\text{FeCoNiCu}_{0.5}\text{Si}_x$  high entropy alloy films:**

Via controlling the power of Si target,  $\text{FeCoNiCu}_{0.5}\text{Si}_x$  HEAFs are prepared by magnetron co-sputtering. The microstructure and magnetic properties of  $\text{FeCoNiCu}_{0.5}\text{Si}_x$  HEAFs have been investigated to clarify the effects of Si concentration on microstructure formation and magnetic properties. It has been found that Si addition strongly affects the magnetic properties of  $\text{FeCoNiCu}_{0.5}\text{Si}_x$  HEAFs of FCC phase by decreasing the coercivity of the films, but hardly affects the phase formation in the films. On the other hand, it has been found that deposition of the films at moderate high substrate temperatures greatly improve the magnetic properties by increasing the saturation magnetization while keeping low coercivity. The addition of Si effectively inhibits the increase in coercivity, which is due to the decrease in magnetostriction.

**Chapter 6 Amorphization of  $\text{FeCoNiCu}_{0.5}\text{Zr}_x$  high entropy alloy films:**

$\text{FeCoNiCu}_{0.5}\text{Zr}_x$  HEAFs with different Zr concentration were prepared by magnetron co-sputtering method. The effects of Zr concentration on microstructure, electrical properties and magnetic properties of HEAFs are systematically investigated. The addition of Zr is found to induce an amorphous structure, the amorphization is due to the atomic radius mismatch between constituent elements. In addition, the amorphization of HEAFs cause the decrease of coercivity, enhancing resistivity and reducing temperature coefficient of resistance. The study also shows that annealing can induce the transformation of Zr-containing HEAFs from amorphous structure to solid solution structure, leading to improved magnetic properties, while maintaining low coercivity (10 Oe or less).

**Chapter 7 Conclusions:** This study has established a method for precisely controlling the composition of HEAFs, and elucidated the relationship between the composition, microstructure, and magnetic properties. We have identified the conditions for fabricating HEAFs with excellent soft magnetic properties. Prepared FeCoNi-based HEAFs exhibit good performance in terms of saturation magnetization, coercivity, electrical resistivity, and thermal stability, making them suitable for applications. Furthermore, the variation of magnetic properties caused by addition of non-magnetic elements are clarified through DFT calculations. This study not only deepens the fundamental understanding of composition-microstructure-property relationships in HEAFs but also highlights pathways for further exploration and development of HEAF in design and application.