様　式　Ｃ－１９、Ｆ－１９－１、Ｚ－１９　（共通）

科学研究費助成事業　　研究成果報告書

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研究課題名（和文）Evolution of new magnetic materials with ultrahigh coercivity

研究課題名（英文）Evolution of new magnetic materials with ultrahigh coercivity

研究代表者
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交付決定額（研究期間全体）：（直接経費）　　3,200,000円

研究成果の概要（和文）：本研究では、化学合成で得られた(Sm,Zr)(Fe,Co,Ti)12単相微粒子を合成することに成功し、これに表面処理を施すことで1Tを超える高保磁力が発現することを見出しました。合成直後の(Sm,Zr)(Fe,Co,Ti)12微粒子では、微粒子表面の元素組成が内部と異なっていますが、表面処理を行うことにより微粒子がほぼ完全なThMn12型構造になることが分かりました。最適な表面処理を行うことにより、(Sm,Zr)(Fe,Co,Ti)12微粒子の保磁力が1.6Tまで向上することも分かりました。これらの知見は、高Fe濃度1-12系希土類磁石の高性能永久磁石としての実用化につながると考えられます。

研究成果の学術的意義や社会的意義
Through these studies, novel nanomaterials science called “nanoscale transformation science” and high performance permanent magnet materials are developed. The research achievement addresses the most pressing challenges of the rare-earth crisis for applications in energy-saving technologies.

研究成果の概要（英文）：We have succeeded in preparing (Sm,Zr)(Fe,Co,Ti)12 (ThMn12, I4/mmm) well-defined particles with ever-larger coercivity (Hc) and remanent magnetization (Mr) based on precise control over their crystallinity and microstructure using chemical synthesis. In particular, the synthesized (Sm,Zr)(Fe,Co,Ti)12 particles have high crystallinity, controllable grain-size and composition, and diverse surface from Sm-enriched and Ti-enriched to depleted surface.
The most important achievements for their Hc and Mr were as large as 1.6T and 1.2T, respectively, at 300 K for isotropic powders. This result is the world's highest performance for SmFe12-based permanent magnet materials. It is a breakthrough in the development of SmFe12-based permanent magnet materials, opening a new era of permanent magnets.

研究分野：Material chemistry

キーワード：SmFe12　Ultrahigh　Coercivity　Permanent　Magnet　Materials

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1. 研究開始当初の背景

SmFe$_{12}$ (ThMn$_{12}$, *I*4/*mmm*) compound has excellent intrinsic magnetic properties, superior to the current high-end-magnet Nd$_2$Fe$_{14}$B compound.[1] Yet, the synthesis of SmFe$_{12}$-based bulk materials with sufficiently large coercivities (H_c) for fabricating permanent magnets has not been realized. The most obvious obstacle to improving H_c is critical issues of microstructure, especially the inevitable formation of magnetically-soft SmFe$_x$ surface. Herein we have already succeeded in preparing Sm(Fe,Co,Ti)$_{12}$ microparticles with ever-larger H_c and remanence (M_r) based on precise control over their microstructure by advanced chemical synthesis, opening a new era of SmFe$_{12}$-based permanent magnet materials.[2-6]

2. 研究の目的

The present research focused on the evolution of new (Sm$_{1-x}$Zr$_x$)(Fe$_{1-y}$Co$_y$)$_{12-z}$Ti$_z$ ($0 \le x \le 0.3$, $0.2 \le y \le 0.3$, $0 \le z \le 1$) (ThMn$_{12}$, *I*4/mmm) permanent magnet materials with ultrahigh H_c and large M_r by a chemical synthesis approach based on nanoscale transformation science. These materials will be used as building blocks for the fabrication of easy-axis aligned permanent magnets with large $(BH)_{\max}$

3. 研究の方法

The synthesis implementing novel nanoscale diffusion and transformation in reduction-diffusion and chemoselective surface refinement reactions is crucial to solving challenging tasks of crystal and microstructure engineering. It included multiple steps as belows.

(1) Chemical syntheses of Sm-, Zr-, and Ti-doped/encapsulated CoFe$_2$O$_4$ nanoprecursors,

(2) Conversion of the nanoprecursors into (Sm,Zr)(Fe,Co,Ti))$_{12}$ particles by reduction-diffusion (RD) processes at high temperatures,

(3) Controlling compositions of the (Sm,Zr)(Fe,Co,Ti))$_{12}$ particles by varying feeding ratios of the starting materials,

(4) Controlling grain size of the (Sm,Zr)(Fe,Co,Ti))$_{12}$ particles bay varying temperatures in the range of 900–1100 ºC and/or utilizing 160 nm CaO particles as a dispersant for RD reaction based on thermodynamics and/or kinetics, respectively,

(5) Chemoselective surface refinement of the (Sm,Zr)(Fe,Co,Ti))$_{12}$ particles, and

(6) Fabrication of the easy-axis aligned (Sm,Zr)(Fe,Co,Ti))$_{12}$ bulk permanent magnets (PMs) by bonding or sintering processes under apply fields for bonded magnets or sintered magnets, respectively.

Microstructure and magnetic properties of the (Sm,Zr)(Fe,Co,Ti))$_{12}$ particles and their easy-axis aligned bulk PMs were elucidated by a wide range of characterization techniques.

4. 研究成果

The synthesized (Sm,Zr)(Fe,Co,Ti)$_{12}$ particles have high crystallinity, controllable compositions and grain sizes (1–3 μm), and diverse surfaces from Sm-enriched and Ti-enriched to depleted surfaces (Fig. 1A). The most important achievement was to turn out that the inevitable formation of magnetically-soft surfaces (SmFe$_x$ compounds) is responsible for very low H_c and the refined surfaces (Fig. 1A) resulted in intriguing H_c. As a result, $\mu_0 H_c$ and $\mu_0 M_r$ values of their surface-refined (Sm,Zr)(Fe,Co,Ti)$_{12}$ isotropic powders at 300 K were as large as 1.6 T and 1.2 T, respectively (Fig. 1B). This result is the world's highest performance for SmFe$_{12}$-based permanent magnet materials.

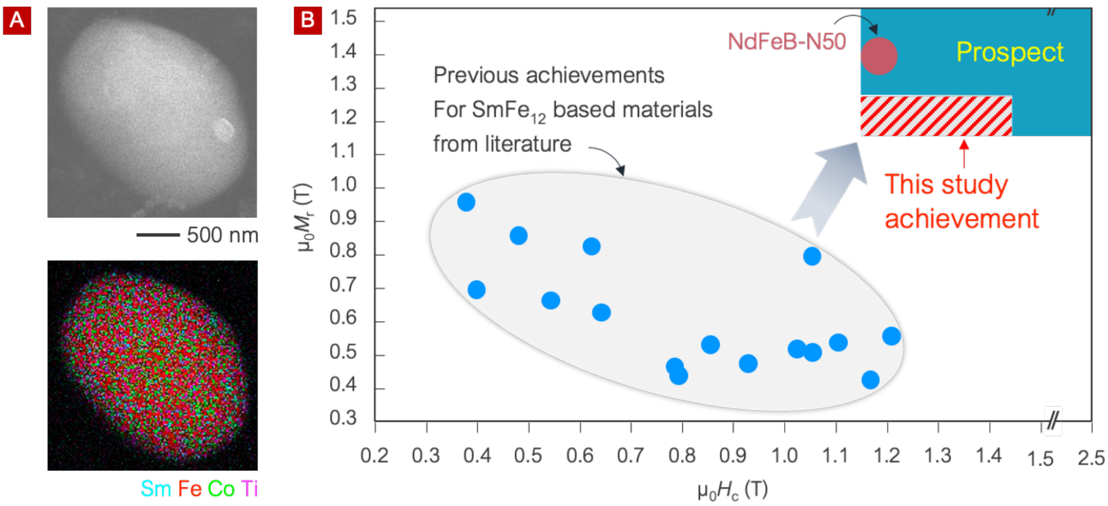

Fig. 1 A. SEM image and EDS elemental map of a Sm(Fe$_{0.8}$Co$_{0.2}$)$_{11}$Ti microparticle. B. Extrinsic magnetic properties of isotropic Sm(Fe$_{0.8}$Co$_{0.2}$)$_{11}$Ti micropowders at 300 K. NdFeB-N50 is a commercial Nd$_2$Fe$_{14}$B-based magnet.

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3. 特願 2021-079367 Method for producing magnetic powders
4. Trinh (*1st author*) *et al.*, Pushing the Performance Limit of SmFe$_{12}$-Based Permanent Magnet Materials, to be submitted to *Nature* **2022**.
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５．主な発表論文等

〔雑誌論文〕　計1件（うち査読付論文　0件／うち国際共著　0件／うちオープンアクセス　0件）

１．著者名 T Thuy Trinh, Jungryang Kim, Ryota Sato, Kenshi Matsumoto, Toshiharu Teranishi	４．巻 22
２．論文標題 Synthesis of Mesoscopic Particles of Multi-Component Rare Earth Permanent Magnet	５．発行年 2021年
３．雑誌名 Sci. Technol. Adv. Mater.	６．最初と最後の頁 37-54
掲載論文のDOI（デジタルオブジェクト識別子） 10.1080/14686996.2020.1862630	査読の有無 無
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 －

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４．発表年 2021年

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４．発表年 2021年

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〔図書〕　計0件

〔出願〕　計1件

産業財産権の名称	発明者	権利者
Method for producing magnetic powders	Trinh Thang Thuy	同左
産業財産権の種類、番号	**出願年**	**国内・外国の別**
特許、PCT/JP2022/01960	2022年	外国

〔取得〕　計1件

産業財産権の名称	発明者	権利者
Method for producing magnetic powders	Trinh Thang Thuy	同左
産業財産権の種類、番号	**取得年**	**国内・外国の別**
特許、2021-079367	2021年	国内

〔その他〕

－

６．研究組織

	氏名 （ローマ字氏名） （研究者番号）	所属研究機関・部局・職 （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