7 Elements Strategy Initiative Center for Magnetic Materials (ESICMM)

– in situ analysis using neutrons and X-rays –

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The objectives of the Elements Strategy Initiative Center for Magnetic Materials (ESICMM) based at the National Institute of Material Science (NIMS) are: (1) to synthesize mass-producible high-performance permanent magnets without using critical rare-earth elements for future generations and (2) to build a framework for use of basic science and technology for industrial R&D. To achieve these objectives, ESICMM is not only focusing on theoretical research and mining of new permanent magnet materials but also simultaneously studying different processing techniques to improve the existing high-performance permanent magnet materials. This is being achieved through collaboration amongst three fields of research namely computer physics, structural and property characterization, and material processing respectively. ESICMM also aims to train scientists who will be able to contribute to the future development of magnetic functional materials.

The CMRC started a project on in-situ analysis using neutrons and X-rays in July 2012 as an analysis group of ESICMM. The complementary use of neutrons at J-PARC/MLF and other facilities as well as use of synchrotron X-rays at the Photon Factory is very useful for analyzing magnetic materials from the atomic scale to micrometer scale.

7-1 Introduction

A comprehensive study was developed to determine the effect of radiations of neutrons and synchrotron on sintered magnets and hot-deformed magnets made of rare earth permanent magnet material and rare earth free magnet material respectively. The following studies were conducted to determine the structure of crystal grains, grain boundaries, sub-phases, and magnetic microstructures respectively.

(1) Structure analysis and magnetic structure analysis of magnetic materials using neutron and synchrotron radiation

Structural analysis was carried out on magnetic materials made of rare earth/ transition metal (1-12 and 2-14-1 systems) through neutron diffraction and synchrotron X-ray diffraction. The magnetic structure of these magnetic materials was determined using temperature ranging from low to Curie temperature respectively. It is clarified that the magnetic moment of each atomic site of rare earth and transition metal sites in a magnet is dependent on the temperature.

(2) In-situ structural analysis of magnet materials at high temperature using neutron diffraction

Structure analysis of the primary phase and sub-phase of the magnet material was carried out at high temperature to study the impact of temperature on phase formation and thermal decomposition of magnetic material. . In-situ structural analysis of 2-14-1 rare earth/transition systems was carried out using neutron diffraction at high temperature. This methodology enabled us to understand the primary phase structure, the crystal structure, the volume fraction, and the phase formation mechanism of the sub-phase material present at the grain boundary phase at high temperature. This also enabled us to determine if this methodology is efficient to enable high-temperature in-situ structural analysis through information science techniques.

(3) Evaluation of the physical properties of magnetic materials

Magneto-crystalline anisotropy and exchange stiffness constant are the fundamental physical parameters of magnetic materials. A method has been devised to determine the magneto-crystalline anisotropy and exchange stiffness constant of magnetic materials using neutron and synchrotron radiation and optical measurement techniques respectively. The spin-wave dispersion of 2-14-1 and 1-12 rare earth/transition metal systems was determined using neutron Brillouin scattering method and a spin-wave dispersion model has been designed for rare earth permanent magnets based on the two-sublattice model.

(4) Study of the magnetization process of permanent magnets

We studied the process of magnetization reversal of magnetic materials was studied using several techniques such as small-angle neutron scattering, micromagnetic simulation, and X-ray microscopy respectively. The details of the process of magnetization and the magnetic interaction between the grains were studied by combining micro magnetic simulation, and information science techniques respectively. Furthermore, a method has also been developed for predicting the process of magnetization through use of materials informatics.

7-2 Results

(1) Structure analysis and magnetic structure analysis of magnetic materials using neutron and synchrotron radiation

The crystal structure and the magnetic structure are the most vital characteristics of magnetic materials in the primary phase. The magnetic structure affects the magnetic properties of the primary phase. Moreover, the impact of temperature ranging from low temperature to the Curie temperature enables us to understand the role of the primary phase in magnetic materials. It is difficult to determine the magnetic structure as the X-ray diffraction does not respond to magnetism. Hence, it is necessary to use neutrons, which act as exemplary magnetic probes. By using neutron diffraction, it is possible to study magnetic structure and its dependence on temperature by using the technique of neutron diffraction. It is also possible to obtain the quantitative value of the magnetic moment that is observed at each site of the primary phase crystal constituting the magnet.

The structural analysis of the rare earth/transition metal with 1-12 and 2-14-1 systems was carried out using the neutron diffraction and synchrotron X-ray diffraction techniques. Powder neutron diffraction experiments were conducted on $(Sm_{1-x}Zr_x)(Fe_{0.7}Co_{0.3})_{11.2}Ti_{0.8}$ (x = 0, 0.2) as a 1-12 magnetic material of rare earths and transition metals. Figure 1 represents the neutron diffraction data of $(Sm_{1-x}Zr_x)(Fe_{0.7}Co_{0.3})_{11.2}Ti_{0.8}$ where, x = 0, 0.2. It was previously assumed that the powder neutron diffraction experiment on Sm compounds will be difficult due to high neutron absorption property of Sm. However, analysis of the neutron diffraction pattern on Sm-based rare earth/transition metal systems enabled us to confirm that neutron diffraction can be used to determine the magnetic structure, site occupancy as well as the atomic position if Zr. In case of 2-14-1 systems of rare earth/transition metal, powder neutron diffraction experiment was conducted for $Nd_2Fe_{14}B$, which is

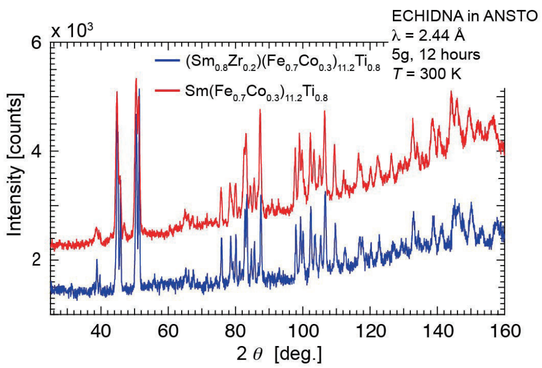

Fig. 1: Neutron diffraction patterns of $(Sm_{1-x}Zr_x)(Fe_{0.7}Co_{0.3})_{11.2}Ti_{0.8}$ (x = 0, 0.2)

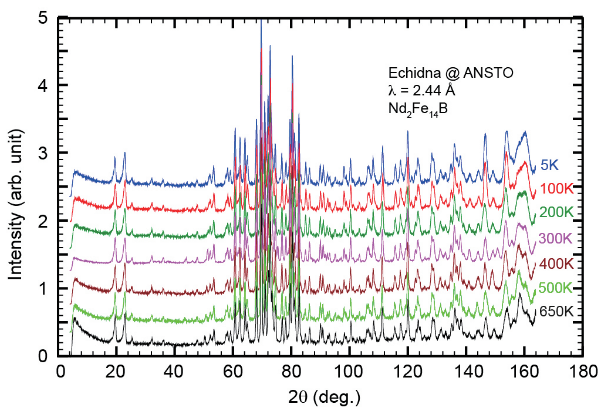

Fig. 2: Neutron diffraction patterns in the temperature range from 5K to 650K of $Nd_2Fe_{14}B$

the primary composite material of the neodymium magnet, under temperature ranging from low temperature to curie temperature respectively Figure 2 represents the neutron diffraction data for $Nd_2Fe_{14}B$ obtained under temperature ranging from 5K to 650K. The magnetic structure of $Nd_2Fe_{14}B$ was successfully determined by analyzing the neutron diffraction patterns as seen in figure 1 and 2 respectively. Hence, the impact of temperature on the magnetic moment at each site was obtained for the rare earth sites and the transition metal sites in rare earth permanent magnets.

(2) In-situ structural analysis of magnet materials at high temperature using neutron diffraction

The structural analysis of the primary phase and the sub-phase of the magnetic material was carried out at a high temperature to study phase formation and thermal decomposition of the magnet material. The methodology was also tested for efficient high-temperature in-situ structural analysis using information science. Concerning rare earth/transition metal 2-14-1 systems, Ga-added Nd-rich Nd-Fe-B sintered magnets, which have been attracting attention for its mechanism of high coercivity, was measured. We conducted an experiment at a high temperature. Figure 3 depicts the result of the in-situ structural analysis with neutron diffraction at high temperature. In-situ structural analysis was carried out to determine the structure of the primary phase and the sub-phase found in the grain boundary phase, the volume fraction, and the phase generation mechanism under high-temperature conditions.

(3) Evaluation of physical properties of magnetic materials

We developed a method to accurately determine the basic physical parameters of magnetic materials, such as magnetocrystalline anisotropy and the exchange stiffness constant. These two parameters can be accurately determined by combining the neutron Brillouin scattering experiment for magnetic materials and the newly developed data analysis method that uses information science. We determined the spin-wave dispersion of rare-earth/transition metal 2-14-1 and 1-12 systems using neutron Brillouin scattering. Fig. 4 shows the spin-wave dispersion of 2-14-1 and 1-12 systems obtained through data analysis of neutron Brillouin scattering experiments. Table 1 shows the exchange interaction energy J_{TT} and magnetic anisotropy energy n_{RT} of rare earth/transition metals 2-14-1 and 1-12 systems obtained from data analysis of neutron Brillouin scattering experiments.

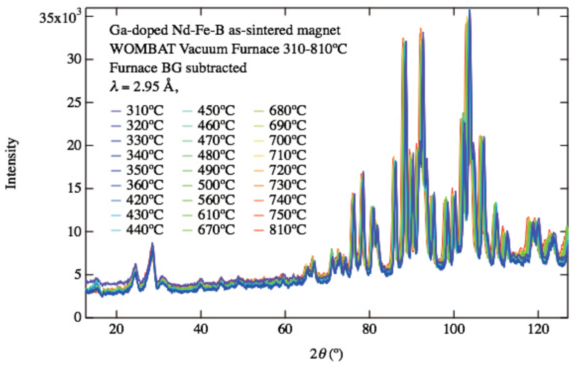

Fig. 3: Result of the in-situ structural analysis with neutron diffraction at high temperatures for Ga-added $Nd_2Fe_{14}B$.

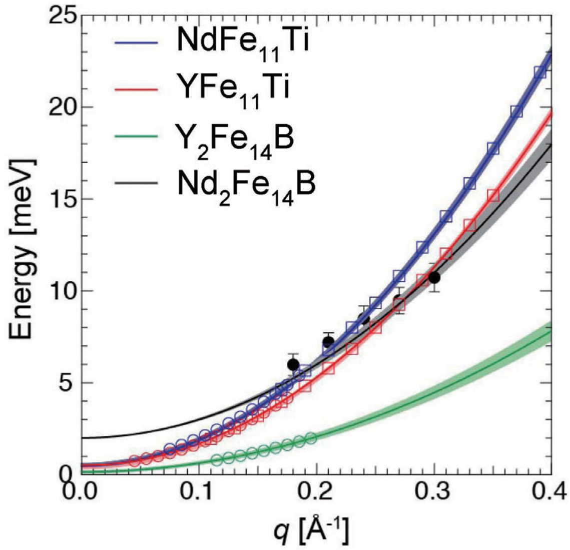

Fig. 4: Spin-wave dispersion of 2-14-1 and 1-12 systems obtained by data analysis of neutron Brillouin scattering experiments.

	J_{TT} [meV Å^2]	n_{RT} [meV]
$Y_2Fe_{14}B$	48(3)	0.15(6)
$Nd_2Fe_{14}B$	100	2
$YFe_{11}Ti$	121(1)	0.6(1)
$NdFe_{11}Ti$	140(2)	0.5(1)

table 1 : Exchange interaction energy J_{TT} and magnetic anisotropy energy n_{RT} of rare earth/transition metals 2-14-1 and 1-12 systems obtained from data analysis of neutron Brillouin scattering experiments.

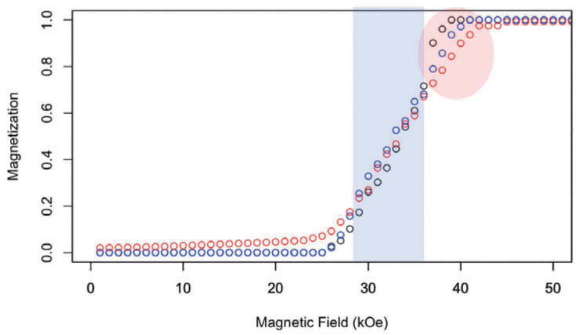

Fig. 5: predicted magnetization process and the actual magnetization process

sition metals 2-14-1 and 1-12 systems obtained from data analysis of neutron Brillouin scattering experiments. From the results, we obtained the spin-wave model of rare earth permanent magnets (rare earth/transition metal 2-14-1 and 1-12 systems) based on the two sublattice models.

(4) Study of the magnetization process of permanent magnets

To elucidate the magnetization process of magnetic materials, we studied the magnetization reversal process using small-angle neutron scattering, micromagnetic simulation, and X-ray microscopy. In order to clarify the details of the magnetization process and the magnetic interaction between grains, we developed characterization methods that combined experiments, micromagnetic simulations, and information science. We built a model to predict the reversal magnetic field for each crystal grain using machine learning. From the evaluation of the machine-learning model, it was possible to predict the reversal magnetic field of crystal grains with high accuracy. Furthermore, we developed a method for predicting the magnetization process using the materials informatics method. Fig. 5 shows the predicted magnetization process and the actual magnetization process. The magnetization process can be accurately predicted by the materials informatics method and can be used for future magnet design.