The Effect of Neutron Irradiation on Sm_2Co_{17} -Based High Temperature Magnets and Nd-Fe-B Magnets

Christina H. Chen [1], Joseph Talnagi [2], Jinfang Liu [3], Payal Vora [3], Ashil Higgins [1], and Sam Liu [1]

1. University of Dayton Research Institute, Dayton, OH 45469,
2. The Ohio State University Research Reactor, Columbus, OH 43212,
3. Electron Energy Corporation, Landisville, PA 17538

***ABSTRACT*: Results from neutron irradiation experiments on rare earth magnets show that the radiation tolerance is very high for Sm_2Co_{17} type magnets and is fairly low for Nd-Fe-B type magnets. The analyses, based on our results and that reported by others show that the major radiation damage in permanent magnets is caused by a radiation-induced thermal spike accompanied by a localized temperature T_L, and, therefore, the radiation tolerance of a magnet is determined mainly by its thermal stability. T_L value depends on the radiation intensity and the magnet composition. The thermal stability of a magnet at temperature below its Curie temperature is related to its intrinsic coercivity H_{ci} and its shape. Finite element analysis is used to show how the thermal stability affects the radiation tolerance.**

I INTRODUCTION

The radiation effects in materials can be categorized into four types: thermal spike, transmutation, ionization, and atom displacement [1]-[2]. Thermal spike is an energy absorption process accompanied by heat. Transmutation creates different atoms and particles. Ionization produces electron-hole pairs; and atom displacement results in interstitial-vacancy pairs. Comprehensive research has produced a great deal of information of radiation effects on semiconductors in the last few decades; however, there was little research for radiation effects on permanent magnets until early 1980s. Rare earth magnets of $SmCo_5$, Sm_2Co_{17}, and $Nd_2Fe_{14}B$ types were developed in 1960s to 1980s [3]-[5]. Since 1983, a dozen groups have reported the radiation effects in rare earth magnets with electron beams, protons, γ-rays, x-rays, and neutrons [6]-[22]. Regardless of the radiation sources, similar results were described - Sm_2Co_{17} type had high radiation tolerance and Nd-Fe-B type had poor tolerance. The mechanism of radiation damage in magnets, however, remained unclear [11]-[13]&[19]-[21]. In the late 1990s, Sm_2Co_{17} type high temperature magnets for use up to 550°C were developed [23]-[24] and used for NASA's space missions [25]. The conditions in this new environment make it necessary to develop a better understanding of radiation effects in the magnets.

II. EXPERIMENT PROCEDURE

All magnets used in this study had intrinsic coercivity (H_{ci}) >21 kOe and L/D =1.25 (L is magnet length along the direction of magnetization and D is magnet diameter). Samples T300C and T500C were Sm_2Co_{17} type with maximum operating temperatures, T_M, = 300°C and 500°C, respectively. $Nd_{13}Dy_2Fe_{77}B_8$ were $Nd_2Fe_{14}B$ type with high coercivity and moderate T_M (~180°C). The Ohio State University Research Reactor (OSURR) was the source of the radiation. The neutron energy ranged from 0.02 to 10 MeV, and the neutron flux ranged from $4.7x10^{10}$ to $2.1x10^{13}$. Gamma radiation accompanied the neutron but was not considered a contributor because gamma rays have no measurable effect on rare earth magnet properties [14]-[16]. The samples were placed in quartz tubes, and some samples had small holes to accommodate embedded thermocouples. Magnetic moments for each magnet were measured with a Helmholtz coil before and after each exposure to the various levels of radiation.

III. RESULTS AND DISCUSSIONS

The losses of magnet flux vs. neutron fluence are plotted in Fig.1. $Nd_{13}Dy_2Fe_{77}B_8$ samples lost 100% of magnet flux at fluence $\geq 10^{16}$ neu•cm^{-2}. The flux losses associated with the T300C and T500C magnets were undetectable at fluence as high as $\geq 10^{20}$ neu•cm^{-2}. The maximum temperature recorded was 267°C in $Nd_{13}Dy_2Fe_{77}B_8$ and 206°C in T500C. Fig. 1 also includes some data from previous literature reported by other groups [6]-[9]. Regardless of the sources of magnets, all Sm_2Co_{17} type specimens had nearly zero loss at fluence as high as $\sim 10^{20}$ neu•cm^{-2}, and $Nd_2Fe_{14}B$ type specimens had significant losses at fluence $\geq 10^{16}$ neu•cm^{-2}. Cost et al [7] showed two different losses for the same Nd-Fe-B magnets. At fluence of $\sim 1.1\text{x}10^{16}$ neu•cm^{-2}, one set lost ~60% with a temperature at 77°C, which had helium in the sample holder. Another set lost ~90% with a temperature at 153°C, which had air in the holder. Helium has five times higher thermal conductivity than air, which obviously helped to disperse the heat. Cost et al. used neutron flux at $4\text{x}10^{12}$ neu•cm^{-2}•sec^{-1}, which was ~20% of the maximum flux used in our experiment. Their recorded temperature in the sample holder filled with air was lower than that recorded in our experiment (with air).

We attribute the localized temperature, T_L, as being induced by the thermal spike resulting from irradiation. The values of T_L were higher than the recorded temperature since it was highly localized in the scale of a cluster of atoms. In our experiment, once neutron flux was increased, several minutes elapsed before the recorded temperature was stabilized, and it did not change as the fluence increased. The recorded temperatures vs. neutron flux in both T500C and $Nd_{13}Dy_2Fe_{77}B_8$ are shown in Fig. 2. The maximum recorded temperature was 267°C in $Nd_{13}Dy_2Fe_{77}B_8$ and 207°C in Sm-Co T500C. It was 249°C in $Nd_{13}Dy_2Fe_{77}B_8$ at flux of $2\text{x}10^{13}$ neu•cm^{-2}•s^{-1}, where the specimens lost 100% of their flux. Since the magnetic flux was fully recoverable by remagnetization after exposure at this irradiation level, we have concluded that radiation-induced damage in the magnets is the result of thermal demagnetization. This effect is comparable to heating a magnet in an oven to a temperature above its Curie temperature, T_c.. The T_c is 309°C for these $Nd_{13}Dy_2Fe_{77}B_8$ specimens, so the minimum T_L must be 309°C when the recorded temperature was 249°C. The minimum T_L values inside $Nd_{13}Dy_2Fe_{77}B_8$ vs. flux were estimated and plotted in Fig. 2, which are 60°C higher than the recorded temperature. The T_c is 875°C for T500C and 820°C for T300C, and the T_L cannot be estimated for them since no magnetic loss was detectable for these Sm_2Co_{17} type magnets in our experiment. Fig. 2 shows that the T_L was the function of neutron flux, rather than the fluence. T_L also depends on magnet types.

In the past two decades, many research groups reported the effects of radiation on rare earth magnets. Some considered the damage as being caused by atom displacement [22]. Several groups, nevertheless, realized the important roles of H_{ci}, the shape, and the T_c of the magnets. Cost et al. [17] and Ito et al. [12] had reported the effects of H_{ci} values and L/D ratios, which showed higher H_{ci} and larger L/D resulting in higher radiation tolerance. Their data are re-plotted in Fig. 3 in a similar scale of this paper. However, it has not been recognized that the major magnetic losses were caused by radiation-induced thermal spike and the radiation tolerance relied on the thermal stability of magnets.

At temperatures below T_c, the H_{ci} values and the magnet shapes play dominant roles for maintaining the magnetic properties during heating, either with or without irradiation. To explain this nature, Fig. 4 shows the demagnetization curves of Sm-Co T500C and Nd-Fe-B N48 at 25 and 150°C. Theoretically, as long as the $T_L < T_c$, a magnet should fully recover its magnet strength after cooling down to room temperature (RT), if it had a perfect linear extrinsic demagnetization curve at temperature exposed. The extrinsic demagnetization curve is the curve of magnetic induction B vs. demagnetizing field (B-curve for short). To maintain a linear B-curve, a high H_{ci}

at RT and a low temperature coefficient of H_{ci} are required. H_{ci} reflects the resistance to demagnetization and the higher the value, the more robust the magnet. If a magnet does not have a linear B-curve, a large permeance coefficient is required [26]. The permeance coefficient is a critical factor in magnetic design. The load line is a function of the L/D ratio for a stand alone magnet. To maintain stable performance, a minimum L/D_m ratio is required, which must correspond to the B/H line above the "Knee" of the B-curve. As Fig. 4 shows, N48 does not have linear B-curves at 25 to 150°C, and its L/D_m ratio should be greater than 0.15 at 25°C, and 3.0 at 150°C for a cylinder magnet. Sm-Co T500C has linear B-curves at temperatures up to 500°C, and no minimum L/D_m ratio is required for applications at all temperatures up to 500°C. Fig. 5 shows 2-D and 1D magnetic flux distributions for T500C and N48 cylinder magnets with L/D =0.4 at 25 and 150°C, which were analyzed by using FEMM finite element analysis software [27]. The plots show that T500C has ~10% loss and N48 has >90% loss in magnet flux from 25°C to 150°C. After cooling down to 25°C, T500C has nearly zero loss and N48 has ~79% losses.

In our experiment, after remagnetization, the recovery rate of magnetic flux of $Nd_{13}Dy_2Fe_{77}B_8$ was 100% after irradiation at 10^{16} neu•cm^{-2}, and 97.5% at 10^{17} neu•cm^{-2}, and 95% at 10^{18} neu•cm^{-2}, which suggested some structural changes likely caused by transmutation or other effects at fluence ≥10^{17} neu•cm^{-2}. Those minor radiation damages will be studied in future research work.

VI. CONCLUSIONS

The major radiation damage is caused by radiation-induced thermal spike accompanied by a localized temperature T_L, which it can exceed the T_c of magnets. The factors determining the T_L during irradiation are: a). the intensity of radiation; b). the magnet type with certain heat capacities and thermal conductivities; c). the conditions of thermal dispersion. The dominant factor for radiation tolerance is the thermal stability of magnets, determined by three factors: 1). H_{ci} of magnets and the temperature coefficient of H_{ci}, which determine the linearity of the B-curve; 2). the load-line of the magnet, related to the L/D ratio of the magnet; and 3) the T_c of the magnet.

The main radiation damage in the semiconductor is caused by ionization and atom displacement, from which the point defects of electron-hole pairs and interstitial-vacancy pairs disrupt the conduction bands. However, point defects in the scale of electrons and atoms are not a concern for all the permanent magnets, including ferrites, Alnico, and others. On the other hand, the thermal stability, related to T_c and H_{ci}, always determines the performance of a permanent magnet in irradiation conditions, thus our conclusions apply to all types of permanent magnets.

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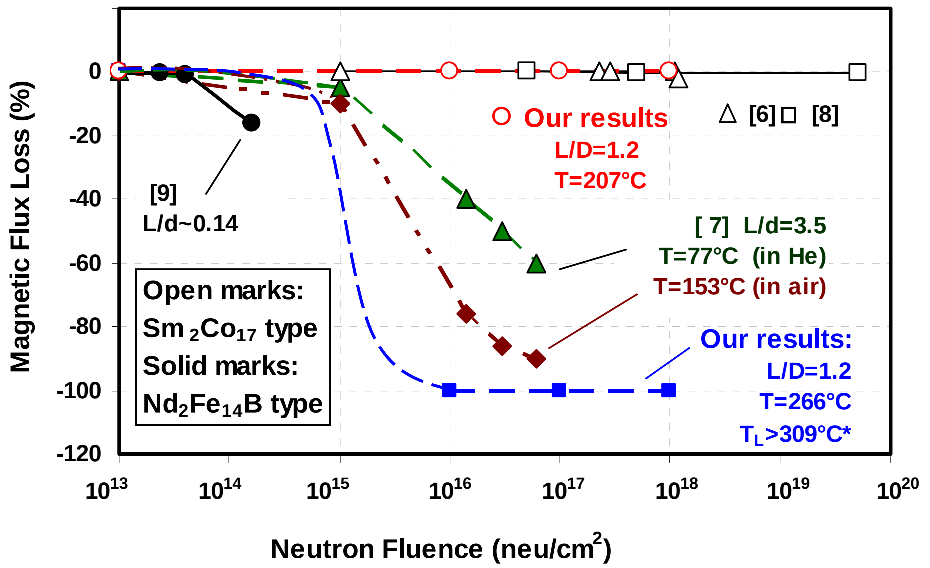


Fig. 1 Magnet losses vs. neutron fluence

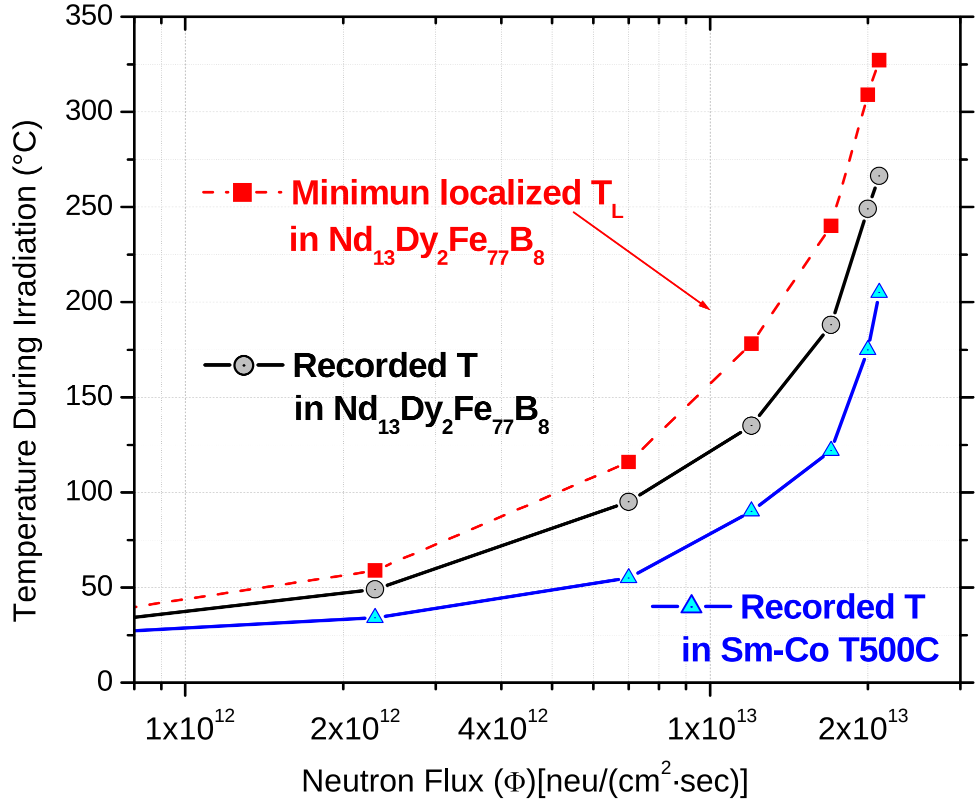
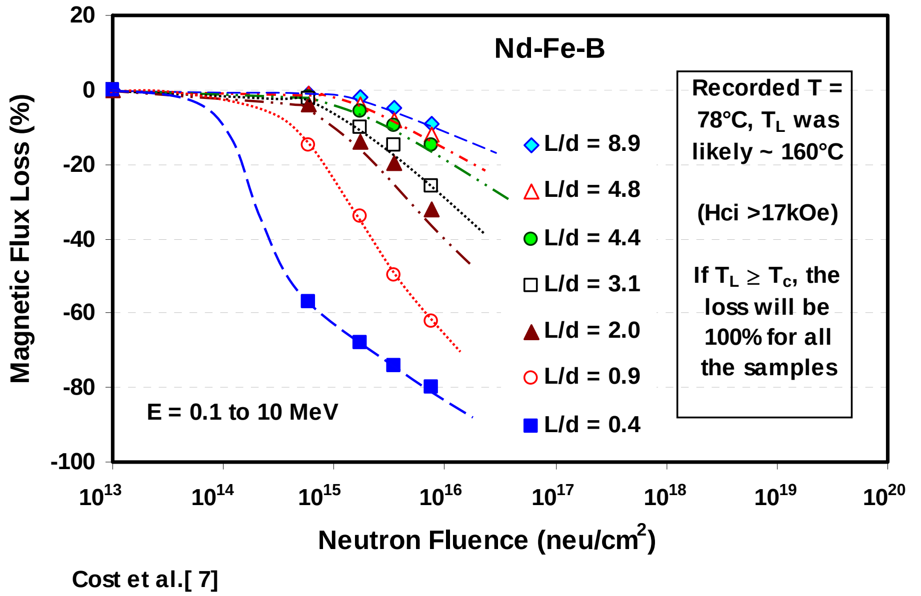
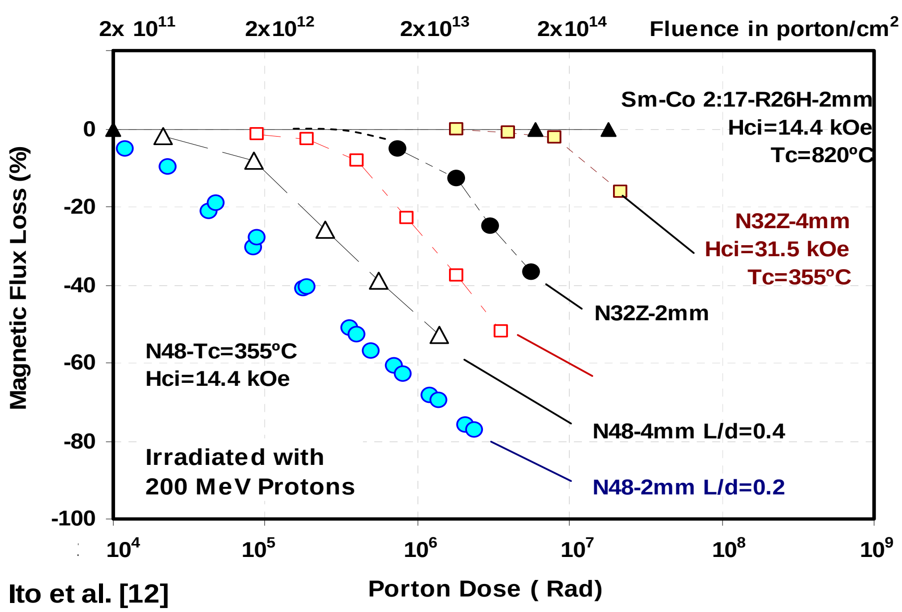


Fig. 2 Temperatures vs. neutron flux in our experiment



(a)



(b)

Fig. 3 Relationship of magnetic flux losses, radiation dose / fluence, and magnet shape L/D (Re-plotted using similar scale in this paper)

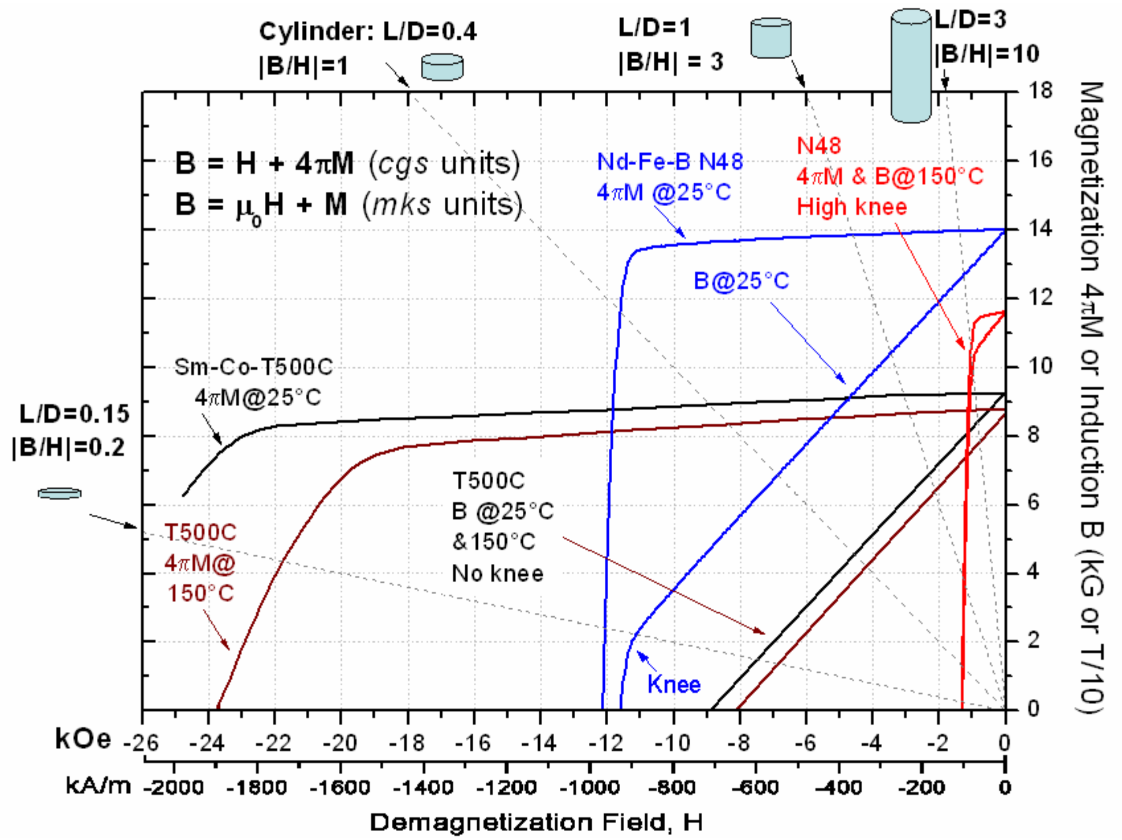
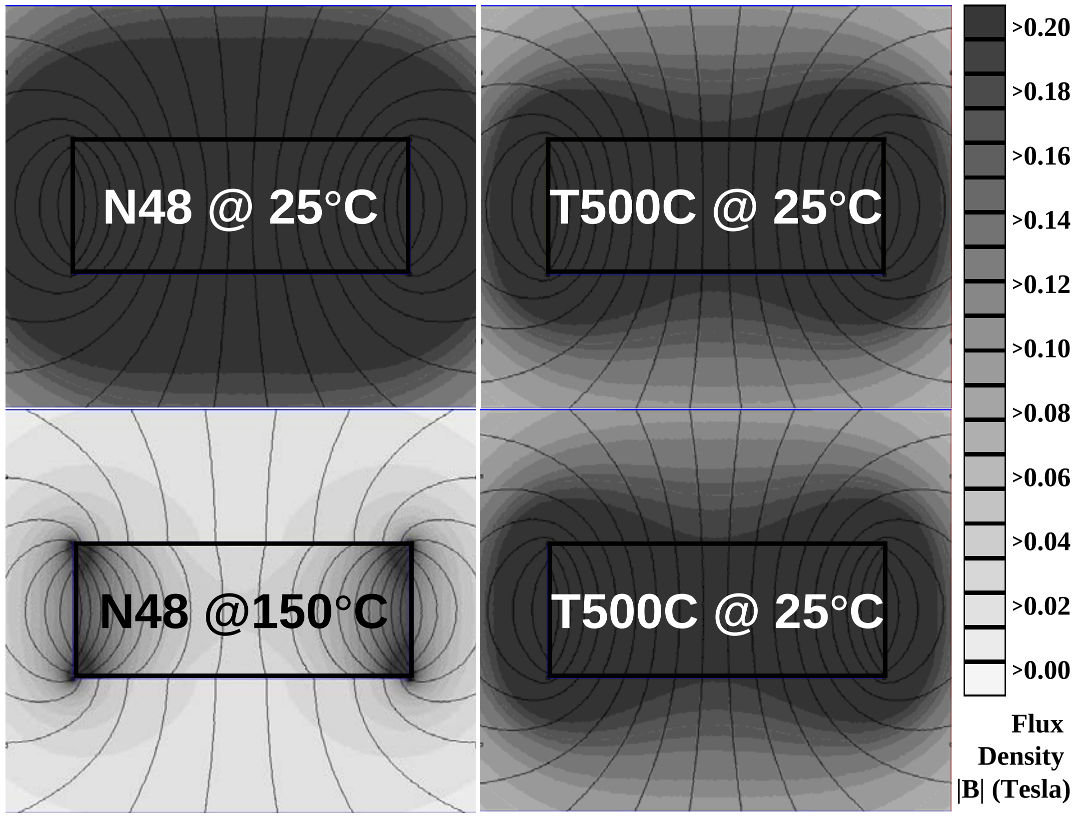
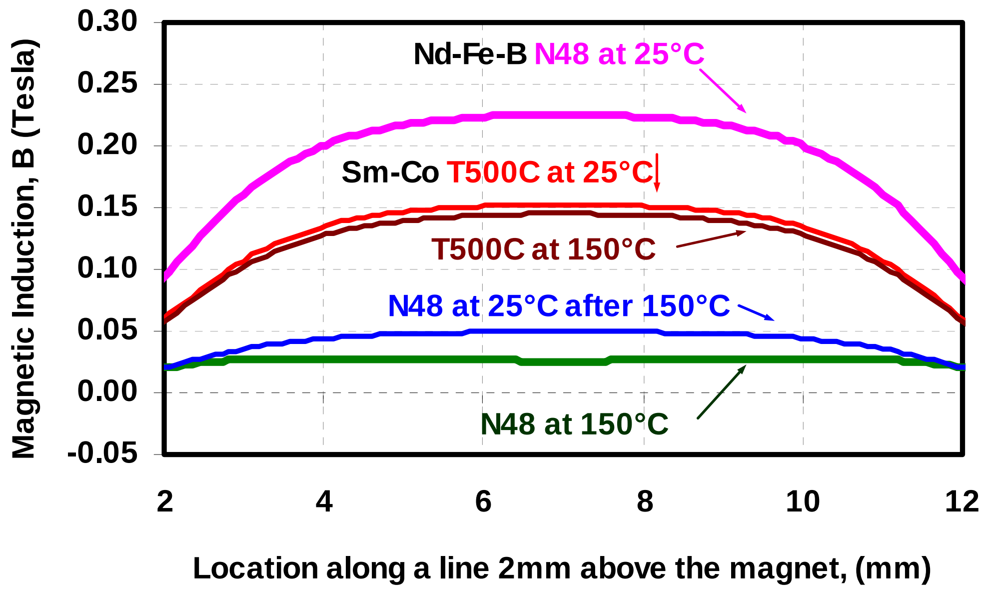


Fig. 4 Demagnetization curves at 25°C and 150°C



(a) 2D distribution of magnetic induction |B|



(b) 1D distribution of magnetic induction B vector (N↑)

Fig. 5 Magnetic flux distributions for Sm-Co T500C and Nd-Fe-B N48 cylinders magnets at 25 and 150°C (L/D = 0.4 and load line or B/H permeance coefficient = 1.0)