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Characterization of magnetic degradation mechanism in a high-neutron-flux environment

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ABSTRACT

Radiation-induced demagnetization of permanent magnets can result in the failure of magnet-based devices operating in high-radiation environments. To understand the mechanism underlying demagnetization, Nd-Fe-B magnets were irradiated with fast and fast plus thermal neutrons at fluences of 10^{12}, 10^{13}, 10^{14}, and 10^{15} n/cm^2, respectively. After irradiation, magnetic flux losses were shown to increase with the fluence. Compared with samples irradiated only with fast neutrons, the samples exposed to the fast plus thermal neutrons have higher magnetic flux losses, which is attributed to the thermal neutron capture reaction of boron. Hysteresis loops of the Nd-Fe-B magnets reveal a slightly increase in the coercivity after irradiation. Full remagnetization of the samples after irradiation was possible, which indicates that structural damage is unlikely an important factor in the demagnetization process at these levels of neutron flux and fluence. Finally, we performed a preliminary Molecular Dynamic (MD) simulation on a cube of ions to obtain a better understanding of the thermal spike mechanism.

1. Introduction

During the past decade, there has been renewed interest in the phenomenon of radiation-induced demagnetization of permanent magnets [1–3]. This interest has been driven by the need for better radiation-resistant magnets in a variety of applications. The high-power ion propulsion engines used by NASA [4], cyclotrons [5], synchrotrons [6], and the high-radiation environments of both the proposed rare isotope accelerator (RIA) and neutrino factory (NF) are among the applications that require radiation-resistant magnets [7,8]. In addition, the accident at the Fukushima Daiichi Nuclear Power Plant, Japan, in 2011 prompted investigations into the ability of robots to conduct sampling, recovery, and rescue missions in intense-radiation environments [7]. The magnets that are used as components in the DC motors of these robots must also be radiation resistant; otherwise, robot joints and wheels will not move even when the battery still provides the power. Therefore, to successfully design radiation-hardened magnets that can withstand high levels of radiation without heavy shielding, it is essential to develop a fundamental understanding of the underlying physics behind the radiation-induced demagnetization mechanism.

The type of radiation, total dose, and dose rate are important factors when considering demagnetization mechanisms. Previous research has indicated that degradation of the magnetic properties of Nd-Fe-B magnets irradiated by gamma rays is small [9–11]. Therefore, the effect of neutron irradiation on Nd-Fe-B magnets is expected to play a bigger role in the process of radiation-induced demagnetization. Since the effects of neutron irradiation on Nd-Fe-B magnets are not well understood, the objective of the present work is to focus on the demagnetization of Ne-Fe-B magnets using irradiation of fast and full-spectrum (fast plus thermal) neutrons from a nuclear reactor. The radiation dose rates at various sites inside a reactor facility where robots are likely to enter are estimated from the Chernobyl reactor site data [12] and from a review of past criticality accidents (an uncontrolled nuclear chain reaction), from which the neutron fluence near the core or near spent-fuel sites is conservatively estimated to be in the range of 10^{14} to 10^{16} n/cm^2 [13]. A previous study applied fast neutron fluence of 4×10^{12} n/cm^2 to Nd-Fe-B magnets [14]. In this work, the fluence levels from 10^{12} to 10^{15} n/cm^2 are reached and their effects are discussed, which corresponds to an absorbed dose estimated at less than 100 Mrad. Although it is known that Sm–Co based magnets have better thermal stability and radiation resistance than Nd-Fe-B magnets, they are brittle, mechanically unreliable, and also have lower energy products with higher material costs, which

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is the reason that Nd-Fe-B magnet, instead of Sm–Co magnet, in focused in this paper.

2. Experimental

2.1. Sample

Commercially available Nd-Fe-B magnet samples manufactured by Apex Magnets Company (M18 × 116DI) were used in the present work. The physical properties of this Nd-Fe-B magnet are listed in Table 1.

Each cylindrical sample has a diameter and thickness of 6.35 and 1.6 mm, respectively. The direction of magnetization was parallel to the cylinder axis.

2.2. Irradiation with neutrons

The irradiation experiments were conducted in the "rabbit" system at The Ohio State University Research Reactor (OSURR). The "rabbit" system is a pneumatic facility designed to rapidly move samples in and out of close proximity to the reactor core. Both the fast neutrons (with an average energy of 1.98 MeV and a most probable energy of 0.73 MeV, which are conveniently referred as 1-MeV neutrons) and fast plus thermal neutrons (with an energy less than 0.4 eV and a peak at 0.0253 eV) were utilized. A cadmium box (1 mm thickness) was used to filter out the thermal neutron component. The temperature during irradiation was approximately 80 °C. Table 2 shows the neutron fluences and the respective neutron flux, *i.e.,* dose rate, achieved for each sample.

2.3. Measurement system

Before and after neutron irradiation, the open-circuit magnetic flux was measured at room temperature using a commercially available Helmholtz coil and an integrating fluxmeter (LakeShore Model 480). Observed flux values for irradiated samples were compared with the original values to determine relative flux losses. Hysteresis loop measurements were performed for each sample before and after irradiation using Physical Property Measurement System with the Vibrating Sample Magnetometer option (PPMS–VSM). The magnetic domain structure was investigated using magnetic force microscopy (MFM). MFM images were obtained using the tapping/lift mode of the Veeco Dimension 3000 Scanning Probe Microscope (SPM). The MFM probe used in this study was coated with a Co–Cr alloy.

3. Results

Fig. 1 shows the neutron-fluence dependence of the magnetic flux loss for the Nd-Fe-B magnet irradiated by fast and fast plus thermal neutrons. The data were plotted against neutron fluence on a logarithmic scale. Notice that the magnetic flux decreased with increasing neutron fluence, regardless of neutron energy. At 10^{12} n/cm^2 neutron fluence, the difference between demagnetization due to fast and fast plus thermal neutron is within statistical insignificance, indicating the thermal neutron contribution is negligible at this level of fluence, which is in agreement with reference [2]. Irradiation by fast plus thermal neutrons resulted in a greater magnetic flux loss compared to the fast-neutron irradiation at 10^{13} and 10^{14} n/cm^2 neutron fluences, respectively, the explanation of which is attributed to the boron consumption by thermal neutron capture and more to the subsequent energetic particle (^{7}Li and ^{4}He) production, which exacerbate the magnetic loss due to fast neutron alone. While the detailed discussion is in the following session, it is noted that the highest fast neutron fluence and the equivalent thermal neutron fluence in our study is about 6.25 times and 272 times higher than that in reference [2], where a radioisotope Cf-252 was the source for fast neutrons and a polyethylene was used as moderator between source and sample to provide thermal-only neutrons.

Other factors such as different dose rate and different irradiation temperature (room temperature in [2] versus 80 °C in our study) may explain the accelerated demagnetization with our higher neutron fluence applied. For example, it took 3 days to reach 3.3×10^{12} n/cm^2 fluence whereas in our case it is 40 s for 10^{12} n/cm^2 neutron fluence. At 10^{15} n/cm^2 fluence, the loss mechanism due to thermal neutron seems to be saturated.

The hysteresis loops for the periods before and after neutron irradiation are shown in Figs. 2 and 3, respectively. There is a clear indication from the enlarged details in Figs. 2 and 3 showing that all 7 samples presented a larger coercivities, albeit minor, than the control sample (except for the one received 10^{13} fast neutron fluence). The difference in the change of coercivity among irradiated samples of the same type are not statistically significant, which could be due to the experimental uncertainty. It is also unclear from Figs. 2 and 3 whether the increase in coercivity is more pronounced when irradiating samples with fast plus thermal neutrons instead of only fast neutrons. Based on the trends of the data in Figs. 3 and 1, the known effect of thermal neutrons from the literature, the sample receiving 10^{13} n/cm^2 fast neutron fluence in Fig. 3 cast doubt on its sample preparation and measurement process. Nevertheless this data point is still preserved in the figure instead of discarded as an outlier.

As previously mentioned, only small changes were found in the hysteresis curves before and after neutron irradiation, which indicates that the neutron irradiation did not cause permanent metallurgical structure changes. The evolution of magnetic microstructure changes was investigated using MFM. Fig. 4(a) and (b) show the MFM images of magnet samples before and after 10^{15} n/cm^2 fast-neutron irradiation, respectively. There is a noticeable difference in the domain structure pattern. The domain pattern in the un-irradiated sample is corrugation and spikes, which are the typical domain structures for sintered Nd-Fe-B magnets. These complex domain structures are formed owing to a balance between the magnetostatic energy and exchange energy [14,15]. Although the corrugation and spike domains still existed after the neutron irradiation, plate domains appeared, as shown in Fig. 4(b). A review of data from the corrugation and spike domains indicates that their easy axes were close to or in the normal of the magnet surface, while the easy axes of the plate domains were parallel or close to the magnet surface [16]. Specifically, the plate domain structure was formed by the nucleation and growth of the reversal domain [17]. This explanation accounts for the loss of magnetic flux, which reached nearly 80% after a 10^{15} n/cm^2 neutron irradiation.

4. Discussion

When interacting with nuclei, fast neutrons typically undergo inelastic scattering: (n, n′)-type reactions. The atom that

Table 1
Physical properties of the Nd-Fe-B sample.

Grade name	Material	Residual magnetization, Br (T)	Intrinsic coercive force, H_{cj} (kA/m)	Coercive force, H_{cb} (kA/m)	Curie temperature, Tc (°C)
N48	$Nd_2Fe_{14}B$	1.38	876	860	320