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TITLE: NEUTRON IRRADIATION STUDY OF Nd-Fe-B PERMANENT MAGNETS MADE FROM MELT-SPUN RIBBONS

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NEUTRON IRRADIATION STUDY OF Nd-Fe-B PERMANENT MAGNETS MADE FROM MELT-SPUN RIBBONS

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ABSTRACT

Radiation-induced changes in the magnetisation of sintered Nd-Fe-B permanent magnets are known to vary widely among specimens produced by different manufacturers. Samples of Nd-Fe-B MAGNEQUENCH magnets, which are made from melt-spun ribbons, have now been studied and show a much reduced sensitivity to neutron irradiation than do sintered Nd-Fe-B magnets. All melt-spun ribbon-based MAGNEQUENCH magnets, i.e., epoxy-bonded, hot-pressed, and die-upset magnets, show essentially the same slow decrease in magnetic remanence with neutron dose. Measurements of the open-circuit remanence B_r/B_{ro} at various times during the irradiation show a decay of only 1.5% of the pre-irradiated value for the MAGNEQUENCH magnets after 1 hour of irradiation, or a dose of 1.4×10^{16} neutrons/cm^2, compared to a 4.6% drop in remanence for the best sintered Nd-Fe-B magnet (Sumitomo 30H) with the same irradiation dose. Moreover, after 5.3 hours of irradiation, the remanence drops by only 3% for the MAGNEQUENCH magnets. Magnets made from melt-spun ribbons are thus the least sensitive to neutron irradiation so far measured for Nd-Fe-B permanent magnets, but are somewhat more sensitive than samarium-cobalt magnets.

INTRODUCTION

Permanent magnets in high radiation environments are known to show changes in magnetic properties, particularly in the magnetic remanence.[1-9] Sintered Sm-Co materials, for example, suffer a 2% loss of remanence with a radiation dose of 1.1×10^{18} neutrons/cm^2 (n/cm^2).[3] Sintered Nd-Fe-B magnets, on the other hand, experienced a 55% loss of remanence after irradiation with 500-MeV protons at a dose of only 10^{14} protons/cm^2.[6] Further studies of sintered Nd-Fe-B magnets using reactor neutrons showed significantly different rates of decay of magnetic remanence with neutron dose for different specimens made by different manufacturers.[7-9] (It should be noted that the flux reported in Ref. 7 is two high by a factor of five, but it is correct in Refs. 8 and 9.) This implies that the sensitivity to neutron irradiation of a Nd-Fe-B magnet depends on the details of its microstructure and on the coercivity resulting from that microstructure. The rate of decay of magnetic remanence for a particular specimen also varies with the temperature at which the irradiation occurs, although the temperature by itself is not high enough to thermally demagnetise the specimen.[7,8] The magnetisation is entirely recoverable upon subsequent remagnetisation,[7,8] implying that no significant damage to the basic magnetic phase occurs during irradiation at doses up to 6.1×10^{16} n/cm^2. The coercivity, however, increases by 20% after irradiation at this dose[7,8] indicating defects are introduced by the irradiation that act as magnetic domain wall pinning sites. Finally, it was found that the rate of decay depends on sample geometry,[9] suggesting a connection between neutron-induced demagnetisation and the geometry-dependent demagnetising field present in the samples. In this report the results of a neutron irradiation study of Nd-Fe-B magnets produced from melt-spun ribbons fabricated at General Motors are presented and compared to those obtained on sintered Nd-Fe-B magnets.

EXPERIMENTAL DETAILS

Samples of epoxy-bonded, hot-pressed, and die-upset Nd-Fe-B magnets were cut into rectangular parallelepipeds with dimensions approximately 1x0.2x0.2 cm. The bonded and hot-pressed samples were obtained from the MAGNEQUENCH plant as production grade materials, and the die-upset sample was fabricated at the GM Research Labs. Each sample was placed in a polyethylene capsule which was fixed to the end of a chromel-alumel thermocouple. The sample could then be lowered into aluminum rabbit tubes at the reactor by the thermocouple. In previous experiments on sintered Nd-Fe-B magnets, reference samples of the same size and shape were also attached to thermocouples and subjected to the same thermal environment but not irradiated.[7,8] This was done to determine any changes in magnetic remanence due to thermal cycling of the samples up to 126°C, and such changes were always found to be negligible.[7,8] In this study the temperature of the samples was typically 80°C during irradiation. No reference sample was therefore used, and we assume all changes in remanence are due to the irradiation.

Irradiation of the samples was carried out at the Omega West Reactor, Los Alamos National Laboratory. The fast neutron flux was about $4x10^{12}$ n/cm^2s. Thermal neutrons were filtered out of the irradiating beam by cadmium shielding so that the energy spectrum was typical for reactor neutrons, but with energies less than 5 eV greatly attenuated. This was necessary with Nd-Fe-B because of the (n,α) reaction between thermal neutrons and ^{10}B which can produce extensive damage not produced for by fast neutrons. The rabbit facility allowed the samples to be removed at selected times during the irradiation for a measurement of the open-circuit remanence at room temperature and then reinserted for further irradiation. Measurements were made by moving the sample through a 500-turn coil. The voltage produced as the magnet moved through the coil was sampled by a Keithley Model 194 high-speed digital voltmeter at a rate of 1kHz. These voltage-versus-time readings were numerically integrated to give a quantity proportional to the open-circuit remanence. Each value of the remanence reported is an average of five or more separate measurements, and the standard deviation of these measurements was typically less than 2% of the remanence.

RESULTS AND DISCUSSION

Figure 1 shows the time (dose) dependence of the open-circuit remanence of irradiated MAGNEQUENCH (MQ) magnets made from melt-spun ribbons. The behavior is essentially the same for epoxy-bonded, hot-pressed, and die-upset MQ magnets, within the scatter of the data. The neutron dose is the flux, $4x10^{12}$ n/cm^2s, times the irradiation time. The decay of the remanence for the MQ magnets is only 3% at 5.3 hours corresponding to a neutron dose of $7.6x10^{16}$ n/cm^2. The best sintered magnet, a Sumitomo 30H, however, lost 4.6% of remanence after only 63 minutes,[9] corresponding to a dose of $1.5x10^{16}$ n/cm^2. Thus, the best sintered magnets exhibit a much faster decay of the remanence than do MQ magnets. These results were analysed for both types of magnets for times up to 63 minutes by fitting the data versus $(time)^{1/2}$ and versus $(time)^{1/4}$ to straight lines. The remanence for MQ magnets decays with time as $(time)^{1/4}$ whereas the data for sintered magnets seem to vary with time as $(time)^{1/2}$. These fits are shown by the solid lines in Figure 1. The scatter of the data for the MQ magnets for times longer than 63 minutes makes it difficult to say whether the $(time)^{1/4}$ behavior is maintained, but the data are at least consistent with such a time dependence.

The decay of magnetic remanence during neutron irradiation is presumably caused by a combination of nucleation of reverse magnetic domains and depinning of domain walls, leading to domain wall motion and demagnetization. The mechanism by which this occurs is either inelastic collisions with the atoms, causing local disruption of the crystal structure and its magnetic anisotropy, or by a magnetic interaction of the neutron's magnetic moment with the magnetization of the material. The former mechanism would be expected to create additional pinning sites, which is seen as increased coercivity in sintered Nd-Fe-B magnets at very high doses.[7,8] Further characterisation of the irradiated magnets is planned in order to directly investigate the effects of neutron irradiation damage on the microstructure. The latter mechanism could be either a magnetic excitation in the magnetisation of a grain, thereby nucleating a reverse domain, or an excitation at a magnetic domain wall pinning site causing depinning of the domain wall, which is then free to move.

The significantly higher resistance to demagnetisation by neutron irradiation of MQ compared to sintered Nd-Fe-B magnets is likely to be associated with the microstructure, and, in particular, how the microstructure gives rise to coercivity in each of these two classes of permanent magnets. From microstructural characterisation,[10,11] both sintered and melt-spun Nd-Fe-B magnets contain $Nd_2Fe_{14}B$ as the magnetically active phase. The crystal grain size in the sintered magnets is typically very much larger than for any of the melt-spun MQ magnets. As a consequence, the coercivity of the sintered magnets relies on the difficulty of nucleating reverse domains in the relatively large grains and on the pinning of domain walls at Nd-rich grain boundaries.[12] In MQ magnets, on the other hand, the nucleation of reverse domains is easier because of the small grains (large demagnetising fields) and the coercivity arises from the strong pinning of domain walls at the larger volume fraction of grain boundaries.[10] If the nucleation of reverse domains and depinning of domain walls are the dominant effects of neutron irradiation, the resulting change in the magnetization of a sintered Nd-Fe-B magnet would be larger than for a MQ magnet. This is because the domain walls can move further in sintered Nd-Fe-B magnets and thereby reverse more of the magnetization for each nucleation or depinning event compared to MQ magnets where the domain walls would quickly become pinned again. It may also be true that the strength of domain wall pinning is important, and that the probability of a neutron causing a depinning event is much less in MQ magnets than in sintered magnets because of the nature of the grain boundary phases and microstructure.

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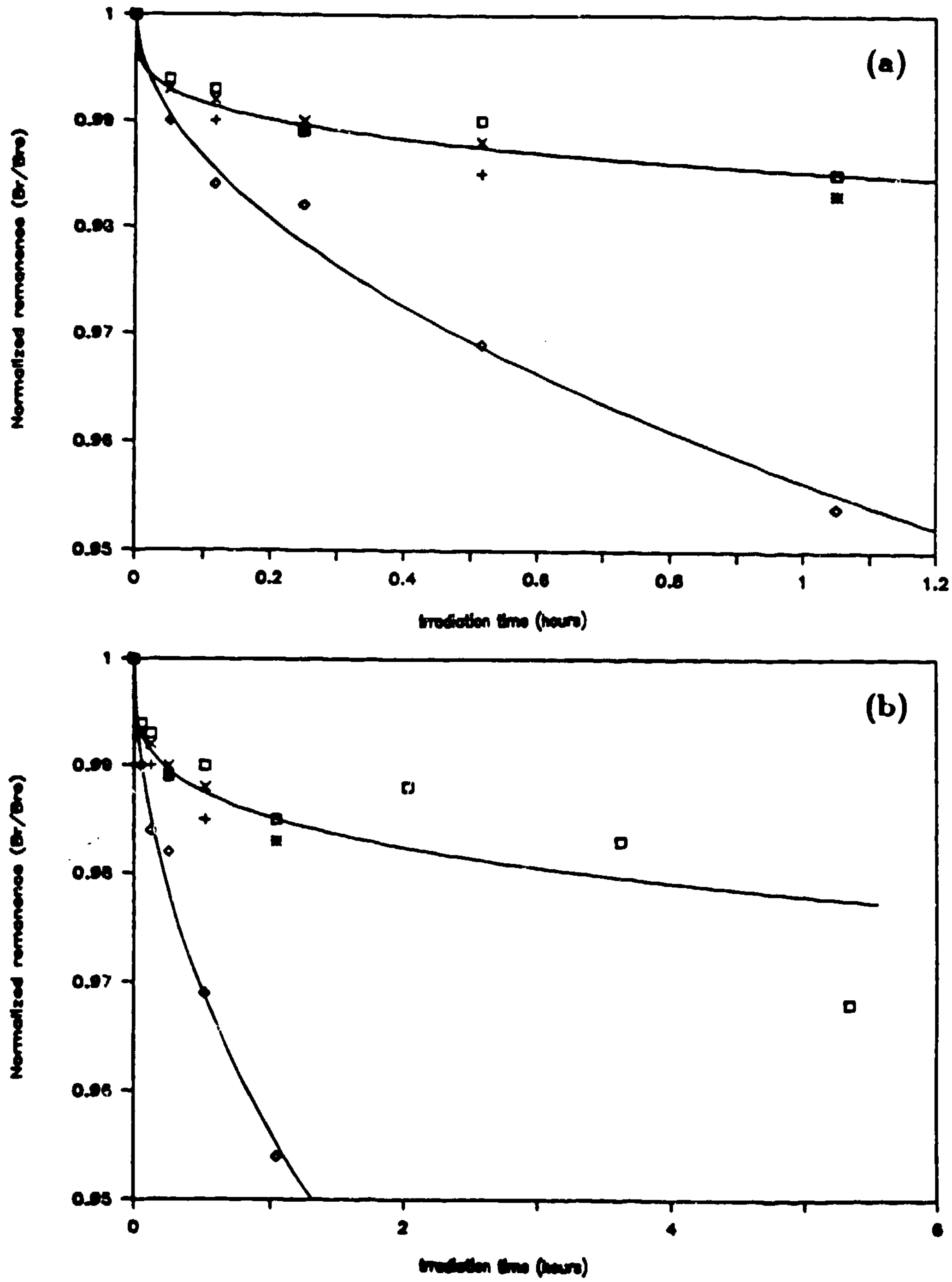

Fig. 1. Decay of open-circuit remanence is shown for epoxy-bonded (+), hot-pressed (x), and die-upset (squares) MAGNEQUENCH magnets, and for a Sumitomo 30H sintered (diamonds) Nd-Fe-B magnet. The data are plotted as normalised remanence versus irradiation time (dose) up to 63 minutes (a) and up to 5.3 hours (b). The solid lines represent fits to the data for irradiation time up to 63 minutes (see text).