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RADIATION EFFECTS IN RARE-EARTH PERMANENT MAGNETS

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ABSTRACT

Nd-Fe-B and Sm-Co permanent magnets have been irradiated with fission neutrons and gamma rays. Irradiated samples were periodically removed for room temperature measurements of the open-circuit remanence. Hysteresis loops were measured before and after irradiation. For neutron irradiation, two magnets showed a rapid loss of remanence, while a third decayed more slowly. Irradiation in the Omega West Reactor at Los Alamos with fast neutrons caused the fast-decay samples to have an initial loss of remanence of 1% for irradiation at 350 K to a fluence of 10^{15} n/cm^2. Both $SmCo_5$ and Sm_2Co_{17} magnets showed excellent resistance to radiation-induced loss of remanence for neutron irradiation to a fluence of 2.6×10^{18} n/cm^2. Results for gamma irradiation are presented and discussed in terms of possible mechanisms for radiation-induced loss of magnetic properties.

INTRODUCTION

The use of permanent magnets in accelerators allows reduction of the weight compared to using electromagnets, both in the magnet itself and in the elimination of the power supply. To be used successfully in a location where scattered radiation is present, the permanent magnets must be capable of resisting decay in remanence due to this radiation. Radiation produced by proton accelerators operating at energies of 50 to 100 MeV is expected to consist of neutrons and gammas produced when the fringes of the accelerated beam strike structures located physically near the beam line. A rough calculation of the total fluence of neutrons expected for a 50 MeV accelerator gives a fluence of about 10^{15} n/cm^2 over the operating lifetime (approximately 200 h) of the accelerator.

Previous applications of permanent magnets in dipole and quadrupole magnets have specified the use of Sm-Co material. Previous work [1-4] has shown that Sm_2Co_{17} magnets are the most resistant to radiation-induced decay of the remanence, but that $SmCo_5$ magnets are also very resistant to doses in the 10^{15} n/cm^2 range. The recently developed Nd-Fe-B magnet family has been of interest to accelerator designers due to the high remanence and intrinsic coercivity of these materials. The sensitivity of these magnets to temperature-produced changes in magnetic properties has led to concern over their stability in a radiation environment. As of 1986, the one study which had been done [4] on radiation effects to Nd-Fe-B magnets showed them to be orders of magnitude more sensitive to charged particle radiation than the Sm-Co magnets. We have previously reported some initial results of irradiations of Nd-Fe-B magnets [5]. We found that while the decay of remanence is more rapid than that of the Sm-Co family, the decay for irradiation at 350 K is only about 1% at a fluence of 10^{15} n/cm^2. A comparison is made with Sm-Co magnets irradiated in the same neutron environment. It is also shown that gamma irradiation by itself has little effect on the remanence of even the Nd-Fe-B magnets.

METHOD

Magnets

The Nd-Fe-B magnets were obtained from three different manufacturers: Crucible Materials Corporation, I. G. Technologies, and Hitachi Magnetics Corporation. The identification and alloy system of the magnets from each manufacturer are given in Table I along with the pre-irradiation values for the residual induction, and intrinsic coercivity specified by the company. The magnets from all three manufacturers were prepared using powder metallurgy combined with thermal treatments unique to each manufacturer.

Two types of Sm-Co magnets were studied, $SmCo_5$, and Sm_2Co_{17}. Both were obtained from I. G. Technologies and were fabricated from powder starting material. The magnetic properties are also given in Table I.

Sample Preparation

The materials were received from the manufacturer in the form of blocks with the largest dimension of roughly 20 mm. Small samples required for the irradiation were cut from these blocks using a low-speed diamond saw. Final samples were typically 7 mm long with a square cross-section from 1 mm to 2 mm on an edge. The magnetization was in the long direction.

After being cut, the samples which were not yet magnetized to saturation were magnetized, either in a magnetic charging unit with charging fields up to 100 kOe, or by measuring the complete hysteresis loop using a vibrating sample magnetometer at 250 K (-23°C). A superconducting magnet applied a final field of at least 55 kOe. Saturation, as determined by the hysteresis loops, typically required a field of 40 kOe or less.

Irradiation

Neutron irradiations were done at the Omega West Reactor at Los Alamos using the epithermal cadmium port. This port is located immediately adjacent to the core fuel rod array and is shielded with cadmium in order to attenuate the thermal neutron flux by roughly 10^4. The flux of fast neutrons ($E \geq 0.1$ MeV) was roughly 2×10^{13} n/cm$^2\cdot$s as measured by dosimetry foils. The neutron energy spectrum for a port symetrically located on the other side of the core, but without cadmium shielding, has been reported [6] and is in good agreement with our measurements for this facility.

For the irradiation, two samples were placed in a polyethylene rabbit in which they were separated by plastic spacers so that they did not interact magnetically. The rabbit was lowered into position in the reactor port with a chromel-alumel thermocouple, the junction of which was attached to a dummy sample for temperature measurement. This arrangement allowed easy insertion and removal of the rabbit from next to the reactor core. Sample temperature during irradiation was kept close to that of the cooling water of the reactor, roughly 350 K (77°C), by introducing a continuous flow of helium gas. Several samples were also irradiated without helium at a temperature of approximately 426 K (153°C). Samples were held in the reactor for the desired irradiation times and then removed for measurement of the remanence. After each measurement they were reinserted for continuation of the irradiation.

To determine whether gamma irradiation by itself could produce an observable decay in the remanence, several of the samples were irradiated in a gamma cell using ^{60}Co. The dose rate in the gamma cell was approximately 16 Rad/s. A measurement of the temperature using a thermocouple indicated no change in temperature compared to the ambient temperature outside the cell. As with the neutron irradiated samples, the same samples were irradiated for increasing periods of time, with the remanence being measured at intervals during the irradiation.

Measurement

Remanence was measured by mechanically moving the sample through a 500-turn pickup coil and monitoring the voltage output of the coil. This voltage was integrated by computer to obtain the area under the voltage-time curve from which a value proportional to the open circuit remanence was calculated. These measured values were less than those obtained from the hysteresis curves because all of the lines of flux were not cut by the pickup coil, so values reported were normalized to the pre-irradiation value. Typically each reported value was obtained from an average of 20 or more separate readings. This resulted in a fractional uncertainty which was usually less than 0.02.

RESULTS AND DISCUSSION

Table II shows the different samples which have been neutron irradiated at 350 K (77°C) along with the total fluence and the remanence normalized to the pre-irradiation value. Several findings can be made from this table. For

Nd-Fe-B magnets we observe that the CRUMAX 282 and NeIGT 27H samples are sensitive to radiation by fast neutrons while the HICOREX 94EB material is relatively insensitive. This finding that some Nd-Fe-B magnets can withstand irradiation is particularly important since previous investigations have only reported that these magnets are highly sensitive to neutron irradiation [4,5]. This new knowledge suggests that the metallurgical processing can markedly affect the microstructure, which is important in determining the radiation-resistance.

In order to better understand the decay of Nd-Fe-B magnets, the effects of temperature, modification of the neutron energy spectrum, and irradiation with gamma rays were investigated. A comparison was also made between radiation effects on the Nd-Fe-B and Sm-Co magnets.

First, non-irradiated samples of Nd-Fe-B magnets prepared from the powder starting material were annealed at 423 K (150°C) to determine if any part of the remanence loss was due to instability at the irradiation temperature. These samples showed an initial 1.5% drop in remanence after heating to this temperature, but no further decay of remanence following periodic measurements for times up to 15,300 seconds. Thus we deduce that the remanence loss is due solely to the irradiation.

Because of the high cross section of boron for thermal neutrons and the relatively high energy of the alpha particles emitted by the reaction, it was desirable to check whether thermal neutrons were contributing a large fraction of the atomic displacement damage. To check this we repeated the irradiation of the two samples which had shown decay, but on two similar samples which were enclosed with 0.4 mm of cadmium which attenuated thermal neutrons by an additional factor of 10^4. The results of this are shown both in Table II and in Fig. 1. In this figure, the remanence normalized to the pre-irradiation value is plotted versus both time of irradiation and neutron fluence. Fig. 1 shows that there is a negligible difference between irradiation-induced decay of remanence with and without the extra cadmium shielding. Thus we eliminate the reaction between boron and thermal neutrons as being important to the results for Nd-Fe-B magnets irradiated in our cadmium wrapped rabbit tube.

It was desirable to know the extent to which Nd-Fe-B magnets are sensitive to other kinds of irradiation. Thus we gamma irradiated samples of the same magnets which showed remanence decay due to fast neutrons. Following irradiation for approximately 35 days, the samples reached a dose of 49 MRad; however, as Table I indicates, this resulted in a decrease in remanence of less than 1%. Since gamma irradiation only displaces individual atoms from their lattice sites, while a single neutron irradiation event results in a cascade of displaced atoms, this tells us that widely separated single displaced atoms are not able to produce a measurable loss of magnetization.

In order to provide further understanding of radiation damage effects we investigated Sm-Co permanent magnets. From Table II we see that Sm-Co magnets, both $SmCo_5$ and Sm_2Co_{17}, are quite insensitive to radiation damage by fast neutrons. The remanence of both of these magnets was only minimally changed after 41 hours of irradiation to a fluence of 2.6×10^{18} n/cm^2. Sm-Co magnets would be the obvious choice for high-irradiation environments except when the higher energy product of the Nd-Fe-B magnets is desired.

Examining Fig. 1 we see that the initial half of the decay curves for the samples from the powder starting material are in good agreement even though the samples are from two different manufacturers. After irradiation for roughly three hours (at which point the magnets have lost 70% of their remanence) these decay curves start to differ; apparently minor differences in magnetic structure become important as full demagnetization is approached.

We have analyzed the full decay curves in Fig. 1 to determine whether the process of irradiation-induced loss of magnetization fits any of several different models for the reaction kinetics. The predicted fits to all for the these models were poor. The reaction clearly does not follow simple first-order kinetics with a single relaxation time. However, first-order kinetics could be involved if there were a fairly broad distribution of relaxation times. From the slope of Fig. 1 we calculate that if this distribution of relaxation times is lognormal, then it has a width parameter, β=2.5, which means that the distribution is slightly less than two decades wide at half maximum.

For some of the samples hysteresis loops were measured before and after neutron irradiation. Fig. 2 shows such a pair of loops for the Nd-Fe-B magnet NeIGT 27H irradiated at 426 K (153°C) and measured at 250 K (-23°C). Note that the remanence, which decreased roughly 80% during the irradiation (see the start of the solid curve in the second quadrant), recovered to the pre-irradiation value after being magnetically saturated. This would seem to indicate that the same magnetic structure is developed after irradiation and thus that irradiation does not appreciably change the metallurgical structure. Interestingly, the coercive force as shown in Fig. 2 was increased roughly 20% by the irradiation. A possible explanation for this is that the damage introduces extra pinning sites for the magnetic domain walls making them more difficult to move.

An estimate of the rate for atomic displacements due to neutron irradiation has been made and compared with the decay rate of the magnetization [5]. For a flux of 2×10^{13} $n/cm^2\cdot s$, an elastic scattering cross section of 3×10^{-24} cm^2, and assuming 500 to 5000 displaced atoms per primary knock-on event the sample will have roughly 10^{-7} displaced atoms per second. Since appreciable remanence decay occurs in the first 1000 seconds, the loss is occuring even though only a small fraction of the atoms is being displaced. A mechanism which agrees with this high loss rate per atomic displacement is radiation-induced nucleation of reverse domains.

This mechanism can be expected to be extremely sensitive to the structure of the magnetic domains. This, in turn, will be determined by the microstructure of the phases. A study of both of these kinds of structures in Nd-Fe-B magnets fabricated from powder starting material [7] has identified the major phase, $Fe_{14}Nd_2B$, as being responsible for the hard magnetic properties. More recently, Hadjipanayis, et. al. [8] have investigated the microstructure of this phase in Nd-Fe-B magnets from different manufacturers and found variations in grain size for this magnetic phase of more than a factor of one thousand within a given magnet; they also found large differences between magnets of the different manufacturers. Such wide differences in microstructure could easily account for the marked differences in resistance to radiation damage which are observed in Fig. 1.

At present our understanding of how coercive properties depend upon structure is not fully clear. We do know, however, that differences in grain size and grain boundary structure can cause different coercivity mechanisms to operate [9]. The determination of how these mechanisms affect the resistance to radiation damage needs further work. Certainly it will be of value to determine how the microstructure of Hitachi HICOREX 94EB differs from that of the other magnets, since it resists neutron irradiation better than its counterparts by a factor of 25. If, as we might expect, a fine grain structure is one of the key factors, then it will be important to investigate magnets fabricated from melt-spun starting material since they have an extremely fine grain size (25 nm) as well as a thin layer of amorphous material at the grain boundaries to promote pinning of domain walls [9].

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REFERENCES

[1] R. D. Brown, E. D. Bush Jr., and W. T. Hunter, Los Alamos National Laboratory report LA-9437-MS, July 1982.

[2] F. Coninckx, W. Naegele, M. Reinharz, H. Schoenbacher, and P. Seraphin, CERN/SPS, TIS-RP/IR/83-07, Feb. 1983.

[3] H. Spitzer and A. Weller, Kernsforschungsanlage Jülich Report SNQ 1 N/BH 22/05/84, May 1984.

[4] E. W. Blackmore, IEEE Trans. Nucl. Sci., NS-32, 3669-3671, Oct. 1985.

[5] J. Cost, R. Brown, A. Giorgi, and J. Stanley, Los Alamos National Laboratory report LA-UR-86-4245. Submitted for publication to IEEE Trans. Magn., Dec. 1986.

[6] W. A. Coghlan, F. W. Clinard Jr., N. Itoh, and L. R. Greenwood, J. Nucl. Mater., 141-143 , 382-386, 1986.

[7] J. F. Herbst, J. J. Croat, F. D. Pinkerton, and W. B. Yellon, Phys. Rev. B, 29, 4176, 1984.

[8] G. C. Hadjipanayis, Y. F. Tao, and K. R. Lawless, IEEE Trans. Magn., MAG-22, 6, 1845, 1986.

[9] R. K. Mishra, J. Magn. and Magn. Mater., 54-57, 450, 1986.

Table I

Pre-Irradiation Properties of Rare-Earth Magnets
Used in This Radiation Damage Study

Alloy	Manufacturer	Material	Remanence (Gauss)	Intrinsic Coercive Force (Oersted)
Nd-Fe-B	Crucible Materials	CRUMAX 282	10,800	>17,000
Nd-Fe-B	I. G. Technologies	NeIGT 27H	10,200	>17,000
Nd-Fe-B	Hitachi Magnetics	HICOREX 94EB	11,500	15,000
$SmCo_5$	I. G. Technologies	INCOR 18	8,800	>16,000
Sm_2Co_{17}	I. G. Technologies	INCOR 22HE	9,600	>16,000

Table II

Results of Irradiations of Rare-Earth Permanent Magnets

Sample	Type of Irradiation	Irradiation Temperature (K)	Irradiation Time (s)	Total Fluence (n/cm^2)	Final Fluence (B_r/B_{ro})
CRUMAX 282	neutron	350	1.26×10^5	2.5×10^{18}	0.209
NeIGT 27H	neutron	350	1.26×10^5	2.5×10^{18}	0.132
HICOREX 94EB	neutron	350	1.92×10^4	3.8×10^{17}	0.860
CRUMAX 282	neutron Cd shielded	350	1.86×10^3	3.7×10^{16}	0.763
NeIGT 27H	neutron Cd shielded	350	1.86×10^3	3.7×10^{16}	0.784
CRUMAX 282	gamma	300	3.05×10^6	48.8*	1.008
NeIGT 27H	gamma	300	3.05×10^6	48.8*	1.003
INCOR 18	neutron	350	1.29×10^5	2.6×10^{18}	1.003
INCOR 22HE	neutron	350	1.29×10^5	2.6×10^{18}	0.998

*Fluence in MRad.

FIGURE CAPTIONS

Fig. 1. Retained fraction of pre-irradiation remanence vs time and fluence for Nd-Fe-B magnets irradiated with fast neutrons at 350 K (70°C). Some samples were wrapped in cadmium to further attenuate thermal neutrons.

Fig. 2. Hysteresis loops at 250 K (-23°C) for a NeIGT 27H sample before and after irradiation.

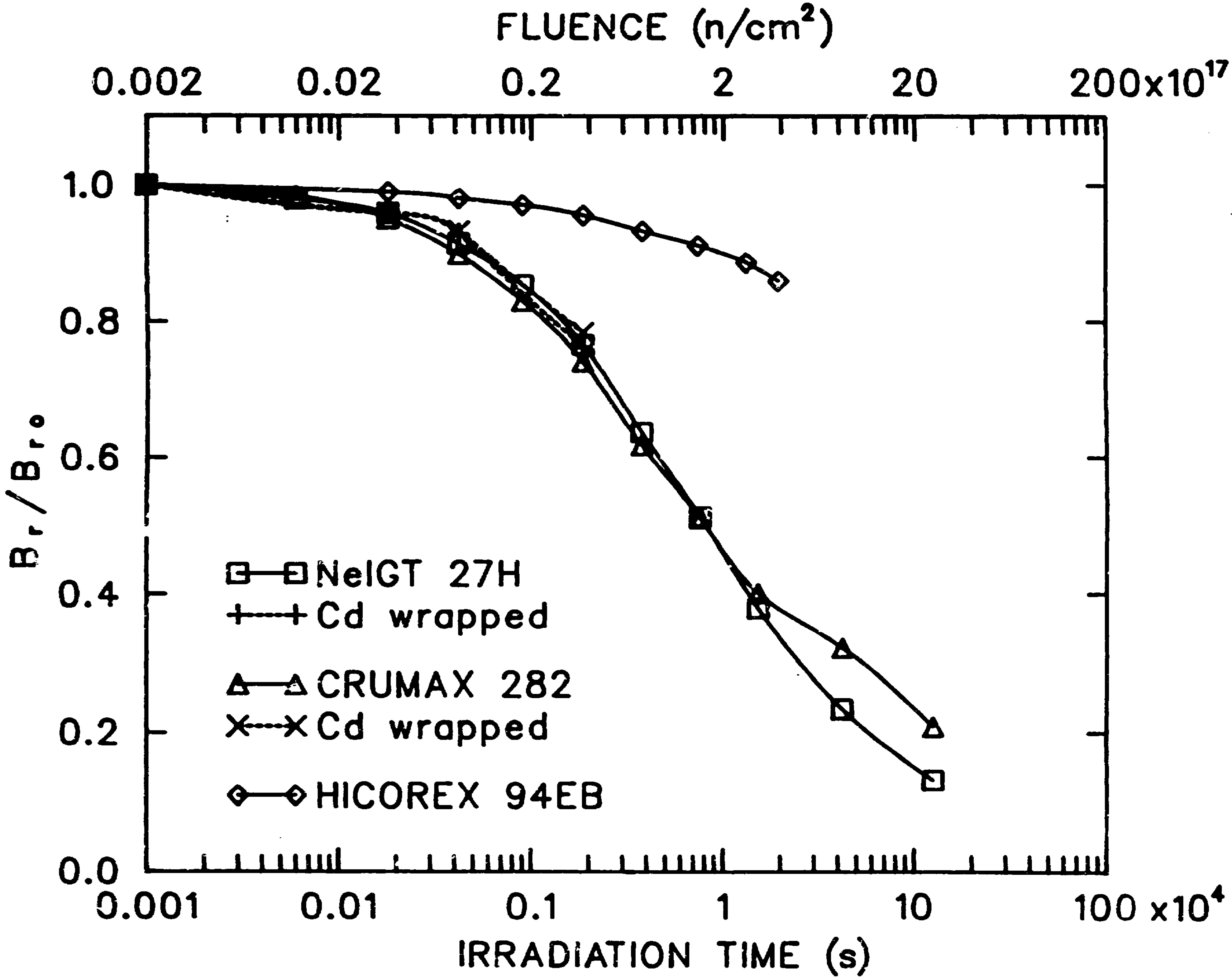
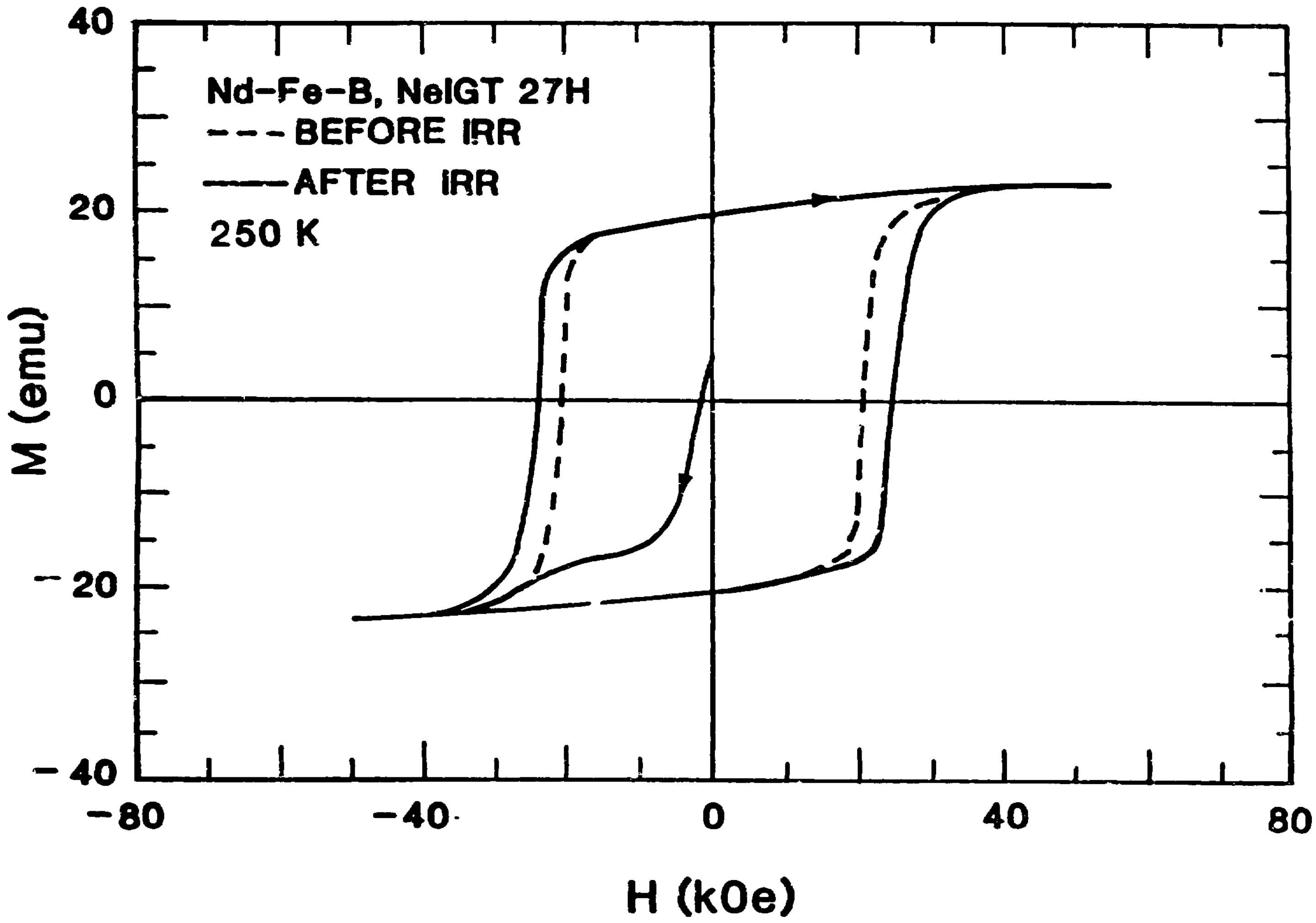

FLUENCE (n/cm²)
0.002
0.02
0.2
2
20
200x10¹⁷
1.0
0.8
0.6
0.4
0.2
0.0
Br/Bro
NeIGT 27H
Cd wrapped
CRUMAX 282
Cd wrapped
HICOREX 94EB
0.001
0.01
0.1
1
10
100 x10⁴
IRRADIATION TIME (s)

FIGURE 1



Nd-Fe-B, NeIGT 27H
---BEFORE IRR
——AFTER IRR
250 K
40
20
0
-20
-40
M (emu)
-80
-40
0
40
80
H (kOe)

FIGURE 2