FAST NEUTRON DAMAGE STUDIES ON NdFeB MATERIALS[*]

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Abstract

Many materials and electronics need to be tested for the radiation environment expected at linear colliders (LC) since both accelerator and detectors will be subjected to large fluences of hadrons, leptons and γ's over the life of the facility[1]. While the linacs will be superconducting, there are still many uses for NdFeB in the damping rings, injection and extraction lines and final focus. Our understanding of the situation for rare earth, permanent magnet materials was presented at PAC03[2]. Our first measurements of fast neutron, stepped doses at the UC Davis McClellan Nuclear Reactor Center (UCD MNRC) were presented at EPAC04[3]. We have extended the doses, included other manufacturer's samples, and measured induced radioactivities which are discussed in detail.

INTRODUCTION

This work is a continuation of work reported recently at EPAC04[3] whose goal is to improve systems such as LCs over their lifetimes by providing a predictive understanding of radiation damage mechanisms based on more controlled and systematic experiments. We give further results for a study[1] of the effects of fast neutrons on $Nd_{2-x}Fe_{14}B$ blocks where x represents substitution of other rare-earths such as Dy, Pr or Tb. Previous studies[4] have shown these substitutions may improve H_{ci} with a high linear correlation of 0.96 as well as radiation resistance (RR) with a correlation of 0.87 and thus, a good correlation of 0.78 between RR and H_{ci}. While temperature was controlled, neither T_s, the stabilization T, nor the effective operating point or load-line were specified although the magnets were in an essentially open circuit configuration. Such questions were addressed in [2] where a new type of two-pole, offset quadrupole was proposed to test the interplay between the H_{ci} of an unloaded block and its loaded, operating point in magnetic circuits which can vary quite dramatically - even over a single PM block in typical magnetic multipoles[5].

The UCD MNRC has a number of areas for irradiating samples with neutron fluxes $\leq 4.5\cdot10^{13}$ n/cm^{2}s. We used a specialized area (NIF) that allows fast neutron irradiations with 1 MeV equivalent neutron fluxes $\leq 4.2\cdot10^{10}$ n/cm^{2}s while suppressing thermal neutrons and γs. We irradiated individual blocks and magnets there as described in [2, 3]. Below, we describe our specific use of the reactor, the measurements and our latest results.

LOGISTICAL AND OTHER PROBLEMS

In reviewing experiments in this area, common characteristics emerge that explain both the difficulty and scarceness of systematic, controlled experiments [2]. In fairness, even a brief consideration of such a program shows many questionable and hard to control circumstances such as the difficulty of handling and measuring the PM test materials even when they are not radioactive. Altogether, this implies that a considerable number of people and jurisdictions become involved. Thus, there is ample opportunity to damage the blocks or change their magnetic properties in ways totally unrelated to radiation damage e.g. most frisking detectors or monitors have steel components that can easily lead to chipped or broken blocks.

While there are many problems, there are many uses for these results. The importance of the work for accelerators is clear[1, 2, 3] but there are also opportunities in space applications and materials research such as defect and domain manipulation e.g. there is evidence that some forms of damage may improve systems after remagnetization.

CHOICE OF PM BLOCKS & MAGNETS

Most experiments use unloaded, single blocks whose results are difficult to interpret[2]. In magnets, PM dipoles should be less susceptible to damage followed by undulators, wigglers, and magnetic multipoles due to variations in $\vec{M}$ over different block types *and* especially variations in $\vec{H}_{ext}$[5]. This is clear from Fig. 2 of [2] but especially Fig. 4 of [5] and explains our design of an asymmetric quadrupole with simple dipole geometry – shown in [2] for a large gap $G \geq l_x, l_y, l_z$ where it was discussed in detail, together with measurements. All of those blocks were nickel-plated but were undoped $Nd_2Fe_{14}B$ as opposed to those in Table 1.

Table 1: Characteristics of open-circuit irradiated blocks

Block	$\mathbf{B}_r$[kG]	$\mathbf{H}_{ic}$[kOe]	$\mathbf{H}_{bc}$[kOe]	$\mathbf{BH}_{max}$
N34Z	11.60	30	11.3	32.7
N50M	14.15	15	13.1	47.7
HS36EH	11.60	24	11.3	32.8
HS46AH	13.41	14	12.9	44.3

Table 1 lists blocks that were open-circuit irradiated. Figure 1 of Ref. [3] gave the demagnetization curves for the Shin-Etsu blocks N50M and N34Z. All blocks had l_z=6 mm, l_x=9 and l_y=25.4 mm with weights of 10.3 g. These dimensions allowed good uniformity of dose throughout the volume by passing the flux perpendicular to the long dimension[2, 3].

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MAGNETIC MEASUREMENTS

For the individual blocks and magnets, a special Hall probe fixture was made to do field scans in combination with Helmholtz magnetization measurements. Examples were given in Fig. 3 and Tables 1–2 of [2]. Here, Table 2 gives results for the block types in Table I where $<\mathrm{M}_y>$ is the average over all runs of one component of **M** where x is the easy axis direction and $\mathbf{M}^i_{xy}$ is the initial measurement, before irradiation, of the easy axis projection in the xy plane. The strength errors are small and repeatable even for the small blocks. The differential damage $\delta\mathbf{M}_{xy}/\delta\mathbf{D}$ is in G/Gy. The Hitachi blocks saw 2 doses totaling 28 Gy(Si) of 1 MeV equivalent neutrons. The Shin-Etsu blocks saw 6 doses totaling 77.8 Gy(Si). Over these ranges the damage is linear. Stepped doses are continuing.

Table 2: Demagnetization results for blocks in Table I

Block #	$<\mathbf{M}_y>$[G]	$\mathbf{M}^i_{xy}$[T]±[G]	$-\delta\mathbf{M}_{xy}/\delta\mathbf{D}$
HS36EH	-62±29	1.1595±0.8	0.00±0.03
HS46AH	-154±18	1.3298±2.0	3.84±0.07
N34Z1	-301±35	1.1106±1.8	0.49±0.02
N50M1	-50±38	1.3706±1.1	1.81±0.02
Ref(#3)	-350±57	1.0748±6.7	0.78±0.08
Ref(#5)	93±177	1.0622±2.2	0.64±0.03
Ref(#7)	466±183	1.0714±0.9	1.04±0.03

DAMAGE MEASUREMENTS

The MNRC provides a number of areas for irradiating samples with neutron fluxes up to 4.5 x 10^{13} n/cm^{2}s. Here, we used the 1 MeV equivalent neutron facility NIF as well as their Ge detector based γ-spectroscopy setup.

Radiation Monitoring

Several methods were used to control and monitor the radiation dose and temperatures that the various PM blocks and magnets were subjected. First, a low power level of 350 kW was set for the MNRC reactor to control heating, dose rate and uniformity of dose. The containment vessel was rotated with a six-sided holder to keep the magnets well isolated from one another and provide dose uniformity. The first run was for 23 minutes and contained an unused neutron/photon dosimeter pair consisting of a PIN diode for the neutron dosimetry that is orders of magnitude more sensitive to neutrons than gammas and MOSFET photon dosimeters where the reverse situation obtains[6]. Also, two sulphur tablets were included whose radioactivity was measured to determine average fluence for each run.

RADIOACTIVITY STUDIES

General Background

While we know that binary $\mathrm{Sm_xCo_y}$ compounds are more radiation resistant than those of NdFeB, they also tend to be weaker, more expensive and can become quite radioactive through the large neutron capture cross sections going to long-lived isotopes Co^{60} [7] and Sm^{151} with 5.3 and 93 y lifetimes. Nevertheless, they may be preferred – especially if damage is important. NASA recently awarded a contract to study radiation and thermal stability effects for SmCo in space applications.

In contrast, NdFeB is generally cheaper and stronger but less radiation resistant with radioactivity characteristics that are not as well understood esp. when doped with other rare earth substitutions. Those candidates are not generally considered from this standpoint. They also tend to be proprietary and changing e.g. Shin-Etsu considers N34Z to be a 5$^{\mathrm{th}}$ generation material that is still under development while N50M is described similarly but as 6$^{\mathrm{th}}$ generation. Their compositions are proprietary.

Several characteristics of rare earths are of interest. $_{61}$Pm has no stable isotope and only the Z,N=odd,even elements have isotopes with 100 % stable fractions i.e. $_{59}\mathrm{Pr}^{141}$, $_{65}\mathrm{Tb}^{159}$, $_{67}\mathrm{Ho}^{165}$ and $_{69}\mathrm{Tm}^{169}$. La, Eu and Lu have two apiece while $_{60}$Nd, $_{62}$Sm, $_{64}$Gd and $_{66}$Dy each have seven with Nd running from A=142-150.

$\mathrm{Nd_2Fe_{14}B}$ is 26.7% Nd by atomic weight, Fe 72.3% and Boron 1%. Fe has 4 stable isotopes ranging from A=54-58 with $_{26}\mathrm{Fe}^{56}$(91.7%) while Ni, the plating material for all samples, has 5 ranging from A=58-64 with $_{28}\mathrm{Ni}^{58}$(67.9%). While only 1 % is Boron, different models[4, 8] suggest it is the major factor in demagnetization. It has two stable isotopes (A=10 & 11) with $_5\mathrm{B}^{10}$ the worst because of its lighter mass and very large neutron capture cross section of 3.8 kb (compare to Co^{59} with 36.6 b) so it provides two mechanisms for demagnetization. $_{64}\mathrm{Gd}^{157,155}$ has 242,61 kb and $_{66}\mathrm{Dy}^{164}$ has 3 kb cross sections. A major difference is that B^{10} neutron induced fission can lead to permanent demagnetization whereas the others lead to similar, heavy, stable, isotopes.

Radioactivity Measurements

Table 2 of Ref. [3] gave results for three different types of blocks from our first 46 min run in NIF at MNRC to obtain trace elements, sources and levels of radioactivity. “Ref” referred to a 3-block magnet with a thin iron return yoke. However, because the overall volumes of material and their geometries in the 3 samples were comparable, for uniformity of dose, the results were directly comparable. For undoped NdFeB in an Fe yoke there was very low induced radioactivity.

The sources of most γ lines were identified e.g. n-capture on Fe^{58}, Nd^{146}, Nd^{150} or Tb^{159}. The latter was the largest source with many lines from Dy^{160} via the β^- decay of Tb^{160}. Most of these were not tabulated. We note that there were many errors and omissions in Ref. [10] used for EPAC04 - most of which were corrected for the Table. Also, while there were many lines greater than 1 MeV, detector efficiencies were not considered.

Table 3 gives our latest results for similar size samples of NdFeB from Shin-Etsu and Hitachi from our 5th, 46 min run. With the reactor again at 350 kW, the blocks received a fluence of fast neutrons of $1.9{\cdot}10^{13}/\mathrm{cm}^2$ (1 MeV equivalent) based on measured sulfur activity. Clearly, the Hitachi blocks are doped differently using Pr^{141} as well as Dy with 7 stable isotopes versus Tb^{159} for Shin-Etsu. The differences are compounded by Shin-Etsu's partial use of Co in place of Fe as well as their doping with Tb^{159} against Hitachi's use of both Pr^{141} and Dy in comparable amounts.

Table 3: Radioactive species by count rate (MNRC Run 6)

Element $_Z X^A$	Decay Prob.[a]	Energy [keV]	Block Type[b]			
			N34Z	N50M	HS36	HS46
$_{65}\mathrm{Tb}^{160}$	0.27	298.6	55.1	29.7	-	-
$_{65}\mathrm{Tb}^{160}$	0.17	879.3	32.9	18.6	-	-
$_{65}\mathrm{Tb}^{160}$	0.12	966.1	22.3	12.4	-	-
$_{65}\mathrm{Tb}^{160}$	0.13	1177.9	14.0	7.9	-	-
$_{27}\mathrm{Co}^{60}$	1.00	1173.2	9.6	7.2	-	-
$_{27}\mathrm{Co}^{60}$	1.00	1332.4	8.7	6.8	-	-
$_{61}\mathrm{Pm}^{151}$	0.23	340.1	26.2	50.2	34.4	50.9
$_{61}\mathrm{Pm}^{151}$	0.09	167.8	11.2	20.6	15.0	22.5
$_{61}\mathrm{Pm}^{151}$	0.07	275.3	9.0	16.9	11.8	17.2
$_{25}\mathrm{Mn}^{54}$	1.00	834.8	0.7	0.7	12.9	26.7
$_{59}\mathrm{Pr}^{142}$	0.04	1575.6	0.1	0.4	12.9	18.5
$_{61}\mathrm{Pm}^{149}$	0.03	285.9	3.3	5.1	3.7	5.1
$_{66}\mathrm{Dy}^{165}$	0.15	94.8	-	-	2.8	0.4
$_{60}\mathrm{Nd}^{147}$	0.28	91.2	2.4	2.9	2.0	2.6
$_{60}\mathrm{Nd}^{147}$	0.14	531.0	2.1	3.1	1.9	2.6

[a]Taken from Ref. [9]. [b]N-series blocks from Shin-Etsu, HS from Hitachi.

Interpretation and Discussion

Essentially all observed lines were identified but many were not tabulated to save space e.g. Tb^{160} levels with higher rates than Co^{60}. Table 4 summarizes characteristics and radioactivities of the main isotopes. There was some confusion with the strong Tb^{160} decay to the Dy^{160} exciting the 298 keV levels since there are other potentially nearby lines at this energy all of which may feed other strong lines e.g. the 966 keV line. Similarly, detector efficiency explains the apparent discrepancy between the 966 and 1179 lines in Table 3. Neutron knockout (n,2n) on Nd^{148} also has a cross section comparable to capture leading to Nd^{147} while (n,p)-exchange reactions on Fe^{54}, Ni^{60} or trace contaminants from the rare earths are also seen. Pm^{151} results from $\mathrm{Nd}^{150}(\mathrm{n},\gamma)\mathrm{Nd}^{151}$ followed by β^- decay. N50Z in Table 3 has about 55% as much Tb as N34Z which improves strength but reduces RR. Based on known capture cross sections for Fe^{58} and Tb^{159} and their relative abundances there is a large substitution in N34Z that greatly improves its RR although HS36E is better whereas HS46A is the most susceptible.

Considering the amount of iron in these magnets it is especially interesting that so few lines appear. Only Mn^{54} appears from Fe^{54}(n,p) charge-exchange having a rather large cross section but a threshold of a few MeV. Even so it has a quit low radioactivity. Mn^{56} appears in a similar way from Fe^{56} but with an even lower intensity and 2 h lifetime. Also, one sees that Shin Etsu clearly made significant substitutions of Co for Fe by the relative intensities of Co and Mn in the different blocks. From Tables 2-4, NdFeB has advantages over SmCo from both the lifetimes, γ energies and relative intensities. Curiously, the Co substitution helped neither the strength nor the radiation resistance.

Table 4: Radioactivities and half-lives of species in Table 3

Element $_Z X^A$	Half Lives	Energy [keV]	Radioactivity[μCi][a]			
			N34Z	N50M	HS36	HS46
$_{65}\mathrm{Tb}^{160}$	72.3d	298.6	0.83	0.44	-	-
$_{27}\mathrm{Co}^{60}$	5.27y	1332.4	0.08	0.06	-	-
$_{61}\mathrm{Pm}^{151}$	28.4h	340.1	0.40	0.77	0.53	0.78
$_{25}\mathrm{Mn}^{54}$	313d	834.8	0.01	0.01	0.09	0.18
$_{59}\mathrm{Pr}^{142}$	19.2h	1575.6	0.01	0.01	0.20	0.29
$_{61}\mathrm{Pm}^{149}$	53.1h	285.9	0.32	0.50	0.36	0.50
$_{66}\mathrm{Dy}^{165}$	2.33h	94.8	-	-	0.19	0.03
$_{60}\mathrm{Nd}^{147}$	11.0d	91.2	0.02	0.02	0.02	0.02

[a]N-series blocks are from Shin-Etsu and HS-series are from Hitachi.

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