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Magnetic Materials Suitable for Fission Power Conversion in Space Missions

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Magnetic Materials Suitable for Fission Power Conversion in Space Missions

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Abstract

Terrestrial fission reactors use combinations of shielding and distance to protect power conversion components from elevated temperature and radiation. Space mission systems are necessarily compact and must minimize shielding and distance to enhance system level efficiencies. Technology development efforts to support fission power generation scenarios for future space missions include studying the radiation tolerance of component materials. The fundamental principles of material magnetism are reviewed and used to interpret existing material radiation effects data for expected fission power conversion components for target space missions. Suitable materials for the Fission Power System (FPS) Project are available and guidelines are presented for bounding the elevated temperature/radiation tolerance envelope for candidate magnetic materials.

Introduction

The Fission Power Systems (FPS) Project has been developing the technological foundations for power generation on the surface of the Moon, an asteroid, or Mars (Ref. 1). These technology development tasks include an investigation of the radiation tolerance of the power system components. NASA guidelines for establishing radiation tolerance with respect to a particular mission specify first to define the environment, then to review the anticipated radiation behavior, and finally to determine that the subsystems will perform as required in the predicted environment (Ref. 2). The Fission Surface Power System Initial Concept Definition document (Ref. 1) provided the baseline mission and environment definition. Other reports have identified candidate materials-of-construction that may be sensitive to the radiation environment resulting from a fast-spectrum fission reactor (Refs. 3 and 4) and reviewed experimental studies relevant to the mission specific radiation tolerance of polymeric components (Ref. 5). The objective of this summary is to provide a more complete review of the magnetic materials that may degrade in this particular space-mission-relevant radiation environment.

The goal of this paper is both to review the relevant radiation literature and also to provide sufficient background to explain how the conclusions can be applied to different mission scenarios. Since the existing radiation-effects experimental data was generated for temperature and/or radiation conditions not necessarily the same as the current mission scenario, this report includes a review of basic phenomena and guidelines for extending the terrestrial focused database to space-mission relevant conditions. The FPS components and component materials have been described previously (Refs. 1 and 3). To summarize, a linear alternator is envisioned which will generate electricity from the relative motion of hard magnets with high coercive force strength/high remanent magnetic flux density, such as NdFeB class or SmCo class, and soft magnetic windings made from FeCo class alloys (Ref. 6). Specific property requirements depend on the details of the alternator design, but the radiation response of NdFeB, SmCo, and FeCo magnetic alloys are important factors to consider when designing a space power system designed for operating extended times in a radiation environment.

Basis of Magnetism

The foundations of material magnetism are based on the electronic behavior of the atoms. Each electron in an atom has a unique quantum number, which characterizes the electron shell (K, L, M, N, etc.), the subshell (either s, p, d, or f), the energy state, and the spin moment (either $+\frac{1}{2}$ or $-\frac{1}{2}$) (Ref. 7). The magnetic response of a material is fundamentally related to these electron positions and especially the spin behavior. All materials respond to applied magnetic fields, but the degree and the usefulness of the magnetic response vary greatly. Atoms that have full electron shells and subshells have only a weak, diamagnetic response to an applied magnetic field. This negative response is due to the applied field's influence on the angular momentum of orbiting electrons (Ref. 8). From a practical standpoint, these materials are often considered non-magnetic. Atoms without completely filled shells/subshells have a permanent magnetic moment based on incomplete cancellation of the electron spin and can be either paramagnetic or ferromagnetic. The atomic-level magnetic moments for paramagnetic materials are random and these materials have no macroscopic magnetic strength except in the presence of an external field. Ferromagnetic materials, however, may possess a permanent magnetic moment in the absence of an external field due to the physical alignment of electron-spin moments in adjacent atoms. The adjacent atoms with aligned spin moments are called atomic dipoles and are the fundamental building block of a magnetic domain. Observations have shown a clear trend between valence electron structure and magnetic moment strength that results from a compromise between Coulomb attraction and kinetic energy (Ref. 8). Narrow d-shell bands have been found to favor magnetism and are prevalent in select transition metals and rare earth metals.

The ferromagnetic materials that are useful in motor or generator power systems are those classified as both soft and hard. In this sense, soft and hard magnetic properties are analogous to ductile and strong in a mechanical system. Soft magnetic materials have magnetic domains that readily align to a magnetic field with minimal wasted energy. Conversely, hard magnetic materials resist change in their magnetic field alignment and can behave as a magnetic spring pushing back on an applied field. The term “permanent magnet” is sometimes used interchangeably with hard magnetic material, although no magnetic properties are permanent in a strict sense. Sufficiently high thermal energies, external magnetic fields, or combinations thereof will lead to magnetic realignment and, hence, loss of “permanence”.

The following terms are useful for describing magnetic material properties. The magnetization, M , is defined either as the net magnetic moment per unit volume in the material or as the contribution of the intrinsic material magnetization to the total magnetic field. The maximum possible magnetic saturation for any material is defined by its atomic chemistry although the measured magnetic saturation, M_s , for a material changes as a function of temperature. The Curie temperature, T_c , represent the temperature at which the magnetic saturation drops abruptly to zero.

The magnetic field strength, H , has the same units as M but represents the strength of the external magnetic field. The magnetic induction, \mathbf{B} , or the magnetic flux density, takes into account both the external field and the material contributions. Induction strength is defined as $B = \mu_0 H + \mu_0 M$ where μ_0 is the permeability in a vacuum. In general, permeability, μ , and susceptibility, χ , are related ways to describe the material magnetic response and $\mu = 1 + \chi$. There are slightly different unit conventions associated with the SI (tesla) or cgs (gauss) unit systems so care must be taken when comparing equations in the literature.

Figure 1 illustrates the response of a ferromagnetic material starting in a demagnetized state ($\mathbf{B} = \mathbf{H} = 0$). The application of a positive external field, \mathbf{H} , leads to an increase in measured magnetic field, \mathbf{B} . A similar plot can be drawn for the material magnetic field response, \mathbf{M} , versus external field. As the external field strength increases there is a realignment of magnetic domains until saturation is reached, B_s or M_s . When the applied field is reversed, the magnetic domains once again rotate along easy directions first and can produce a response curve with a hysteresis. The initial slope of the curve is the initial permeability, μ_i . The residual induction, B_r , or remanence, M_r , represent the remaining magnetic field strength when there is no applied field. The coercivity, H_c , is the reverse (or negative) field strength that

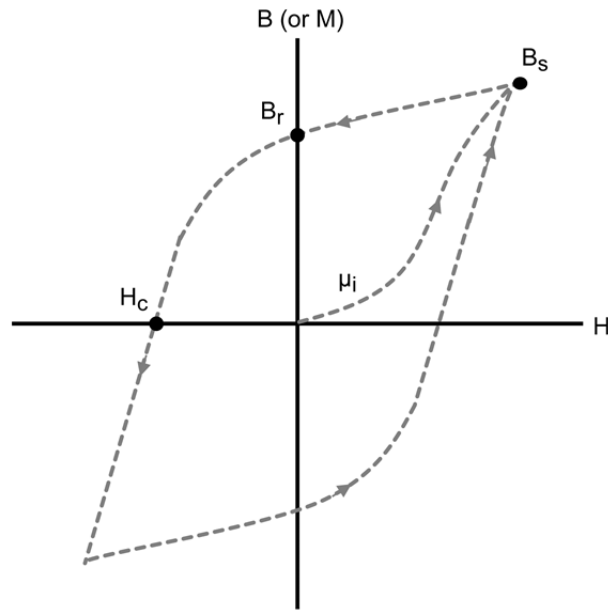


Figure 1.—Illustration of magnetic field strength, H , plotted versus magnetic induction strength, B . Points on the curve are labeled for B_r , which is the residual induction and H_c , which is coercivity or coercive field. Initial permeability, μ_i , is the slope B/H when $H \approx 0$. Soft magnetic materials should have a large permeability and low coercivity to minimize the hysteresis loop and hence energy loss on each cycle. High coercivity, remanence, saturation, and low permeability are desirable for hard magnets.

must be applied to return B to zero. Similarly the intrinsic coercive field, H_c , is the applied field needed to return M to zero for a H versus M relation. The shape of this B - H curve and the values of B_s , B_r , H_c , and μ_i describe the engineering properties of the magnetic material.

As the magnetic domain (region of aligned dipoles) increases, the magnetic strength of the material grows. There is a maximum possible magnetic strength, or magnetic saturation, of each ferromagnetic material based on its atomic structure. Therefore, chemistry changes either can raise or lower the magnetic saturation of a given metal alloy depending on how the minor element(s) atoms incorporate into the host lattice. The magnetic saturation is also a function of temperature because the increasing thermal energy causes atomic vibration which opposes the magnetic alignment forces. The Curie temperature is the temperature at which the long-range ordering of dipole spin alignment no longer occurs in ferromagnetic materials. Although the measurable magnetic strength disappears at the Curie temperature, it is interesting to note that neutron scattering experiments show that some very localized magnetic moments still exist (Ref. 8).

The structure of the magnetic material at the atomic level, also known as the crystallographic structure, influences many of the magnetic parameters. Magnetic domains are bounded by domain walls that are similar to crystallographic grain boundaries in that they move more freely as temperature rises and their motion can be limited, or pinned, by microstructural features. Limiting grain boundary size is one common way to control domain size. Magnetic permeability or susceptibility (the proportional constant between M & H) and coercivity (remnant field at $B = 0$) are affected by microstructure as well as chemistry and temperature. The influence of the crystal structure on magnetic reorientation has to do both the metallic crystal symmetry and the atomic spin orbital shape. Domain wall movement, and therefore magnetic field reorientation, is easier along some crystallographic directions than others. For example, the

easy direction, or easy axis, for body center cubic iron is along the [100]-type direction, which is the cube edges. The magnetic anisotropy constants, K_i , represent the difference in energy density required for magnetic saturation along an easy direction and a hard direction in the same alloy. $K_1 \sim 0$ is best for soft magnets (e.g., ordered FeCo) and negative K_1 implies a easy plane (family of directions), not just one easy direction.

Microstructural pinning features that restrict domain wall movement typically are desirable for “hard” magnets and undesirable for “soft” magnets. These microstructural features include grain size, the presence of secondary phases or precipitates, and the presence of cold work. Crystallographic texture that results from non-random grain orientations can either improve or impede magnetic response depending on the orientation of the texture with respect to the magnetic field lines. Induced electrical currents also lead to energy loss in soft magnetic materials, so higher electrical resistivity is a desirable material property for soft magnets.

Soft Magnetic Materials

When domain wall movement and domain magnetic orientation occur in weak applied fields, $H_c \leq 1000$ Amp/m, then the material is considered a soft magnet (Ref. 8). In addition to low coercivity, high initial permeability is desired to have a tight B-H hysteresis loop. Soft magnetic material performance is improved by alignment of crystallographic directions which allow easy movement of magnetic domains with the anticipated applied field directions. Soft magnetic material performance is improved by properties that increase remanence and lessen magnetic anisotropy. Anisotropy in the crystal lattice is important because the slope near saturation is dominated by the last remnants of alignment and because increasing anisotropy impedes domain wall movement. Impurities, especially in the form of precipitates, and residual stress can also pin domain walls and decrease soft magnetic performance. Proper alloying benefits magnetic properties by decreasing resistivity or improving other properties such as grain size. If grain size is too large, there are too few domain walls and micro-eddy current losses become high. Grain sizes that are too small restrict domain movement.

Soft magnetic alloys based on equiatomic FeCo alloys have a very high saturation strength, ~ 2.4 T, and relatively low magnetic anisotropy. This class is the preferred soft magnetic material for high flux density, high temperature applications and Hiperco 50 is a commercially available alloy in this class. Proper heat treatment produces a B2-type long range order which gives Hiperco 50 a high yield strength even at elevated temperature as well as high magnetocrystalline anisotropy (Refs. 9 and 10). Unfortunately the ordered microstructure does have low ductility. Because this composition goes through an order-disorder phase transformation around 730°C , the magnetic and mechanical properties are highly sensitive to manufacturing processes. Additions of up to 2 percent V and 4 percent Ni can improve fabricability and mechanical properties with minimal negative impact on magnetic properties. For sheet products, this alloy class is hot rolled above 912°C , quenched for cold rolling without long-range order, and then heat treated to regain long range order. Rolling into thin sheets for laminations can produce crystallographic textures that impede alignment of the easy magnetization directions with applied fields. Annealing and intermediate cooling rates after annealing are important to minimize adverse crystallographic texture and optimize magnetic properties (Ref. 8).

Hard Magnetic Materials

Hard or “permanent” magnetic materials are those which retain a relatively strong magnetic alignment and resist realignment from a reverse applied field as the result of a large residual induction, B_r , and coercivity, H_c . Common permanent magnets have coercivities in the range of 10^4 to 10^6 Amp/m (Ref. 8). It should be noted that for soft magnetic materials, the B-H curve and M-H curve are functionally equivalent, but that there is a distinction in hard magnetic materials. The intrinsic coercivity derived from the M-H curve is the shape independent description of coercivity. The B-H knee, or $(B-H)_{\text{max}}$ in the

second quadrant (upper left in Fig. 1) is a common descriptor to characterize the strength or energy density of hard magnetic materials. Magnetic permanence is enhanced by and magnetic reversal suppressed by:

1. Using chemistry or microstructure to increase magnetic anisotropy,
2. Maximize remanence through preferred grain texture,
3. Pinning domain walls or making single domain particles or grains,
4. Minimizing opportunities for domain exchange between particles or grains, such as non-magnetic grain boundaries.

Magnetic coupling of transition metal elements and lanthanide series (rare earth) elements vary in a systematic and predictable manner based on standard quantum physics (Ref. 8). There are several useful alloys of the RT_5 and R_2T_{17} series, where R = rare earth and T = transition metal. The phase boundary separation in the Sm-Co phase diagram is such that most SmCo alloys are in fact a combination of both $SmCo_5$ and Sm_2Co_{17} and will be generically referred to here as SmCo except when it is necessary to distinguish the phases. Based on a hexagonal structure (rather than a cubic structure), SmCo has a large magnetic anisotropy, but domain walls move readily unless pinned. However, limited substitution of Fe for Co and microstructure refining additions of Cu & Zr have led to SmCo alloys with B-H energy products up to 24 to 30 MOe. In summary, the simple alloys have coercivity that is grounded in anisotropy and nucleation of reverse domains is the limiting step. The more complex, higher energy-product SmCo alloys rely on complex microstructure for grain refinement, domain pinning and thus are tailored based on both chemistry and heat treatment (Ref. 11).

Alloys based on $Nd_2Fe_{14}B_1$ have a high anisotropy based on a tetragonal crystal phase, a large magnetism due to the Rare Earth-Transition Metal coupling of Nd-Fe, and the ability to grow non-magnetic B-rich and Nd-rich phases to decouple the magnetic grains. Very small grains and preferred grain orientation are required for optimal magnetic properties. Magnetic reversal appears to be dominated by nucleation and growth of reverse domains at temperatures up to 175 °C. Additional refinement options include partial substitution of Co for Fe to increase T_c and substitution of Tb for Nd to stabilize magnetic anisotropy to higher temperatures.

The Curie temperature is higher for the Co-based rare earth magnets than the Fe-based magnets. T_c also increases with increasing Co content and decreases with increasing Fe content. Apparently an antiferromagnetic exchange dominates Fe-Fe bonds at smaller bonding separation which also explains why increasing pressure can also depress T_c in Fe-based alloys (Ref. 8).

Radiation Damage to Magnetic Properties

The Fission Surface Power System Initial Concept Definition document provided a baseline radiation environment description for system. Shielding was proposed that would limit dose at the power conversion components to 5 Mrad gamma radiation and 2×10^{14} n/cm² high-energy neutron fluence (Ref. 1). The nominal magnetic materials operating temperature, 150 °C, can also be obtained from this system description. Therefore this review can focus on radiation damage at specific fluence and temperature ranges. The magnetic materials relevant to this program are metallic alloys with secondary phases that can be intermetallic or ceramic. Radiation damage processes and effects on metal alloys are covered in great detail by Was (Ref. 12) and are summarized here. Gamma radiation is a type of ionizing radiation because the gamma photons have insufficient energy to affect an atom nucleus but can displace electrons and thus ionize atoms. Typically, ionizing radiation has minimal effect on alloys since the electrons redistribute readily in the metallically bonded crystal structure. Conversely, neutron radiation has sufficient incident energy to damage metallic materials.

When a high-energy neutron strikes a material, a large energy transfer can take place. The struck atom is referred to as the primary knock-on atom (pka) and its energy is distributed to other nearby atoms by a series of secondary collisions in what is called a displacement cascade. These collisions produce

empty lattice sites (vacancies) and atoms injected into the interstices between lattice sites (interstitials). Fortunately, most vacancies and interstitials recombine; those that do not are responsible for radiation-induced microstructural and mechanical property changes. The continuous production and subsequent diffusion of point defects lead gradually to the formation of point-defect clusters and other extended defects. The evolution of such extended defects in turn is responsible for the physical and mechanical property changes of the material. Through this basic damage process, incident radiation changes defect diffusion rates, dislocation microstructures, void/bubble formation, and elemental segregation resulting in localized chemistry changes and changes in phase stability (Ref. 12). Note that this is a stochastic process and radiation can pass through a material without striking any atoms at all.

Radiation Studies With Soft Magnetic Materials

In the early 1960s, Gordon and Sery studied the effect of radiation on numerous soft magnetic materials. Figure 2 shows their results for in-pile magnetic property measurements on a range of soft magnetic materials up to 2×10^{18} n/cm² (fast fluence $\sim 10^{17}$ n/cm²) and sample temperatures of ~ 80 °C (Ref. 13). Alloy “2V Permendur” in these plots is chemically similar to the modern Hiperco 50. The iron-nickel alloys optimized for very high initial permeability appeared to be most sensitive to irradiation damage. The iron-cobalt alloy appeared to be unaffected. Gordon and Sery continued to study the iron-nickel alloys in various radiation fields and found that combinations of radiation and applied magnetic field could affect the magnetic domain alignment in much the same way as the combination of elevated temperature and magnetic fields (Refs. 14 and 15). These studies suggest that radiation can influence microstructural-controlled magnetic properties analogously to thermal treatments even when the bulk temperature remains low. Research specific to modern Hiperco 50 has shown that operating above 450 °C induces magnetic property degradations resulting from long-range order instability (Ref. 16).

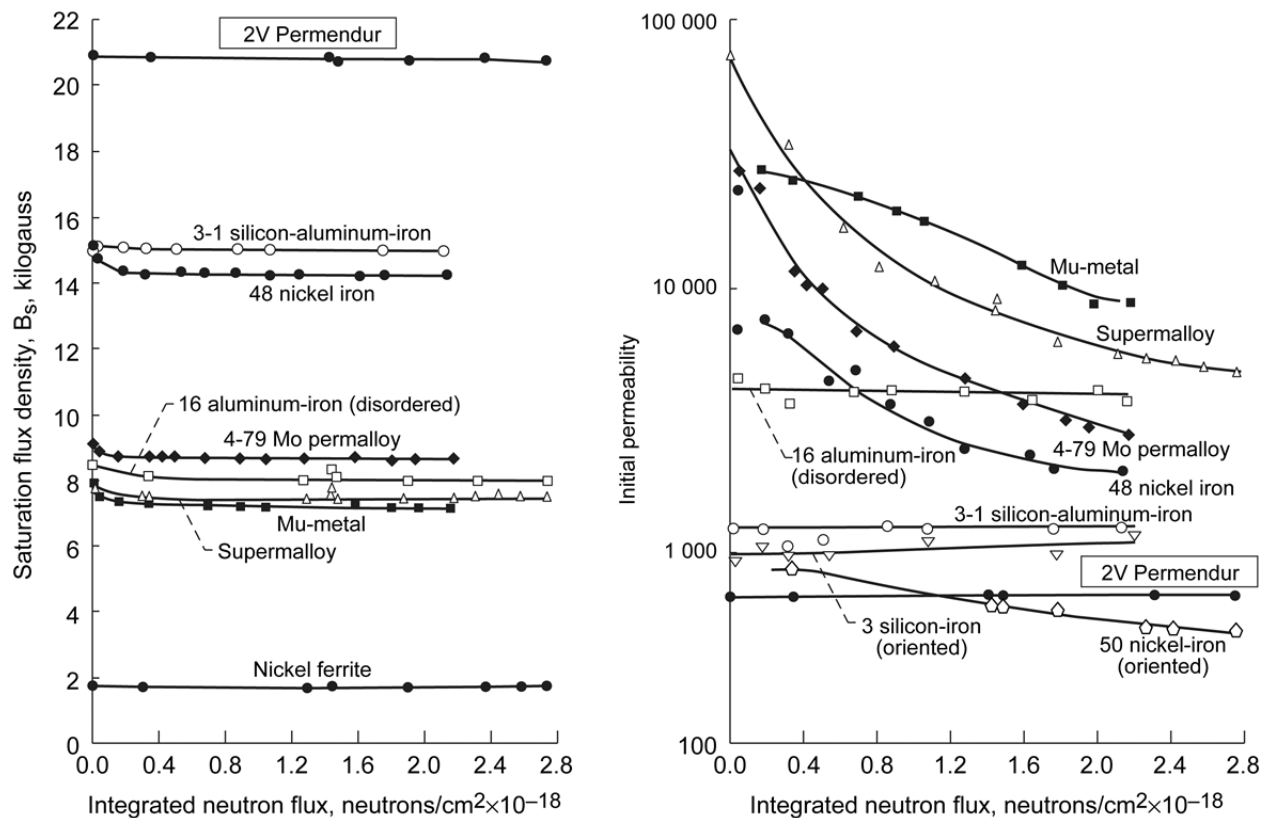


Figure 2.—Magnetic parameters of soft magnetic materials measured during neutron irradiation (Ref. 13). 2V Permendur, highlighted with boxes around name, is chemically similar to modern Hiperco 50.

In his chapter on phase stability, Was (Ref. 12) reviewed irradiation induced order-disorder reactions for several alloys and suggest that the influence of radiation on the ordering parameter was dependent on more factors than just fluence or dose rate. A combination of elevated temperature and radiation environment would likely have synergistic impact on the phase stability in Hiperco 50 and magnetic property degradation seen reported by Lin (Ref. 16) may occur at lower bulk temperatures under irradiation.

Radiation Studies With Hard Magnetic Materials

Early radiation studies on SmCo magnets were performed using proton beam and proton induced neutron spallation radiation. Brown et al. found negligible deterioration in residual induction in SmCo after exposure to fluences up to 10^{18} n/cm² for neutrons with $E > 0.1$ MeV (Ref. 17). Yet nearly concurrent studies in France subjecting similar SmCo alloys to lower fluences ($\sim 10^{16}$ /cm² or 10^9 rad) of 400 GeV high energy protons resulted in nearly complete demagnetization (Ref. 18). Blackmore performed similar studies using fluences from 10^6 to 10^{10} rad of lower energy, 500 MeV, protons and distinguished the higher stability of the Sm₂Co₁₇ alloys compared to the earlier SmCo₅ alloys (Ref. 19). These results, shown in Figure 3, confirmed that radiation energy level is a key factor and that both bulk chemistry (SmCo₅ versus Sm₂Co₁₇) as well as specific chemistry/processing (Crucore 18 versus other SmCo₅ magnets) are important.

Cost et al. studied the effects of neutron irradiation on Nd-Fe-B magnetic properties (Ref. 20). NdFeB permanent magnets from two different manufacturers were irradiated in the Omega West Reactor at Los Alamos National Laboratory with fast neutrons, $E > 5$ eV, at temperatures of 77 and 153 °C (350 and 426 K) to fluences up to 6×10^{16} n/cm². The Curie temperature for both magnets was stated as 312 °C (585 K), therefore the exposure temperatures were nominally 60 and 73 percent of the Curie temperature. At intervals during the irradiation the samples were removed from the reactor and remanence measured at room temperature. Cadmium shielding around the port was used to minimize the thermal induced (n, α)

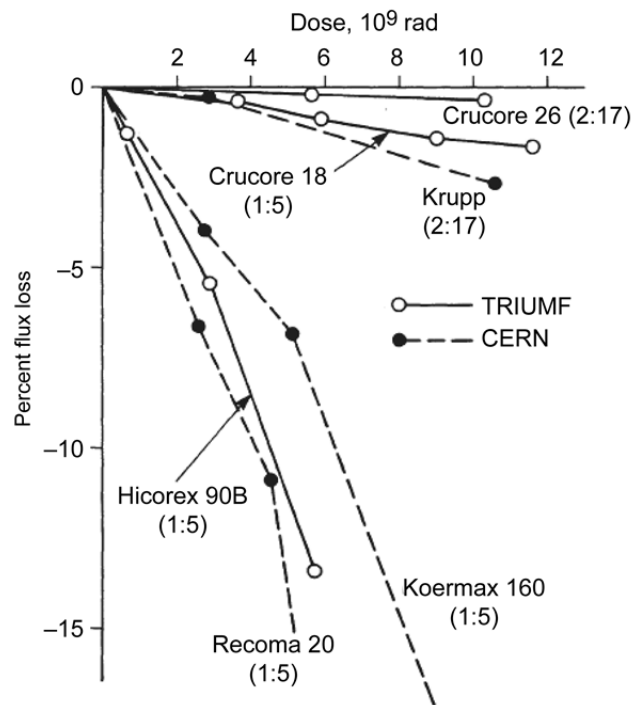


Figure 3.—Flux loss in SmCo₅ and Sm₂Co₁₇ class magnets as a function of high energy proton fluence, from Reference 19.

reaction with the ^{10}B isotope. The authors believed this damage would be different than that caused by higher energy neutron irradiation alone. The authors stated that the initial loss of remanence for irradiation to 10^{15} n/cm² was 10 percent at the higher temperature (~73 percent T_C) and roughly 5 percent at the lower temperature (~60 percent T_C). The loss rates were nearly the same for samples obtained from two different manufactures. The authors concluded that these losses are due to irradiation since the remanence did not decay with thermal exposure alone at 153 °C. Cause for greater decay at the higher temperature was speculated to be that a collision cascade has a higher probability of nucleating a reverse domain when the exposure temperature is closer to the Curie temperature. Also there was an increase in coercivity after radiation and re-magnetization, presumably due to agitation and relaxation of domain wall boundaries. This research demonstrated that there was a decrement in hard magnet permanence due to fast neutron exposure.

Brown and Cost extended this research by focusing on fast neutron, $E > 5$ MeV, flux of 4×10^{12} n/cm²/s and a temperature of 77 °C for several NdFeB alloys with various length-to-diameter (L/D) ratios and grain sizes (Ref. 21). It is well established that the aspect ratio of a magnetic material coupon affects the magnetic field shape and influences magnetization/demagnetization processes (Ref. 8). As expected, the rate of remanence decay in this research decreased monotonically with increasing L/D. The authors tried various comparisons to find correlation between material characteristics and decreasing remanence. They could not correlate the grain size and radiation-induced decay; however, the range of grain sizes investigated was not large. They found that remanence decay rate did decrease with increasing intrinsic coercivity. The linear correlation coefficient (R) between H_{ci} and remanence decay rate was 0.78. A slightly better correlation was found between the decay and knee field ($r = 0.83$). The authors specifically point out that these correlations are at best rough guidelines due to the scatter in the results. These results confirm that irradiation induced degradation is sensitive to chemistry, processing, and the magnetic field shape/strength during irradiation. This highlights the need to understand the experimental specifics when attempting to compare results from various studies.

Luna et al. exposed commercially available $\text{Sm}_2\text{Co}_{17}$ and $\text{Nd}_2\text{Fe}_{14}\text{B}$ magnets and an experimental PrFeB alloy to gamma, electron beam, and mixed beam (electron/photon) radiation fields (Ref. 22). Exposure of $\text{Sm}_2\text{Co}_{17}$ and $\text{Nd}_2\text{Fe}_{14}\text{B}$ magnets to a ^{60}Co gamma source resulted in no measurable remanence loss. Direct exposure of $\text{Sm}_2\text{Co}_{17}$ and $\text{Nd}_2\text{Fe}_{14}\text{B}$ magnets to an 82 MeV electron beam to a cumulative dose of 36 krad resulted in no measurable remanence loss in the SmCo but a 1.5 percent loss in the NdFeB. Samples of SmCo, NdFeB, and PrFeB were also exposed to a mixed electron/photon radiation field produced by the electron beam striking a tungsten alloy target. Results were slightly confused by the loss of water cooling during part of the experiments, but the SmCo was virtually unchanged even without active cooling and yet there was remanence loss in the NdFeB samples even when actively cooled. The PrFeB suffered the largest remanence loss as a result of irradiation. This paper also presents an often-referenced summary table which ranked alloy radiation hardness as $\text{Sm}_2\text{Co}_{17}$, SmCo_5 , followed by $\text{Nd}_2\text{Fe}_{14}\text{B}$ as the least radiation tolerant.

Zeller made some interesting comparisons between the character of demagnetization induced by elevated temperature and by irradiation (Ref. 23). He illustrated the field shape effects discussed previously by measuring magnetic field as a probe traverse a specimen normal to the magnetization direction. Elevated temperature exposures up to 54 °C produces more severe demagnetization in the center of the specimen than near the edges. This same shape relation was noted after exposure to ^{60}Co gamma radiation and to a more notable extent after irradiation with 106 MeV ^2H charged particles. He noted that the presence of light elements such as boron should increase the susceptibility of the magnets to radiation-induced thermal spikes since the recoil energy is so much higher for light elements than typical metals. The author did not note any temperature measurements made during irradiation exposure and thus the possible effects of temperature and radiation damage cannot be separated.

Finnish researchers published radiation studies in the early 1990s using 20 MeV protons (Refs. 24 to 26) and delved further into the mechanisms of irradiation induced demagnetization. Talvitie et al. compared Nd-Fe-B to Nd-Fe-B doped with Nb and Dy, irradiated from -258 °C to room temperature, and with magnetization along either the short or long sample dimension, followed by positron defect

measurements (Ref. 24). Their observations, including a 1000 times increase in sensitivity between $-258\text{ }^{\circ}\text{C}$ and room temperature as well as insensitivity of hysteresis curves, suggested that the magnetic losses were temperature driven and not related to defect formation. In a companion paper, they expanded their results with cube shaped specimens and introduced a simple theoretical frame work for modeling the irradiation induced changes (Ref. 25). The model suggests that the energy of the impinging radiation particle is transferred to the lattice in a spherical region surrounding the primary knock-on atom. The temperature then locally rises above the Curie temperature and the presence of a demagnetizing field can nucleate a reverse magnetic domain. They calculated the critical radius needed for reverse magnetic domain nucleation as a function of impacted atom type, initial temperature, grain size (actually domain-limiting microstructural feature size), and magnitude of the reverse magnetic field. In 1994 Kähkönen et al. published additional results that included magnetic packages rather than individual magnets, varied the impinging proton energy from 14 to 20 MeV, and studied the effects of alpha particle radiation at numerous temperatures from 22 to 295 K (Ref. 26). Their theoretical work suggests that the right combination of low demagnetization field and low global specimen temperature will preclude irradiation-induced demagnetization. They calculated that there would be no irradiation induced demagnetization in the fine grained, sintered $\text{Nd}_2\text{Fe}_{14}\text{B}$ magnets they studied for temperatures less than 50 percent of the T_c when magnetization is perpendicular to the short dimension of thin rectangular magnets, $B/\mu_0 H = -10$. They reported that the irradiation induced demagnetization resulting from alpha particle radiation was also temperature dependent, although correlation rate was different and was attributed to a stronger energy transfer from the alpha particles to the lattice compared to the lighter proton particles.

In the early 2000s researchers from Argonne National Laboratory (Refs. 27 and 28) and Fermi National Accelerator Laboratory (Refs. 29 to 31) published numerous studies addressing radiation tolerance of permanent magnets. Alderman and Job irradiated NdFeB magnets with x-rays, gamma-rays, fast neutrons up to $1.61 \times 10^{14}\text{ n/cm}^2$, and with thermal neutrons up to $3.34 \times 10^{12}\text{ n/cm}^2$ (Refs. 27 and 28). Subsequent to irradiation, sample flux densities were measured. The fast neutron irradiation from 1 to 2 MeV ^{252}Ca exposure revealed residual induction measurements with no significant changes in flux densities after $1 \times 10^{12}\text{ n/cm}^2$ exposure, but there was a 0.6 and 10 percent flux density degradation respectively after the 2×10^{13} and $1.61 \times 10^{14}\text{ n/cm}^2$ steps. Thermal neutron irradiation was achieved with polyethylene-moderation of the ^{252}Ca and fluences of up to $3.34 \times 10^{12}\text{ n/cm}^2$ had no statically significant effect on residual induction. The authors claimed that the irradiations were performed at ambient (room) temperature, however it did not appear that there was active cooling or *in situ* temperature measurement.

Volk reviewed permanent magnet radiation studies, emphasized the theoretical framework presented by Kähkönen et al., and proposed research to test the Kähkönen theory (Ref. 29). He proposed radiation testing magnetic materials with the same grain size and know magnetization direction but with significantly different coercivity. He emphasized testing magnetic materials in component configuration so that the demagnetization fields would be representative. A subsequent paper provided additional background discussing why existing literature did not have sufficient detail to confirm or refute the Kähkönen theory and they proposed a model magnet configuration that was a variation on the offset-quadrupole magnet design. This multipole would consist of three dipoles and a variable gap (Ref. 30). The arrangement of the dipoles would allow control of the self-demagnetization fields. Several papers presented preliminary radiation results for individual magnet blocks (Refs. 31 and 32). Unfortunately the research required to confirm the basic understanding was not completed due to insufficient funding (Ref. 33).

Chen et al. studied the effect of neutron irradiation effect on $\text{Sm}_2\text{Co}_{17}$ and NdFeB magnets using a water-moderated reactor with a broad, heavily thermal, energy range. They showed that $\text{Sm}_2\text{Co}_{17}$ -type magnets had significantly higher resistance to neutron irradiation than Nd-Fe-B-type magnets (Ref. 34). Their results supported the Kähkönen thesis that radiation-induced thermal spike leading to an increase in localized temperature, T_L . For both $\text{Nd}_{13}\text{Dy}_2\text{Fe}_{77}\text{B}_8$ and SmCo magnets the temperature raised by thermal spike was dependent on the neutron flux, or reactor's power, rather than neutron fluence. The recorded temperature in the NdFeB sample was $266\text{ }^{\circ}\text{C}$ for the neutron flux of $2.1 \times 10^{13}\text{ n/cm}^2/\text{s}$. The NdFeB magnets lost all their magnetic strength, and authors concluded that the local temperature at samples

interior must exceed $T_c = 309\text{ }^\circ\text{C}$. The $\text{Sm}_2\text{Co}_{17}$ magnets, however, had imperceptible change in magnetic flux after up to $\sim 10^{20}\text{ n/cm}^2$.

Additionally, there are a number of papers that summarize either magnet degradation as the result of in situ operation or test individual magnets destined for a specific application (Refs. 35 to 38). As was observed by previous authors (Ref. 30), simply cataloging the existing data does not provide the basic understanding to provide fundamental guidance. Very few of the papers report factors such as grain size and specimen temperature during irradiation. Most of the data is reported on commercially available magnets which have been found to vary chemically and in physical characteristics. Thus the community still has only a general guidelines to predict radiation induced demagnetization.

The Effect of Gamma—Irradiation on the Magnetic Properties

Boockmann et al. studied the effect of γ -radiation from a ^{60}Co -source on the magnetic properties of sintered Rare earth- magnets. The magnetic properties of sintered SmCo_5 , $\text{Sm}_2(\text{Co,Cu,Fe,Zr})_{17}$ and $(\text{Nd,Dy})_{15}\text{Fe}_{77}\text{B}_8$ magnets were not affected by γ -radiation from a ^{60}Co -source up to accumulated dose of 50 Mrad (Ref. 39). In addition to studying neutron irradiation, Alderman et al. performed parallel studies on NdFeB using x-radiation and γ radiation (Ref. 27). Even with doses of 280 and 700 Mrad, respectively, the photon radiation had negligible impact on magnetic properties. Furthermore researchers study a synchrotron beam source detected no magnetic degradation due to x-ray exposure estimated to be 12 Mrad (Ref. 40). Therefore, permanent magnetic degradation appears minimal due to x- or γ -radiation.

The Effect of Electron—Irradiation on the Magnetic Properties

Bizen et al. (Refs. 41 and 42) investigated the magnetic field change of undulator magnets when exposed to a 2.0 GeV electron beam. They characterized magnetic field intensity for specimen magnetized both parallel and perpendicular to the aspect ratio as well as individual and stacked magnets. Magnetic intensity decreased monotonically with accumulation of electron dose; approximately 2.5 percent intensity loss for the accumulated electron dose of 40×10^{13} (Ref. 41). Subsequent experiments utilized vacuum and air thermal stabilization treatments prior to irradiation. Heat treatment at $142\text{ }^\circ\text{C}$ for up to 24 hr led to a loss of magnetic intensity of up to 0.69 percent, but greatly reduced the subsequent irradiation-induced changes of the $\text{Nd}_2\text{Fe}_{14}\text{B}$ magnets (Ref. 42). This result is similar to the increased stability seen with thermal treatments (Ref. 8) or controlled irradiation (Ref. 15).

Summary and Conclusions

The research for the radiation effect of permanent magnets was sparse before the 1980s, and primarily performed at the Department of Energy laboratories in order to evaluate the insertion devices for the synchrotron radiation facilities. In recent decades, numerous researchers investigated rare earth magnets exposed to neutrons, electron beams, protons, gamma-radiation, and X-radiation. Studies have shown that accumulation of neutron or high energy particle radiation can degrade magnetic properties. Numerous studies suggest that radiation damage affects both soft and hard magnetic properties in a manner that is analogous to and additive with thermal damage. The parallel or anti-parallel spin alignment of the orbiting electrons is sensitive to energy balance. Increases in kinetic energy in the material increases the mobility of magnetic domains and increases the likelihood of reverse domain nucleation, which promotes a realignment along external magnetic field lines. Increased kinetic energy can also alter the crystal structure of the magnetic material by standard processes such as grain growth, precipitation or solutioning. Alloys with highly engineered crystal structures can achieve the most desirable magnetic properties, but also have greater driving force to destabilize in response to increased thermal/radiological energy.

Currently no physics-based models or experimental data are available to fully predict the effect of thermal and radiation conditions of a FPS mission on baseline magnetic materials. Significantly more studies have addressed the radiation tolerance of hard magnetic materials and theoretical models have been proposed, but not verified. There is a general consensus in the literature that bulk temperature, radiation, and demagnetization fields all influence magnet radiation hardness. A guideline of staying below 50 percent of Curie temperature ($T \leq 300$ K when $T_c = 583$ K) for small demagnetization condition was suggested for minimal radiation-induced degradation in specific NdFeB-type permanent magnets (Ref. 26). Note that this theoretical estimate with very specific assumptions including the size of reverse magnetic domains and the strength of the demagnetizing field.

Although there is no direct experimental verification for the Kähkönen model, the trends in the literature support it. Figure 4 summarizes some examples of neutron irradiation experiments for rare earth permanent magnets. The measured bulk temperatures of three NdFeB experiments were 77, 153, and 267 °C, which represented approximately 60, 73, and 86 percent of the Curie temperature. There was measurable post-irradiation remanence degradation in each case. In similar SmCo material studies, magnet bulk temperatures did not exceed ~40 percent of their Curie temperature and experience no measurable degradation. Thus it is suggested that, in absence of targeted experiments to define precise temperature/radiation limits, the permanent magnets should not be used at a bulk temperature exceeding 50 percent of their Curie temperature in conjunction with neutron fluences that exceed 10^{16} n/cm². If the demagnetization field of the magnetic component is not low, then the bulk temperature should be lower than 50 percent of T_c . Modern Sm₂Co₁₇-type alloys have Curie temperatures on the order of 800 °C (1073 K) (Ref. 34). These alloys should be functional in the modest neutron environments envisioned for the FPS convertors at temperatures up to ~265 °C (535 K).

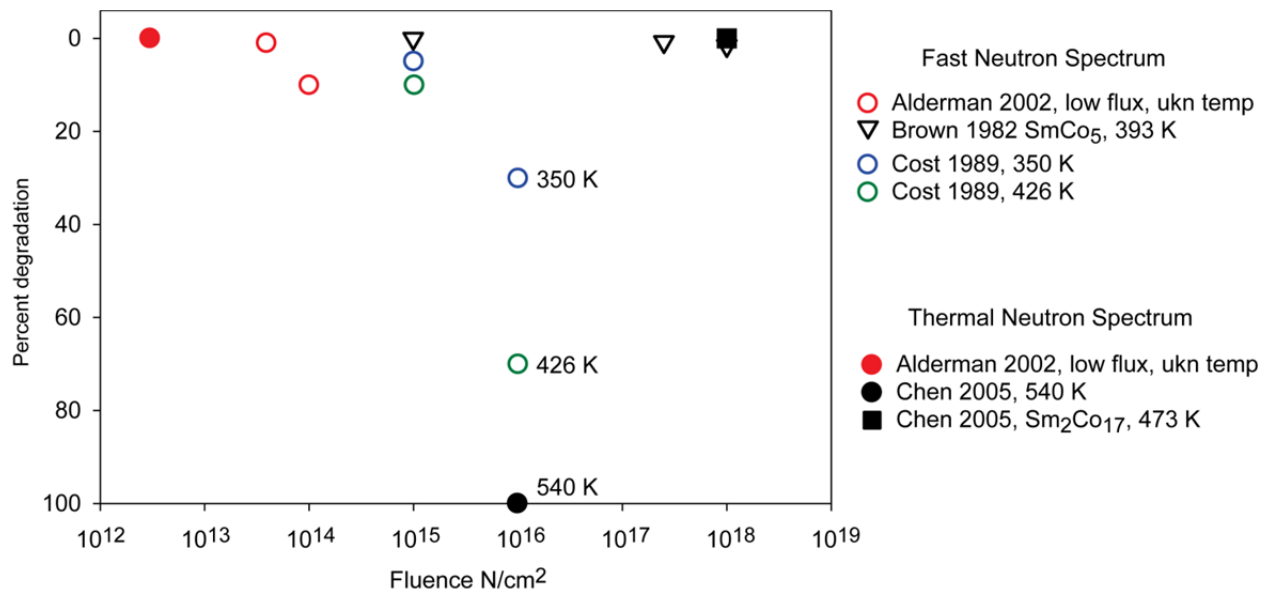


Figure 4.—Summary of remanence loss as a function of neutron fluence for rare earth permanent magnets. Circles are NdFeB alloys, up-side-down triangles are SmCo₅ alloy, and squares are Sm₂Co₁₇-based alloys. Open symbols represent primarily fast neutron exposure and filled symbols are from experiments with minimal fast neutrons.

Radiation damage in soft magnetic materials has not been studied as extensively as the damage in hard magnetic materials. The most extensive radiation review suggested that the more highly engineered alloys with the highest initial permeability were the most susceptible to radiation induced degradation (Ref. 13). However, the data was reported for fluences of 10^{18} n/cm² and higher, which is well above the approximately 10^{14} n/cm² mission fluence currently considered. A reasonable supposition would be that the degradation mechanisms affecting soft magnetic materials would be dominated by the energy transfer. The published Curie temperature for candidate alloy Hiperco 50 is 938 °C (1211 K). If the 50 percent T_c limit discussed relative to hard magnets could be applied, the combined conservative radiation-temperature limit would be ~327 °C. However Hiperco 50 experiences discontinuities in the B-H curve after very short times at 580 °C and induction loss after extended times at 450 °C (Ref. 16). In a review of phase transformations in radiation environments, the influence of thermal spike distribution on order-disorder transformation was described. The fundamental mechanism appears to be the same as that driving reverse domain nucleation in the hard magnetic material, although the particular temperature/fluence relation need not be the same. In the absence of specific temperature/radiation interaction data, limiting Hiperco 50 application in a radiation environment to a bulk temperature of 160 °C (~50 percent of the general temperature limit) should be a conservative guideline to maintain optimum magnetic properties. Higher temperature applications are possible if the design can accommodate higher core losses resulting from decreased induction and increased coercivity. Studies on magnetic loss due to temperature effects alone can provide non-conservative guideline (Refs. 16 and 43) and additional performance losses can be anticipated due to radiation interactions for temperatures greater than 160 °C. The magnitude of the additional degradation could be estimated from modeling the radiation capture thermal distribution or could be measured experimentally.

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14. ABSTRACT Terrestrial fission reactors use combinations of shielding and distance to protect power conversion components from elevated temperature and radiation. Space mission systems are necessarily compact and must minimize shielding and distance to enhance system level efficiencies. Technology development efforts to support fission power generation scenarios for future space missions include studying the radiation tolerance of component materials. The fundamental principles of material magnetism are reviewed and used to interpret existing material radiation effects data for expected fission power conversion components for target space missions. Suitable materials for the Fission Power System (FPS) Project are available and guidelines are presented for bounding the elevated temperature/radiation tolerance envelope for candidate magnetic materials.					
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