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RADIATION DAMAGE THRESHOLDS FOR PERMANENT MAGNETS

UNITED STATES NAVAL ORDNANCE LABORATORY, WHITE OAK, MARYLAND

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RADIATION DAMAGE_THRESHOLDS
FOR PERMANENT MAGNETS

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ABSTRACT: Several sets of permanent magnets, representative of commercially important magnet materials, were irradiated at Brookhaven National Laboratory and Argonne National Laboratory to integrated neutron flux levels from $3 \times 10^{17}$ to $4 \times 10^{20}$ epicadmium n/cm$^2$. In spite of this relatively high dose, Alnicos II, V and XII showed negligible change in properties whether irradiated at 60°C, 235°C, or 325°C. Cunico I, though affected, showed changes less than a threshold of radiation damage of ±10%. Cunife I and 3-1/2 Chromium Steel showed slight improvements in properties. The Barium Ferrites, Silmanal, 36 Cobalt Steel and others exceeded the 10% damage threshold by various amounts which extended up to severe demagnetization. Differentiation between temperature and radiation effects was accomplished by the use of control magnets, and by the 600°C irradiation. Limitations on the use of Alnicos II, V, XII and Cunico I in combined heat and nuclear radiation environments may be imposed by the higher vulnerability of associated soft magnetic circuit components, e.g., pole pieces of soft iron, to radiation damage and by high gamma heating which can occur if a magnetic circuit must be used in a sealed container (for protection from corrosion or other reasons).

Of the two most widely used groups of permanent magnets, the Alnicos exhibit the highest resistance to radiation, while the barium ferrites show the least.

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The first irradiation experiment in this survey of the effects of nuclear radiation on permanent magnets showed that none of the materials tested were affected by the same amount of radiation which caused some soft magnetic materials to deteriorate. The initial permanent magnet results were presented in NAVORD Report 6276 and the work on soft magnetic materials in NAVORD Report 6127. This report gives the results for the behavior of the same materials at ~10, 100 and 1000 times the initial dose to which they were subjected. The work was performed as part of a broad program for developing magnetic materials (Task No. RRMA-02008/2121/R007-01-001).

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INTRODUCTION

1. What are the effects of nuclear radiation on permanent magnet performance? To answer this question, a series of experiments was performed to test a number of materials to predetermined values of integrated neutron flux or until the magnetic properties of most of the magnets showed major degradation. However, materials such as Alnicos II, V and XII showed little or no change in properties after high level irradiation and, except for adverse temperature effects, Cunife I and 3-1/2 Chromium Steel showed a slight tendency toward improvement. Eleven materials in all were subjected to irradiations up to $5 \times 10^{22}$ epicadmium nvt at maximum temperatures of 60, 235 and 325°C. Two materials showed changes of the order of a $\pm 10\%$ threshold of radiation damage, Cunife I and Cunico I. The two Barium Ferrites (oriented and unoriented) Platinum Cobalt, 36 Cobalt Steel and Silmanal showed severe property degradations.

2. In the first of these experiments the total integrated epicadmium flux ($n/cm^2$ or nvt) reached was only about $3 \times 10^{17}$. None of the thirteen materials irradiated were affected by this relatively low dose. This was not an unexpected result since previous work with soft magnetic materials revealed a rough rule of thumb measure that materials having coercive forces less than 0.5 oersted tended to be degraded by irradiation to this level, whereas those having coercive forces greater than 0.5 oersted were not affected. Permanent magnet coercive forces lie between 50 and 4500 oersteds (see Fig. 1). Subsequent irradiations of $\sim 10^{18}$, $10^{19}$ and $10^{20}$ nvt were performed until some of the magnets not only crossed the threshold of damage but were in some cases severely demagnetized.

3. The initial experiment with permanent magnets to $3 \times 10^{17}$ nvt was performed at the Brookhaven National Laboratory. (NOTE: Unless otherwise specified, all values of integrated flux are for epicadmium neutrons (neutrons whose energies $> 0.4$ ev)). All succeeding experiments were performed in the CP-5 reactor at Argonne National Laboratory (ANL). Since this work was of interest to the Reactor Engineering Division at Argonne, the experiment was conducted as a joint effort by the U. S. Naval Ordnance Laboratory and the Argonne National Laboratory. Projected applications by ANL included the use of permanent magnets as components of electromagnetic flowmeters.
immersed in the coolant of a reactor in high nuclear radiation fields for extended periods of time. ANL provided the facilities such as space in CP-5, the use of hot cells, temperature monitoring equipment, and dosimetry fabrication and evaluation, while NOL conducted the experiments on the permanent magnets.

EXPERIMENTAL

4. Because reactor space available is limited, the number of permanent magnet materials to be tested in this survey was restricted to a representative group of thirteen materials. Figure 1 lists these materials and shows their nominal demagnetization curves. Table I gives additional information. Two materials, the Elongated Single Domain (ESD) magnets, were eliminated after reaching the $10^{19}$ nvt level because one of them had completely disintegrated into a powder sometime before this value had been reached and had clung to the other test specimens. This disintegration was probably due to the melting of the lead alloy matrix under high temperature. The Platinum Cobalt magnet was eliminated from the normal temperature experiment due to lack of space.

5. In order to be able to differentiate between radiation induced changes and changes caused by high temperatures, duplicate sets of magnets were used as controls. Temperature levels and temperature drops (which occurred at reactor shutdowns) encountered by the in-pile test magnets were simulated for the control magnets in ovens. To further insure that the test and control assemblies had the same treatment, both sets of magnets for each experiment were, wherever practicable, stabilized, tested and handled in the same manner and at the same time. They were also packaged in identically designed containers. Most of the containers were approximately 12" long by 1.25" in diameter. These dimensions were dictated by the geometry of the CP-5 holes available and by the necessity for minimizing the length of container monitored by a single set of dosimeters.

6. The buildup of temperature by gamma heating to high values in the can type of container shown in Fig. 2A was prevented in the normal temperature experiment by the use of the tube type of assembly shown in Fig. 2B. (The gamma flux was estimated to be appreciably greater than $10^7$ roentgens per hour). The magnets and their aluminum spacers fitted closely enough in the tubes of this assembly so that adequate cooling was effected but not so closely that they could not be removed without forcing. A non-magnetic stainless steel spring in each tube provided positive contact of the magnets with the assembly both
at the ends of the tube and, to some extent, along the length. The magnets were aligned with the north poles in the same direction.

7. In-pile temperatures were recorded continuously on a strip chart. The sensing elements were chromel alumel thermocouples inserted in holes in dummy samples of type 304 (non-magnetic) stainless steel. A set of three dosimeters was included in each container and in the last three experiments (at $10^{26}$ nvt) two such sets were included. The three dosimeters per set included a cobalt aluminum foil or wire for monitoring the thermal flux, a second Co-Al wire in a cadmium jacket for the epicadmium flux, and an aluminum sulphate pellet for the neutrons having energies greater than 2.9 Mev.

8. No in-pile magnetic measurements were made because existing measurement techniques and presently available in-pile space are incompatible with such measurements. Because of the high induced activities in these magnets, all post-irradiation measurements were made in hot cells. All of the magnetic properties measured were made with specially designed search coils which allowed data to be taken by remote handling in the hot cell. Since pre- and post-irradiation measurements were made with the same or identical search coils, leakage flux errors as high as ± 2% were eliminated.

9. For most of the magnets only one property was measured, the open magnetic circuit induction (BOMC). BOMC is the operating induction value of the magnet under open circuit conditions. It is represented by a point on the demagnetization curve or within it. For a given material, it depends on the shape and dimensions of the magnet, as well as on the geometry and material of the entire magnetic circuit. Each magnet had that length to diameter (L/D) ratio which fixed its initial operating point at or above the knee of the demagnetization curve. This insured operation of the magnet at an optimum point for open circuit conditions. Experimental work at the $10^{17}$ and $10^{18}$ nvt levels included an additional set of magnets which was used for the closed magnetic circuit tests in which demagnetization curves were obtained.

RESULTS AND DISCUSSION

Irradiation to $5 \times 10^{26}$ nvt, Changes Less Than 10%

10. Three sets of magnets were irradiated to a total integrated flux of about $4 \times 10^{26}$ nvt, one at a normal temperature of 60°C ± 10°C, one at an intermediate temperature of 230°C ± 20°C, and one at a high temperature of 330°C ± 20°C. The results of
these three experiments are shown in Table II and Fig. 3. There are two main groupings of the results. The first group includes those magnets which were unaffected by this high value of integrated flux or at the least showed radiation damage of less than an arbitrarily chosen threshold change of \( \pm 10\% \). The second group consists of those materials in which changes greater than this 10\% threshold occurred.

11. Alnico II, Alnico V, Alnico XII and Cunico I belong to the first group. The first two showed negligible changes in properties to within an experimental error of \( \pm 2\% \). The Alnico XII magnet irradiated at a normal temperature showed a -6.5\% change which is still considerably below the 10\% threshold of damage. The three Cunico I samples approached the 10\% threshold value. For Cunico I comparison of the results (see Table II or Fig. 3) for each of the three test magnets with its control reveals that the change produced was due to irradiation. The average of the differences between percentage changes of the test and control magnets is about -7\% (Note the similarity in behavior of this material and 36 Cobalt Steel, particularly with reference to the analysis on 36 Cobalt Steel in the next section).

**Irradiation to \( 5 \times 10^{20} \text{nvt} \), Changes Greater Than 10\%**

12. The three irradiated 36 Cobalt Steel magnets showed changes of -37, -37.5 and -34.5\% in BOMC at normal, intermediate and high temperatures respectively. The changes in the three corresponding control magnets were -1, -20.5, and -22\%. An analysis of the demagnetizing effects of nuclear radiation and temperature on BOMC for this material is illustrated with the aid of Fig. 4.

13. The nominal operating point for each of the six magnets after magnetization and stabilization was point A. If we consider the high temperature control sample alone, heating to 325^\circ C may be considered equivalent to a fictitious demagnetizing field of magnitude \( -\Delta H_T \). This field causes a shift in the operating point from A down the curve to C. At the first simulated reactor shutdown, the drop in temperature, i.e., the removal of the equivalent demagnetizing field, \( -\Delta H_T \) will cause the point to move along the idealized minor loop CD until it intersects the load line AO at A'. With succeeding rises and drops in temperature the point commutes between C and A'. Its final position at the end of the temperature simulation period is A. The net change in BOMC caused by temperature alone is \( \Delta B_T \). For the control sample run at 235^\circ C the same analysis holds with the exception that \( \Delta B_T \) would be smaller.

14. For the irradiated sample at 325^\circ C the same temperature induced change, \( \Delta B_T \), occurs initially since the first reactor
shutdown takes place within one or two days after irradiation begins. However, the cumulative effect of sixteen weeks of neutron bombardment causes the operating point to move from A' through C to C'. The net change in BOMC ($\Delta B = 34.5\%$) can be considered as resulting from a fictitious demagnetizing field $-\Delta H_1$ which is the equivalent of the demagnetization influence of irradiation. This field $-\Delta H_1$ may be considered as being permanently applied since the effects of irradiation are cumulative whereas those of temperature are partly reversible, i.e., BOMC moves from C to A' once the equivalent field $-\Delta H_1$ is removed. It does not move up a similar minor loop parallel to CA' since the fictitious field cannot be removed upon cessation of irradiation except perhaps by remagnetization of the magnet in which case temperature effects could also be erased (provided that no irreversible metallurgical changes had occurred in either case). For the magnet irradiated at 235°C the same equivalent demagnetizing field $-\Delta H_1$ gives rise to approximately the same $\Delta B_1$ ($-37.5\%$).

15. $\Delta B$ in Fig. 4 merely indicates the difference between irradiation and temperature effects. That $\Delta B_1$ is actually the magnitude of the change produced by irradiation becomes evident when the results for the two magnets subjected to temperatures of only 60°C are examined (see Fig. 3). The control magnet showed a negligible change indicating no effects due to temperature. Its operating point remains at A. However, the change in the test magnet of $\Delta B = 37\%$ was due entirely to irradiation. This corresponds to the application of an effective, permanent field $-\Delta H_1$ which caused the operating point to move from A to C'. The radiation induced change in BOMC occurred independently of the presence of temperature. The fact that various demagnetizing influences can operate independently of each other makes it possible to stabilize a magnet against any changes which are smaller than the change caused by the stabilization process itself. This analysis indicates therefore that this material may be amenable to stabilization such that it would not be affected by irradiation to the nvt's achieved in spite of the large changes in BOMC which actually occurred.

16. An alternative method of illustrating the change in 36 Cobalt Steel is to consider the operating point BOMC as remaining always on the load line. Since the load line is a function only of the geometry of the magnet it does not change. Therefore, the demagnetization curve must change with temperature or irradiation. Its change in shape is unspecified. However, its intersection with the load line marks the position of BOMC and thus the net change $\Delta B_1$ or $\Delta B_T$. As before the changes due to temperature and irradiation are independent of each other; the larger of the two changes masks out the presence of the other.
17. The oriented and unoriented Barium Ferrite magnets are identical in composition but have marked differences in magnetic properties (Fig. 1). However, both were affected to the same extent by irradiation. For approximately the same integrated fluxes the changes in the magnets irradiated at normal temperatures were \(-63\%\) and \(-54.5\%\) respectively; but at intermediate and high temperatures the percentage changes were, for both materials, within a range of \(-23 \pm 2\%\). At the intermediate temperature (\(\approx 235^\circ C\)) the two ferrite controls showed no changes. This is to be expected since normally they can withstand temperatures up to \(450^\circ C\) without appreciable changes. The high temperature controls exhibited peculiar behavior, however. They were completely degraded by prolonged heating at the \(325^\circ C\) temperature. It should be pointed out that both the irradiated samples and their controls were not only subjected to the above temperatures but also to periodic drops in temperature which followed reactor shutdowns (for the controls quenching in air by abrupt removal from the oven simulated the reactor temperature drop). These thermal shocks may have caused the deterioration in the high temperature controls. No explanation has suggested itself for the constancy of the \(-23\%\) values (see Fig. 3), for the irradiated magnets at intermediate and high temperatures as compared to the lack of change in the controls at the intermediate temperature and the almost complete degradation for the controls at the high temperatures.

18. Although no Platinum Cobalt magnet was irradiated at a normal temperature there is some evidence of radiation damage in this material (Fig. 3). Based on the analysis for 36 Cobalt Steel the damage should be at least as great under normal temperature irradiation as that which occurred at the elevated temperatures. The control magnet at \(325^\circ C\) showed anomalous behavior similar to that of the Barium Ferrite controls at this temperature. Actually all eleven of these controls were checked after only two weeks of the simulation of the in-pile temperatures. All but one (3-1/2 Chromium Steel, \(-13.7\%\) change) showed negligible or slight effects, e.g., Platinum Cobalt \(-4.6\%\), Silmanal \(-3.2\%\), the ferrites \(-1\%\). However, at the end of the 16-week simulation of reactor temperatures, all but Cunico I and the three Alnico magnets showed major changes in BOMC (see columns 8, 9, Table II and Fig. 3). This was not unexpected for 3-1/2 Chromium Steel or Silmanal which are affected by temperatures above \(120^\circ C\) (Table I) and \(235^\circ C\), respectively. But materials such as the ferrites and Cunife are not appreciably affected at temperatures below \(450^\circ C\). The presence of thermal shocks was probably a factor in causing the degradation which occurred in some of the magnets.

19. Silmanal is normally annealed by a slow bake at about \(250^\circ C\). Therefore, the high temperature alone, which peaked at
325°C, caused an almost complete loss of magnetization in both the test and control magnets. However, at a more normal temperature of operation (∼60°C) there was a change in BOMC of -53% due almost wholly to radiation damage. The magnets at 235°C showed opposing trends. The test magnet was adversely affected by radiation; the control magnet was helped by the simulated reactor temperature. But, since none of the Silmanal magnets had been given an optimum heat treatment initially, this control magnet had been improved only because of a fortuitous heat treatment which occurred during the in-pile temperature simulation. The test magnet did not show improvement because of the large radiation damage effect.

Irradiation to 5 x 10²⁰ nvt, Possible Improvement in Properties

20. For high level irradiation at a normal temperature, 3-1/2 Chromium Steel showed a slight improvement in BOMC of 2.4%. Ordinarily a percentage change of this magnitude would not be significant. However, this material has the lowest coercive force (Fig. 1) of all the materials tested and proved to be the most easily affected by demagnetizing influences. The positive percentage change, which ran counter to the expected tendency to demagnetize, may have been even larger than that shown in Fig. 3. However, this magnet had become jammed in its close-fitting aluminum jacket and had to be subjected to the demagnetizing effect of being forcibly punched out before its BOMC value could be measured. The effect of intermediate temperature irradiation on chromium steel was about the same as that of temperature alone on the control magnet, and this was expected since this material is permanently affected by temperatures above 120°C. It is significant though, that the radiation induced change was smaller than the change produced by temperature alone (-68% as compared with -79%). The additional fact of a sizeable increase in BOMC at the high temperature irradiation corroborates this tendency for 3-1/2 Chromium Steel to improve with radiation (see Fig. 3). But the magnitude of the percentage increase is somewhat misleading. Because a preliminary dry run had indicated an in-pile temperature of about 325°C, the two sets of test and control magnets of this experiment were temperature cycled (stabilized) at this temperature. The magnetic induction, BOMC, for both the test and control specimens therefore decreased from initially optimum values by -89% and -79% respectively. The subsequent irradiation induced increase of 67% in the test magnet yielded a value of BOMC which was still 82% below its initial optimum value.

21. At high temperatures (∼325°C) the Cunife I magnet was almost completely degraded by temperature alone since the test and control magnets showed the same decrease in BOMC. At the intermediate temperature there was some evidence of a radiation
induced change greater than that caused by temperature alone. At a normal temperature the control sample was essentially unaffected but the test magnet showed a 13% increase. Its final $B_{OMC}$ value was, in fact, 3% higher than the initial optimum value for this sample. This magnet also had to be forcibly ejected from its jacket, which treatment would tend to demagnetize it.

**Stability of Changes**

22. In order to determine whether or not the changes caused by irradiation were stable, the two sets of magnets irradiated to $\sim 10^{20}$ nvt at elevated temperatures were retested 4-1/2 months after irradiation. As a check, the control sets were also retested four months after their temperature run. The data are shown in Table III. Most of the magnets maintained, within the experimental error, their immediate post-irradiation values. The 36 Cobalt Steel of one set (see column 1 of Fig. 3) was a notable exception. However, because of the intense gamma radiation resulting from the induced activity in the irradiated magnets the wooden containers in which they were stored broke down in places allowing some of the magnets to move closer together. The 36 Cobalt Steel was in tandem with the Alnico V and its drop in $B_{OMC}$ may have been due to the demagnetizing influence of the latter. Moreover, the control magnet of the second set (see 36 Cobalt Steel, column 4, Table III) showed considerably deterioration after four months of storage. Thus, the post-irradiation changes in both cases may be due to an instability inherent in magnets which have undergone major changes in $B_{OMC}$.

23. The two 3.1/2% Chromium Steel test magnets changed by appreciable amounts, +13, -18%. The controls changed also, although to a lesser extent. These changes are not significant since the $B_{OMC}$ values from which the changes occurred are only about 20% of what they would be for normally optimum values.

**Effects of $3 \times 10^{17}$ and $2 \times 10^{18}$ nvt on Demagnetization Curves**

24. Initially, two sets of thirteen magnets were irradiated in the Brookhaven National Laboratory Reactor to $3 \times 10^{17}$ nvt epicadmium. Both sets were then shipped to Argonne National Laboratory where they were irradiated a second time but in separate holes of the CP-5 reactor. The set used for demagnetization curve determinations was irradiated in hole VT-9 at a temperature of 500°C ± 20; the second set for the measurement of open magnetic circuit induction, $B_{OMC}$, was irradiated simultaneously in hole VT-5 at 300°C ± 20. The integrated flux for each set was about $2 \times 10^{18}$ nvt.
25. No changes due to neutron irradiation were detectable in the demagnetization curves for the experiment at \(3 \times 10^{17}\) nvt\(^{-1}\). At \(2 \times 10^{18}\) nvt the differences in the before and after demag-curves for eight of the irradiated magnets were the same as the before and after curves for the corresponding controls (e.g., see the curves for Alnico V and Alnico XII in Figs. 5, 9 and 10). The magnets which were not affected included the Alnicos II, V, XII, Cunife I, Cunico I, Platinum Cobalt and the Barium Ferrites (oriented and unoriented).

26. Of the remaining five of this set of thirteen, the ESD, Fine Iron and Fine Iron-Cobalt irradiated magnets (Figs. 7 and 8) showed changes smaller than those that occurred in their controls (Figs. 9 and 10); the 36 Cobalt Steel (Fig. 5), 3-1/2 Chromium Steel (Fig. 7) and Silmanal irradiated magnets showed changes which were larger than those of their controls (Figs. 9 and 10). The disparity in behavior between test and control magnets is probably due, primarily, to temperature differences in the test magnets which are not reflected in the value of \(500^\circ C \pm 20\) quoted above. This quantity represents the temperature monitored at one point in the assembly (\(\pm 20\) shows the range of variation in temperature caused by variation in-pile power, etc., and not the experimental error). The amount of gamma heating which can occur in a material is a function of its geometry, thermal conductivity, area of contact with the assembly in which it is mounted, etc. Therefore, the temperatures of some of the irradiated magnets may have exceeded \(520^\circ C\) or fallen below \(480^\circ C\) by substantial amounts. In contrast, the controls were uniformly heated in an oven and experienced the same temperatures to within a fraction of a degree.

27. One other consideration confirms the conclusion that the influence of temperature was largely responsible for the changes in the demagnetization curves. The changes in BOMC of the magnets irradiated simultaneously to \(2 \times 10^{18}\) nvt (but at the lower temperature of \(310^\circ C\)) together with the above set were due to temperature. Of the five materials which showed such pronounced differences between the test and control magnet demagnetization curves the two ESD materials showed negligible changes in BOMC for both test and control magnets; changes in BOMC for the Silmanal, 3-1/2 Chromium Steel and 36 Cobalt Steel irradiated magnets were the same as or less than those which occurred in the control magnets.

28. The set of magnets used for determining changes in the properties of the demagnetization curves was not irradiated further because temperature alone had caused large irreversible changes in five of the materials.
Effects on $\text{BO}_{\text{MC}}$, $3 \times 10^{17}$ nvt ($900^\circ\text{C}$)

29. None of the magnets tested gave any indication of the presence of radiation damage greater than an estimated experimental error of $\pm 2\%$. This together with the results for the demagnetization curves determined at this level sets a limit of about $3 \times 10^{17}$ nvt below which one should not expect to find any radiation damage.

Effects on $\text{BO}_{\text{MC}}$, $2 \times 10^{18}$ nvt, ($3100^\circ\text{C} \pm 5$)

30. Alnicos II and XII, Cunico I, 36 Cobalt Steel, Platinum Cobalt, Silmanal and the ESD magnets, Fine Iron and Fine Iron-Cobalt were not affected by irradiation to this level.

31. Two Barium Ferrite magnets showed changes of $-3\%$ in $\text{BO}_{\text{MC}}$. This is close to the $\pm 2\%$ experimental error but is considered significant because this material has exceptional stability even at elevated temperatures (compare percentage changes with those of the corresponding controls in Tables II and IV and Fig. 13).

32. $\text{BO}_{\text{MC}}$ for Alnico V decreased by 9%. This change is not considered typical in the light of all the data obtained for this material. At higher doses of integrated flux no radiation damage effects were observed.

33. The large changes in properties in Cunife I and 3-1/2 Chromium Steel were caused by the high temperatures. In fact, the changes in the controls were larger than those in the test magnets (Fig. 12). This is consistent with the tendency toward improvement in $\text{BO}_{\text{MC}}$ which was mentioned in the section describing the irradiation experiments at $\sim 10^{20}$ nvt (Fig. 3).

Effects of $3 \times 10^{19}$ nvt on $\text{BO}_{\text{MC}}$, ($2500^\circ\text{C} \pm 20$)

34. The following magnets were not affected by this irradiation: Alnicos II, V, XII, Cunico I, Platinum Cobalt, Cunife I, 3-1/2 Chromium Steel, ESD Fine Iron and Silmanal.

35. Materials such as Cunife I, 3-1/2 Chromium Steel and Platinum Cobalt - both test magnets and controls - were affected by the temperature of $3100^\circ\text{C}$ in the previous irradiation and yet were not affected by the $2500^\circ\text{C}$ temperature of this irradiation. The explanation for this is that the temperature-induced decreases in $\text{BO}_{\text{MC}}$ stabilized these magnets to any succeeding temperature excursions whose peaks were less than $3100^\circ\text{C}$. By contrast, the previously unaffected 36 Cobalt Steel magnet changed by $\sim -9\%$. Its controls did not change, thus indicating that the test magnet approached the threshold of radiation damage.
36. The ESD Fine Iron-Cobalt test sample disintegrated completely, probably due to the melting of the lead alloy matrix. Its controls also showed signs of deterioration, i.e., corrosion and swelling (difficulty was experienced in placing the search coil on the controls).

37. The two Barium Ferrites both showed near-threshold changes of -8%. At the highest integrated fluxes of these experiments, $5 \times 10^2$ nvt, the high temperature magnets showed changes of $\sim 22\%$. The progressive damage with integrated flux occurs over three orders of magnitude in nvt - $10^{17}$ to $10^{20}$. This is in sharp contrast to the effects of radiation on a typical soft magnetic material such as Supermalloy for which order of magnitude changes occurred in some properties for a change in nvt of only a factor of 2.

CONCLUSIONS

Results up to $5 \times 10^{20}$ Epicad nvt

38. Alnico II, Alnico V and Alnico XII are not affected by nuclear radiation up to $\sim 5 \times 10^2$ epicad nvt (and $\sim 2 \times 10^{19}$ nvt for neutrons with energies $\sim 2.9$ Mev) at temperatures ranging from $25^\circ C$ to $325^\circ C$.

39. Cunife I, Cunico I and 3-1/2 Chromium Steel should be operable at this level since they showed less than threshold of damage changes. Proper stabilization techniques could reduce expected radiation induced changes to negligible amounts. These materials would have to operate at temperatures of about $70^\circ C$ or less to minimize changes due to gamma heating. Tenzer has shown that Alnico V can withstand temperatures of $550^\circ C$ for one thousand hours with little or no change in remanence (BOMC). The Alnico magnets in this experiment showed no changes in the demagnetization curves for irradiation to $2 \times 10^{18}$ nvt at $500^\circ C$ (although no BOMC measurements were made at this temperature). This suggests the possibility of stabilizing Alnico V, II and other materials to withstand not only high temperatures but still higher integrated fluxes than were achieved or an environment combining both.

40. The Barium Ferrite magnets withstand irradiation to these levels better if the temperatures are about 235 to $325^\circ C$ than they do at a normal temperature of operation. Proper stabilization could reduce irradiation induced changes or, perhaps, eliminate them.

41. It is questionable whether stabilization techniques would compensate for the large changes which occurred in Platinum Cobalt, 36 Cobalt Steel, and Silmanal since large knockdowns of the order of -35% by stabilization may result in subsequent erratic behavior.
Results at $3 \times 10^{17}$ to $3 \times 10^{19}$ nvt

42. None of the magnets were affected by nuclear radiation of $3 \times 10^{17}$ nvt (at 300°C). Most were unaffected by the $2 \times 10^{18}$ nvt irradiation; only three showed changes not attributable to temperature. The change in Alnico V, -9%, is probably not representative. The changes in the Barium Ferrites, -3%, are tolerable or can be eliminated by stabilization. The results for the demagnetization curves at $3 \times 10^{18}$ nvt were not clear cut for five of the materials because of the high temperatures which were present. The differences between the curves of the test and control magnets were attributable to temperature effects.

43. BOMC for ten of the materials remained unaffected by irradiation to $3 \times 10^{19}$ nvt. The 36 Cobalt Steel and the oriented and unoriented Barium Ferrites approached but did not exceed the threshold of radiation damage. They would therefore be amenable to stabilization treatment to minimize changes caused by irradiation.

General

44. Various limitations precluded the performance of experiments in which adequate statistical results could be achieved. Nevertheless, a small measure of statistical success was attained. The three Alnico materials, for example, although somewhat different in composition, gave essentially the same results under six different irradiations (two for demagnetization curve determinations, four for BOMC measurements). The two Barium Ferrite materials - identical in composition - likewise showed the same changes after several irradiations. These experiments moreover, provide boundaries for further work in this field. Irradiation to integrated fluxes lower than $3 \times 10^{17}$ epicadmium would not be rewarding (excluding perhaps experiments which would reach this value by means of very high pulses of flux of very short duration). Future work on those materials which were unaffected could be started at the $10^{20}$ nvt level and pursued until the damage threshold was attained and exceeded. Another experiment of interest would be to irradiate permanent magnets in the presence of intense magnetic fields to see whether their nominal magnetic properties could be improved. Prerequisites of this work would include ample in-pile space for the equipment which produces the magnetic field and the necessity that such equipment would itself be resistant to radiation damage.

45. Two other factors must be considered with reference to the behavior of permanent magnets in radiation fields. First, adequate cooling must be taken into account even for materials
like the Alnicos which operate successfully in combined environments. The temperature equilibrium in a sealed container, which is inadequately cooled could exceed the Curie temperature of a magnet, this would result in its complete demagnetization. Second, the soft magnetic materials (high permeability materials) used in conjunction with permanent magnets in magnetic circuits may be more susceptible to radiation damage than the magnets themselves. Although previous work\textsuperscript{6} has shown that $B_{\text{max}}$ (see Fig. 5) the saturation induction, of soft magnetic materials is not affected at $3 \times 10^{17}$ episcad n/cm\textsuperscript{2}, not enough is known about how this property may be affected at the integrated fluxes of $\sim 5 \times 10^{20}$ episcad n/cm\textsuperscript{2} achieved in these experiments.

46. The following relations between changes due to temperature and radiation have been observed in these experiments (assuming thermal shocks are not of primary importance).

a. Radiation effects are independent of temperature effects, e.g., in 36 Cobalt Steel and Cunico I.

b. The presence of high temperatures during irradiation counteracts to some extent the effects of radiation, e.g., Alnico XII, both Barium Ferrites.

c. Irradiation effects counteract temperature induced changes: 3-1/2 Chromium Steel. Also, possibly, Cunife I and Barium Ferrites.

47. Alnicos II and V were not affected at the integrated fluxes achieved. Other materials for various reasons showed no conclusive trend.

48. Calculations of the formation, by transmutation, of isotopes which would act as impurities in the magnets show that this is a negligible factor contributing to radiation damage. The primary factor is that of physical damage to the lattice structure by the more conventional damage mechanisms.

ACKNOWLEDGEMENTS

49. The authors wish to thank the following individuals and organizations for their help and cooperation in performing these experiments:

a. P. Brown, V. Hall, A. D. Rossin, R. J. Armani and others of the Reactor Engineering Department of ANL. The cooperative effort between ANL and NOL was initiated by Mr. Brown. When Messrs. Hall and Brown left ANL, Dr. Rossin and R. J. Armani continued to provide for the time, facilities, and personnel in the Reactor Engineering Department, the Hot
Cell Laboratory and at the CP-5 reactor to continue the irradiation studies.

b. Dr. W. H. McCorkle, John Condelos and other personnel of CP-5 for carrying on the irradiations, handling the hot containers, etc.

c. W. Doe for providing the Hot cell facilities in the Hot Cell Laboratory, R. Enders for assisting in the measurements of the irradiated magnets in the hot cell, and other personnel for their assistance.

d. L. Steele of the Naval Research Laboratory for advice on methods of cooling containers placed in a reactor.


f. E. A. Bye of Simonds Saw and Steel Company for furnishing samples of 3-1/2 Chromium Steel and 36 Cobalt Steel.
REFERENCES


# TABLE I. PERMANENT MAGNET MATERIALS IRRADIATED - MAGNETIC CHARACTERISTICS, DESIGN FACTORS, HEAT TREATMENT

<table>
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</thead>
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<td>10 Al, 13 Co, 18 Ni, 6 Cu, 53 Fe</td>
<td>1.7</td>
<td>GOOD</td>
<td>340</td>
<td>C</td>
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<td>GOOD</td>
<td>340</td>
<td>CN</td>
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<td>EXCEPTIONAL</td>
<td>480</td>
<td>C</td>
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<td>&gt;500†</td>
<td>CR</td>
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<td>.25</td>
<td>POOR</td>
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<td>HR</td>
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<td>HR, C</td>
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<td>76.1 Pt, 13 Co, .1 Mn</td>
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<td>310†</td>
<td>A</td>
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<td>86 Ag, 8.4 Mn, 5.2 Al</td>
<td>.06</td>
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<td>CR, B</td>
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<td>480</td>
<td>S</td>
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<td>BaO - 6 Fe₂O₃</td>
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<td>450</td>
<td>S, N</td>
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<td>Fe, Lead Alloy Matrix</td>
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<td>320</td>
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<td>ESD FINE IRON COBALT</td>
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<td>320</td>
<td>ESD</td>
</tr>
</tbody>
</table>

* Adapted, in part, from Indiana Steel Permanent Magnet Manual No. 6
† As determined in these experiments
C - Cast, heat treated
CR - Cold reduced
CR,B - Cold reduced (by swaging), s ow baked at *500°C

HR - Hot rolled and formed
M - Melted in a magnetic field
S - Pressed from powder and sintered
ESD - Ultrathin lamellated single domain particles of iron or iron-cobalt compacted in matrix and baked or coined
A - Arc melted into rod shape, machined, annealed
TABLE II. PERCENTAGE CHANGES IN OPEN MAGNETIC CIRCUIT INDUCTION (BOMC)
OF PERMANENT MAGNETS AS A RESULT OF IRRADIATION WITH \( \sim 10^{20} \) n/cm² (EPICADMIL™)

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<tr>
<th>Material</th>
<th>( ~ 60^\circ C )</th>
<th>( ~ 235^\circ C )</th>
<th>( ~ 325^\circ C )</th>
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<td>Control</td>
<td>Sample</td>
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<td>&lt;.5</td>
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<td>-6.5</td>
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<td>-4.5</td>
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<td>-1</td>
<td>-11.5</td>
</tr>
<tr>
<td>CUNIFE I</td>
<td>+13</td>
<td>-5</td>
<td>-52.5</td>
</tr>
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<td>3-1/2 CHROMIUM STEEL</td>
<td>+2.5</td>
<td>-1.5</td>
<td>-68</td>
</tr>
<tr>
<td>36 COBALT STEEL</td>
<td>+37</td>
<td>-1</td>
<td>-37.5</td>
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<td>-</td>
<td>-</td>
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<td>SILMANAL</td>
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<td>-1.5</td>
<td>-72.5</td>
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<td>+1</td>
<td>-24.5</td>
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<td>BARIUM FERRITE (UNORIENTED)</td>
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<td>&lt;.5</td>
<td>-21</td>
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<th>ACTUAL ENVIRONMENTAL CONDITIONS</th>
<th>Sample</th>
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<th>Control</th>
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<td>PEAK TEMP. (°C)</td>
<td>72</td>
<td>71</td>
<td>247</td>
<td>270</td>
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<td>355</td>
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<td>AVERAGE TEMP. (°C)</td>
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<td>58</td>
<td>235</td>
<td>225</td>
<td>325</td>
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<td>EPICAD nvt</td>
<td>(4 \times 10^{20})</td>
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<tr>
<td>FAST nvt (E &gt; 2.9 MeV)</td>
<td>(1.7 \times 10^{19})</td>
<td>(1.1 \times 10^{19})</td>
<td>(1.8 \times 10^{19})</td>
<td></td>
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<tr>
<td>THERMAL nvt</td>
<td>(1.6 \times 10^{20})</td>
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<td>DURATION (DAYS)</td>
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<td>65</td>
<td>65</td>
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TABLE II

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<th>Sample</th>
<th>Control</th>
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<tr>
<td>ALNICO XII</td>
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<td>-1</td>
<td>-.5</td>
<td>-6.5</td>
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<tr>
<td>CUNICO I</td>
<td>&lt;+.5</td>
<td>-.5</td>
<td>+.5</td>
<td>&lt;+.5</td>
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<tr>
<td>CUNIFE I</td>
<td>+.5</td>
<td>-.5</td>
<td>+13*</td>
<td>+3</td>
</tr>
<tr>
<td>3-1/2 CHROMIUM STEEL</td>
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<td>-8*</td>
<td>-18*</td>
<td>-3</td>
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<tr>
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<td>-1.5</td>
<td>-3</td>
<td>-28</td>
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<td>-.5</td>
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<td>~0</td>
<td>+.5</td>
<td>+23*</td>
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nvt, epicadmium 2 x 10^20

Temperature °C | ~235°C | ~235°C | ~325°C | ~325°C

* BOMC so reduced by previous events (irradiation or heating) that these large changes are the result of inherent instability of open magnetic circuit induction at low levels.

† Improvement due to heating at temperature of annealing.
**TABLE IV. PERCENTAGE CHANGES IN OPEN MAGNETIC CIRCUIT INDUCTION OF IRRADIATED MAGNETS AT INTERMEDIATE VALUES OF INTEGRATED NEUTRON FLUX COMPARED WITH CONTROLS**

<table>
<thead>
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<th>(\sim 10^{18})nvt</th>
<th>(\sim 10^{19})nvt</th>
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<td>ALNICO XII</td>
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<tr>
<td>CUNICO I</td>
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<td>-3</td>
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<tr>
<td>CUNIFE I</td>
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</tr>
<tr>
<td>3-1/2 CHROMIUM STEEL</td>
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<td>-39.5</td>
</tr>
<tr>
<td>36 COBALT STEEL</td>
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<td>-10</td>
</tr>
<tr>
<td>PLATINUM COBALT</td>
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<td>-33</td>
</tr>
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<td>SILICANAL</td>
<td>-10</td>
<td>+12.5</td>
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<tr>
<td>BARIUM FERRITE (ORIENTED)</td>
<td>-3</td>
<td>+5</td>
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<tr>
<td>BARIUM FERRITE (UNORIENTED)</td>
<td>-3</td>
<td>-5</td>
</tr>
<tr>
<td>ESD FINE IRON</td>
<td>-3.5</td>
<td>-5</td>
</tr>
<tr>
<td>ESD FINE IRON-COBALT</td>
<td>-6.5</td>
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<tr>
<td>nvt, epicadmium</td>
<td>(3 \times 10^{18})</td>
<td>---</td>
</tr>
<tr>
<td>Temperature (^\circ C) (peak)</td>
<td>315</td>
<td>310</td>
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* Physically Disintegrated
Figure 1. Nominal Demagnetization Curves of the Permanent Magnet Materials Irradiated
Figure 2A. High Temperature Irradiation Assembly - Partially Disassembled
Figure 2B. Container Used for Optimum Utilization of Reactor Water Cooling to Hold Gamma Heating Temperature to ~60°C. Magnets and Aluminum Plugs Alternately Spaced
<table>
<thead>
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<th>Material</th>
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<td>$2 \times 10^{12}$ NVT, 235°C</td>
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<td>Control, 60°C</td>
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<td>Control, 235°C</td>
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<td>Control, 325°C</td>
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<td></td>
<td>Epicadmium</td>
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<td>Barium Ferrite (Unoriented)</td>
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<td>Platinum Cobalt</td>
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<tr>
<td>3 ½ Chromium Steel</td>
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<tr>
<td></td>
<td>To 67%</td>
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<td>Cunife I</td>
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**Figure 3.** Comparison of Percentage Changes in Open Magnetic Circuit Induction, $B_{omc}$, for Magnets Irradiated to $\sim 10^{20}$ epicadm nvt at Normal and High Temperatures.
Figure 4. Method of Analysis of Changes in BOMC Produced by Fictitious Demagnetizing Fields, $\Delta H$'s, on Nominal Demagnetization Curve for 36 Cobalt Steel
Figure 5. Alnico V, Alnico XII, 36 Cobalt Steel, Cunife I: Effect on Demagnetization Curves of Irradiation with $2 \times 10^{18}$ epipad mt at $\sim 500^\circ$C
Figure 6. Platinum Cobalt, Barium Ferrites Oriented and Unoriented, Silmanal: Effect on Remagnetization Curves of Irradiation with $2 \times 10^{18}$ Epicad mvt at $\sim 500^\circ$C
Figure 7. ESD Fine Iron, Cunico I, 3-1/2 Chromium Steel: Effect on Demagnetization Curves of Irradiation with $2 \times 10^{18}$ Epical nvt at $\sim 500^\circ$C
Figure 8. ESD Fine Iron-Cobalt, Alnico II: Effect on Demagnetization Curves of Irradiation with $2 \times 10^{18}$ Eupid at $\sim 500^\circ C$.
Figure 9. Alnico V, 36 Cobalt Steel, ESD Fine Iron-Cobalt: Effect of Temperature (\( \sim 500^\circ \text{C} \)) on Unirradiated Control Magnets
Figure 11. Alnico V, Alnico XII, Alnico II: Effects of Irradiation and Temperature Control Events on Open Magnetic Circuit Induction (OMC).

- **Irradiated Magnets**
- **Control Magnets**

<table>
<thead>
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<th>Before</th>
<th>After</th>
<th>2nd Event</th>
<th>Before</th>
<th>After</th>
<th>3rd Event</th>
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Uranium, Thorium, and Plutonium
Figure 13. Cunico I, Barium Ferrites Oriented and Un-oriented, Silmanal: Effects of Irradiation and Reactor Temperature Simulation Events on Open Magnetic Circuit Induction (BOMC)
Figure 14. ESD Fine Iron-Cobalt, ESD Fine Iron: Effects of Irradiation and Reactor Temperature Simulation Events on Open Magnetic Circuit Induction (BONC)
Figure 15. 36 Cobalt Steel, Platinum Cobalt: Effects of Irradiation and Reactor Temperature Simulation Events on Open Magnetic Circuit Induction (BOMC)
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