UNCLASSIFIED

AD 262 261

UNCLASSIFIED

NRL Report 5686

THE EFFECTS OF IRRADIATION ON MAGNETIC PROPERTIES OF ALLOYS AND FERRITES

A. I. Schindler

Metal Physics Branch
Metallurgy Division

August 11, 1961

DEPARTMENT OF THE NAVY · OFFICE OF NAVAL RESEARCH

U. S. NAVAL RESEARCH LABORATORY
Washington, D.C.

Copies available at OTS $1.00

CONTENTS

PROBLEM STATUS

This is an interim report on the problem; work is continuing.

AUTHORIZATION

NRL Problem M01-10
Project RR 007-01-46-5408

Manuscript submitted July 27, 1961.

THE EFFECTS OF IRRADIATION ON MAGNETIC PROPERTIES OF ALLOYS AND FERRITES

A. I. Schindler
U.S. Naval Research Laboratory
Washington, D.C.

ABSTRACT

Studies have been conducted on the effects of neutron irradiation on the magnetic characteristics of metal alloys and ferrites. The magnetic properties examined include the coercive force, remanence, shape of the hysteresis loop, Curie temperature, and magnetic moment. Only the Curie temperature was found to be relatively insensitive to neutron irradiation; all the other properties were modified by varying amounts. The radiation sensitivity of metal alloys and selected ferrites appears to be primarily related to the dependence of magnetic properties on the short-range ordering of the atoms along certain crystallographic directions. Materials which can be easily ordered show the greatest sensitivity. It has been found that the application of a magnetic field during irradiation can cause directional ordering to take place which in many cases produces more desirable magnetic characteristics. The studies of the ferrites are more complicated since the effects appear to be related to atomic displacements as well as possible radiation induced oxidation. This has been deduced from magnetic measurements of irradiated samples coupled with neutron diffraction data of the same material.

BACKGROUND

When a crystalline solid is exposed to neutron irradiation, significant changes in many of its physical properties are observed (1). In general, these modified properties are a consequence of the resultant ionization, transmutations, thermal spikes, and atomic displacements. Atomic displacements can be further subdivided into formation of vacancies and interstitials, replacements, and displacement spikes which may collapse to form dislocation rings. It would appear that radiation-modified magnetic properties are caused primarily by the atomic displacements and thermal spikes which are responsible for increasing the atomic mobility or diffusion, for changing the concentration gradients, and for introducing coalesced defects and their resultant strains.

In studying the effects of irradiation on magnetic properties, observation of the changes in the magnetic hysteresis loop is extremely convenient and relatively easy. Figure 1 shows a typical magnetic hysteresis loop. As the magnetic field, H, is increased from zero, the value of the magnetic induction, B, of a demagnetized sample increases with a non-zero slope, called the initial permeability, μ_o. As H is further increased, the slope passes through a maximum and then decreases to approximately one. The maximum slope is called the maximum permeability, μ_{max}. Technical saturation is defined as the state represented by that portion of the curve where the slope is equal to one, and the induction attains a maximum value, known as the saturation induction, B_S. If now the applied field is reduced to zero, a residual induction or remanence, B_R, is found to remain. To reduce the magnetic induction to zero, a field must be applied in the negative direction. The value of the applied field which gives zero magnetic induction is called the coercive force, H_C. The shape of the hysteresis loop and the values of the loop parameters are strongly dependent upon such properties as atomic arrangement and state of strain and are consequently affected by neutron irradiation.

METHODS OF MEASUREMENT

The types of experiments that have been performed include measurements of the effects of irradiation upon

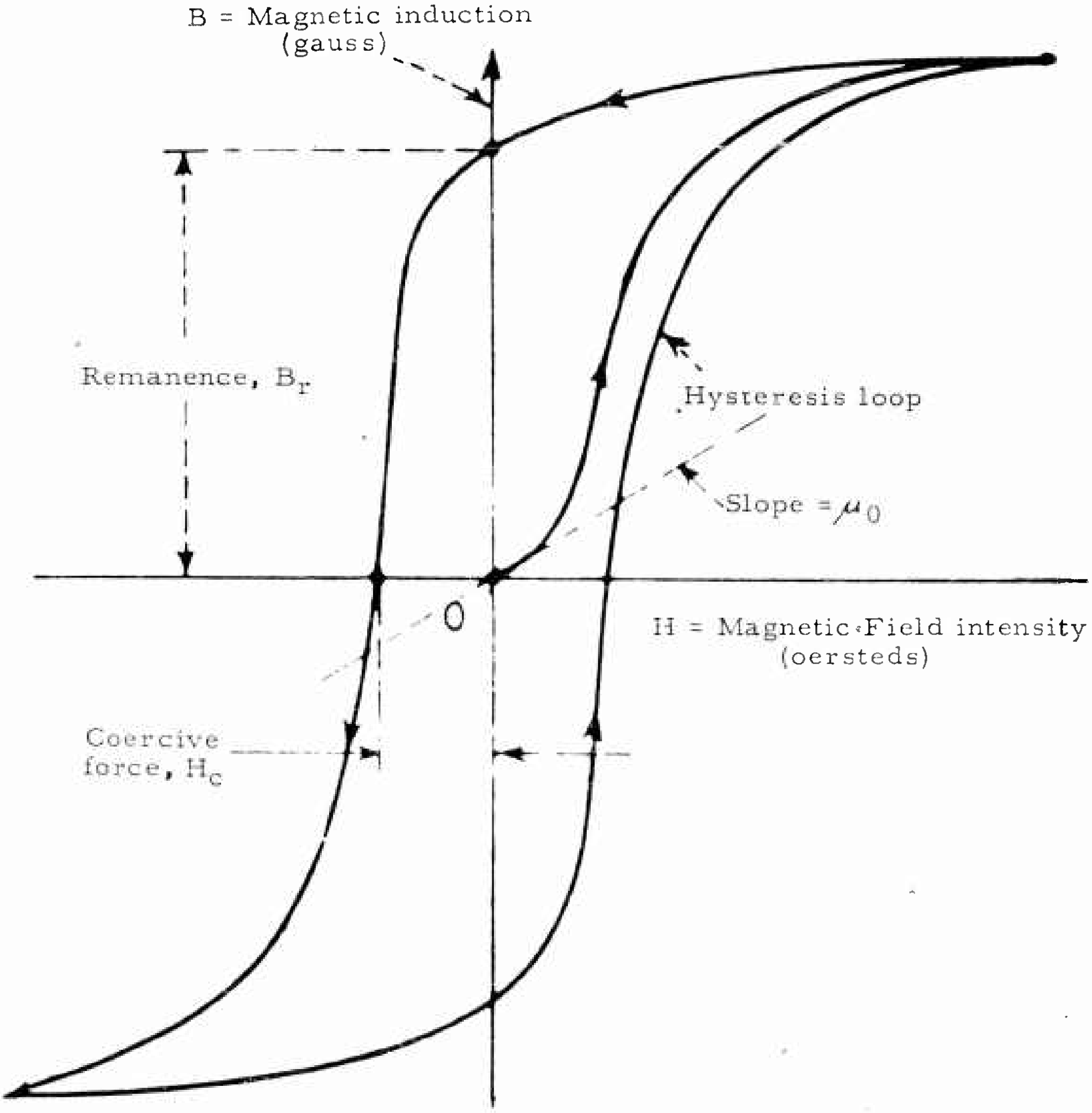


Figure 1 - A typical magnetic hysteresis loop.

the hysteresis loop parameters, the magnetic moments, and the Curie temperatures. In all cases, pre-irradiation measurements were made and then the samples were placed in aluminum cans and irradiated at the Brookhaven Graphite Reactor for multiples of a two-week period. Radiation monitors and thermocouples were used during irradiation for the measurement of the neutron flux and sample temperature during the irradiation period. To prevent overheating, compressed air cooling was employed. The samples were then returned to NRL for post-irradiation measurements.

For the measurement of the hysteresis loops, toroidal samples with appropriately wound B and H coils were employed; Curie temperature measurements were made on rods and on toroids which were cut in half; magnetic moment measurements were made on oxide powders. Details of the measurement and irradiation procedures have been reported elsewhere (2-6) and will not be repeated here.

In the course of this program two distinctly different classes of magnetic materials were investigated: ferromagnetic metal alloys and ferrimagnetic metal oxides commonly called ferrites. Since the structures, the composition dependence of magnetization, and the magnetization mechanisms are so different for these two types of materials, it is desirable to treat the results separately. The first portion of this paper will consider the metal alloys, while the second portion will be devoted to the results obtained on ferrites.

I. METAL ALLOYS

The most direct mechanism relating magnetic properties of metals and neutron irradiation is based on the strain sensitive nature of the initial and maximum permeability. Kersten (7) has shown that if the process of magnetization is entirely by domain rotation, and if the internal strains are randomly oriented, then the initial permeability can be written:

$$\mu_{\circ} = 1 + \frac{(B_S - H_S)^2}{6\pi\lambda_S\sigma_i} ,$$

where H_S is the applied field necessary for technical saturation, λ_S represents the saturation value of the magnetostriction, and σ_i is the internal strain. Therefore as the internal strain is increased by any means, μ_o would be expected to decrease. The value of μ_{max} should be affected in a similar manner. The introduction of strains by irradiation-induced defect production would of course result in a reduction of the value of μ_o and μ_{max}, in strain-sensitive magnetic materials.

In addition to being dependent upon strain, μ_o, μ_{max}, H_C, and B_R are all known to be sensitive to the state of atomic order. Our knowledge of the effects of order on magnetic properties is perhaps most complete for a large class of commercially significant magnetic materials known as permalloys. Permalloys are iron-nickel alloys of two basic compositions: 50% Fe-50% Ni and 75% Ni-25% Fe, i.e., NiFe and Ni_3Fe, plus, in many cases, small amounts of Mo, of Si, or of Cu and Cr additions. In 1923, Ellman (8) found that superior magnetic properties could be obtained for the permalloys by employing a double heat treatment consisting of heating to 950°C for an hour and furnace cooling, then heating to 600°C and air quenching at a maximum rate of 1500°C per minute. The compositions most sensitive to this type of treatment range from 50% Ni-50% Fe to 90% Ni-10% Fe, with a maximum sensitivity occurring at 78% Ni-22% Fe. It was later determined that Ni_3Fe was an order-disorder alloy with an ordering temperature of 490°C and the magnetic properties were in some way a function of the state of order. Quenched or disordered permalloy was observed to have low magnetic anisotropy and magnetostriction and high magnetic permeability. Slow cooling or ordering, on the other hand, caused the absolute value of the anisotropy and magnetostriction to increase and the permeability to decrease.

A series of experiments was performed by Kaya (9) which illustrates the effects of order on the hysteresis loop. The results of this investigation are shown in Figs. 2 and 3. A sample of quenched Ni_3Fe (76% Ni) was annealed for various lengths of time at 490°C and the hysteresis loops were examined as a function of annealing time. As the ordering of the Ni_3Fe sample proceeds, it can be seen that initially the remanence decreases and the coercive force increases. At some critical stage of the ordering process a constricted or kinked hysteresis

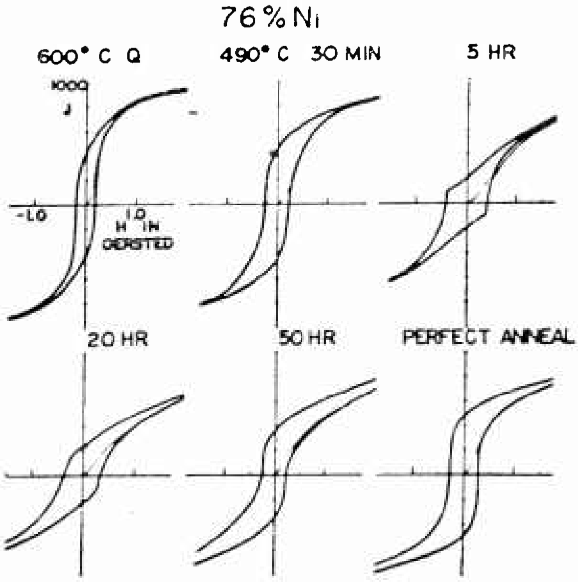


Figure 2 - Variation of the shape of the hysteresis loop for 76 Permalloy with time of annealing. (Annealing temperature - 490°C.)

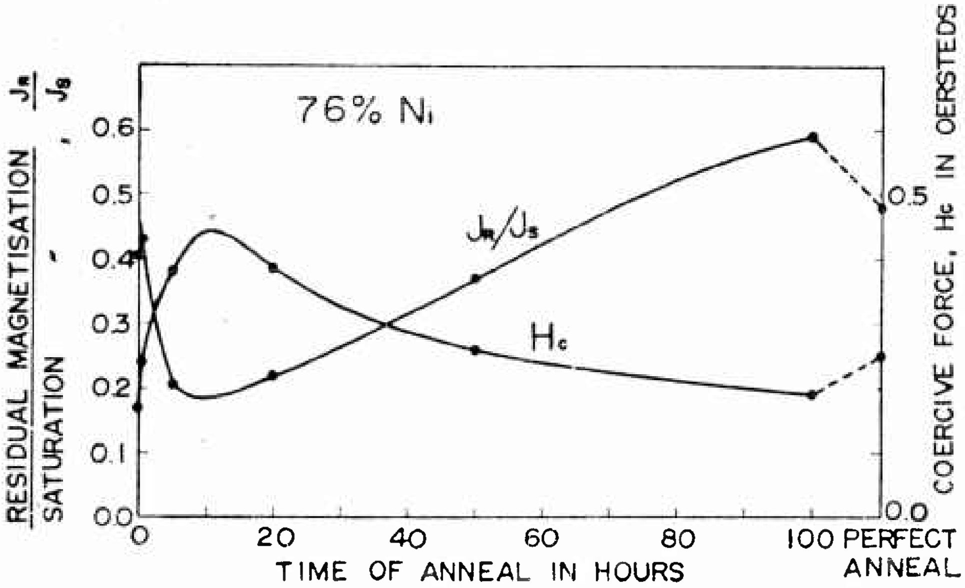


Figure 3 - Dependence of the coercive force and the remanence of 76% Ni-Permalloy on the time of annealing. (Annealing temperature - 490°C.)

loop is obtained. Further ordering causes the coercive force to decrease and the remanence to increase. In Figure 3 the coercive force, H_C, and the remanence, B_R, are shown as a function of the annealing time, or consequently, the state of order. (B_R here is defined in terms of J_R/J_S, where J is the intensity of magnetization, subscript S refers to a saturation value and R to remanence value, $J_R/J_S = B_R/(B_S-H_S)$.) Although the extrema of these curves are not clearly defined it appears that the minimum in B_R and the maximum of H_C occur at the same stage of the ordering process.

If these samples are partially ordered thermally in the presence of an externally applied magnetic field, a rectangular hysteresis loop is obtained. In the case of such a loop, the preponderance of easy directions of magnetization of the individual domains are aligned parallel or antiparallel to the field applied during the thermal annealing. An alternate statement phenomenologically describing this result is that uniaxial anisotropy has resulted from the magnetic annealing. L. Neél (10) and S. Taniguchi (11) have independently proposed an explanation for the uniaxial anisotropy created in a binary alloy annealed in a magnetic field which can also explain the results of the Kaya experiment carried out in zero field. According to their model, directional diffusion takes place and local or short-range anisotropic atomic configurations result. This model is schematically illustrated in Fig. 4. Consider a solid solution of two types of atoms, where the solid circles represent the A atoms, and open circles represent the B atoms. In the completely random state, equal numbers of similar-atom pairs are to be expected in all directions, as shown in Figure 4a. For perfect long-range order no similar-atom pairs will be found, as in Fig. 4b. The directional short-range ordering is shown in Fig. 4c. In this latter case, because of the directional diffusion, there is a definite directionality to the alignment of similar-atom pairs. Such short-range directional ordering can stabilize the magnetization in a direction related to the atomic anisotropy with consequent growth in uniaxial magnetic anisotropy. Taniguchi (11) has further proposed that directional ordering effects can occur even if the alloy is heat treated in the absence of a magnetic field. In this case, the internal magnetic field of each domain will influence the directionality of diffusion. Now,

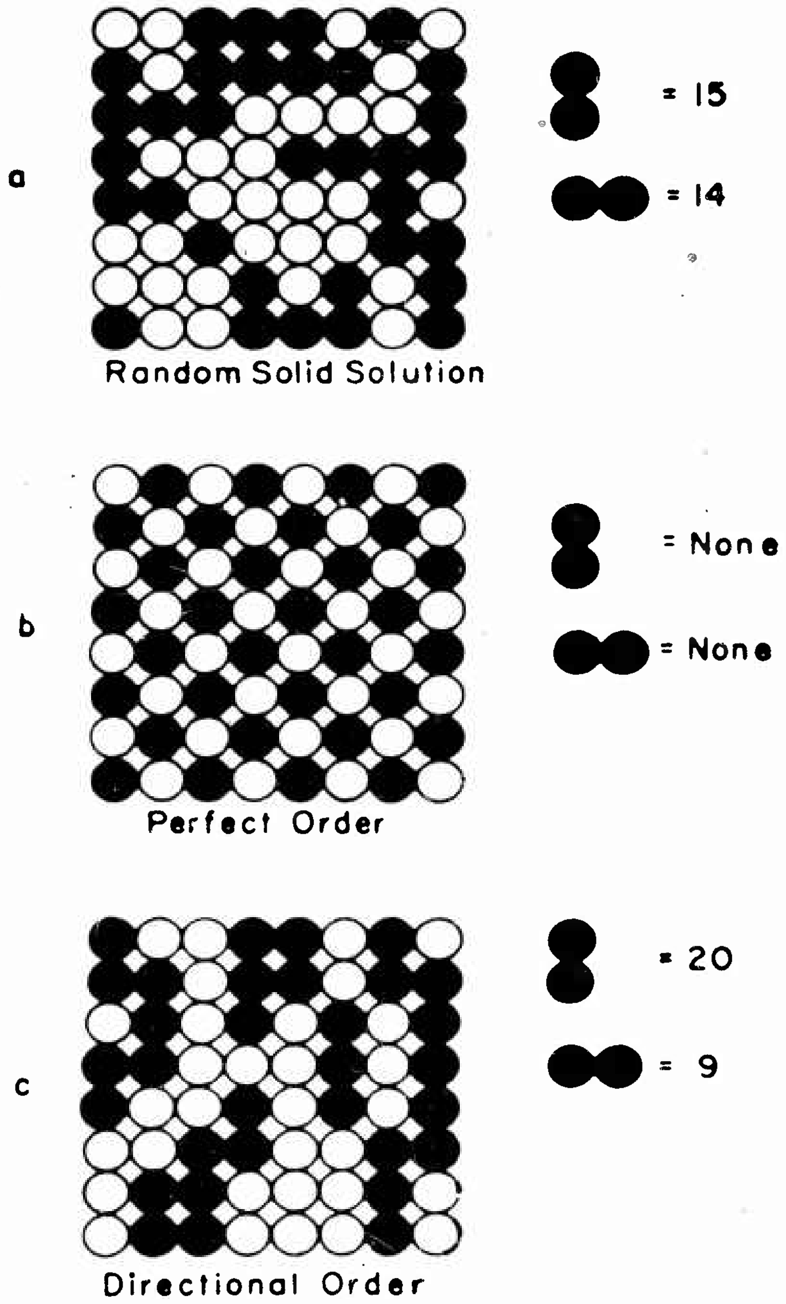


Figure 4 - Possible arrangement of atoms in a 50-50 alloy of black and white atoms.

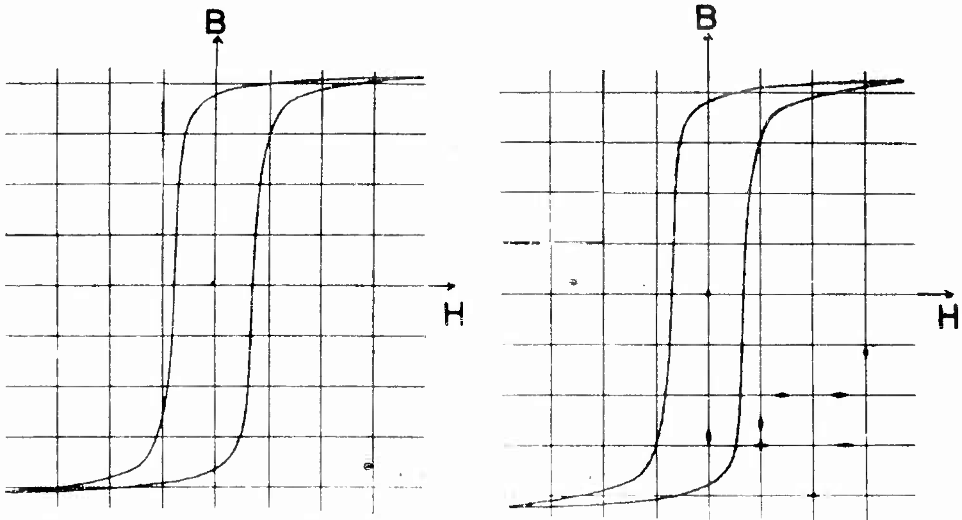
however, this direction will change from domain to domain, in accordance with the internal field distribution during the annealing.

If the normal magnetic anisotropy is small, as in the case of permalloys, then any additional uniaxial anisotropy has a strong effect on the magnetization process. The direction of the uniaxial anisotropy that is created by annealing in a magnetic field will be the same for all of the domains, and this will result in a square hysteresis loop. Annealing in the absence of a magnetic field also causes uniaxial anisotropy, but the direction of the anisotropy will be different from domain to domain, and a constricted or kinked hysteresis loop will be obtained. An indirect method of examination of the growth of uniaxial anisotropy can then be made by a study of the magnetic hysteresis loop. However, for iron-nickel alloys there is also a tendency for long-range superlattice formation, i.e., formation of Ni_3Fe. Long-range ordering, on the other hand, impedes the formation of directional short-range ordering. As the formation of the Ni_3Fe superlattice occurs, the short-range directional ordering and, consequently, the uniaxial anisotropy decreases, so that the more normal hysteresis loop is obtained. The new hysteresis loop will, of course, have different values of coercive force and remanence consistent with the values of cubic anisotropy and magnetostrictive constants of the ordered alloy.

Thus, according to the models presented, neutron irradiation would be expected to affect magnetic characteristics of metal alloys as a result of the introduction of local strain into the lattice of strain-sensitive materials and as a result of changes in the state of local order for order-disorder alloys.

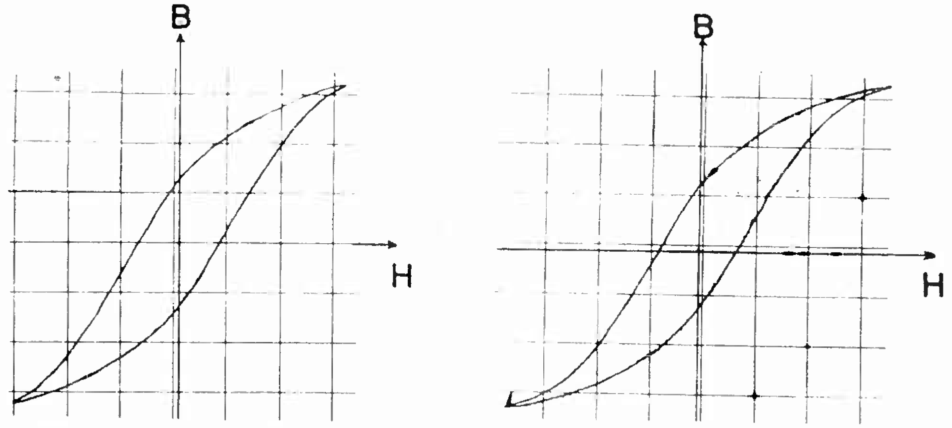
RESULTS

In general, large changes due to irradiation were found for those samples which are known to experience order-disorder transformations. Some samples, however, were found to have magnetic properties relatively unaffected by neutron irradiation. Figure 5 shows the results of neutron irradiation on a grain-oriented, 3% silicon steel. This steel is similar to that used in electrical transformer cores. The sample had been



BEFORE IRRADIATION AFTER IRRADIATION

Figure 5 - Effect of irradiation on the 60 cycle hysteresis loop of grain oriented silicon steel.



BEFORE IRRADIATION AFTER IRRADIATION

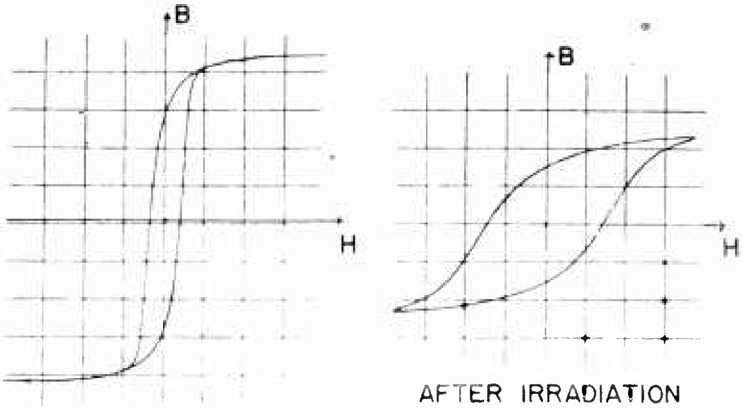
Figure 6 - Effect of irradiation on the 60 cycle hysteresis loop of Heusler alloy.

irradiated at the Brookhaven Reactor for 4 weeks to a total dosage of between 6×10^{16} nvt and 3×10^{17} nvt. The pre- and post-irradiation 60-cycle hysteresis loops indicate that to an accuracy of 5%, no changes resulted.

Figure 6 shows the pre- and post-irradiation 60-cycle hysteresis loops of a Heusler alloy, Cu_2MnAl. This material is ferromagnetic in the ordered state, but only feebly ferromagnetic in the disordered state. The initial state of order is unknown, but the pre-irradiation hysteresis loop indicated that the sample was partially ordered. After a four-week irradiation period, no significant changes in the magnetic properties were detected. More positive results might have been obtained if initially this sample were completely ordered or disordered, and such experiments are being planned.

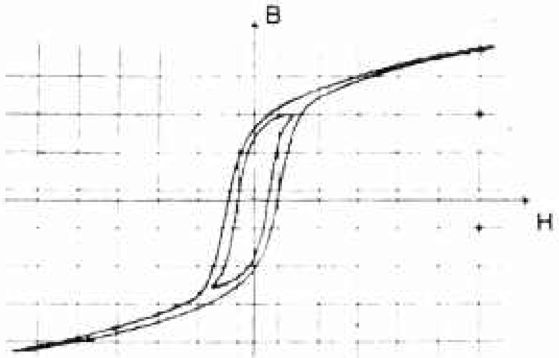
Figure 7 shows the pre- and post-irradiation hysteresis loop of a sample of supermalloy (5% Mo-79% Ni-16% Fe). This sample, which can be considered similar to Ni_3Fe, with added molybdenum, is strain sensitive and can be thermally ordered. The before-irradiation hysteresis loop, as shown in the upper left hand curve, is typical of a partially disordered material. After irradiation, the 60-cycle hysteresis loop obtained using the same maximum applied field is as shown in the upper-right-hand curve, and is characteristic of a magnetically unsaturated sample. This indicates that the magnetic permeability of this sample has been drastically lowered by the neutron irradiation. The lower curve shows the saturated hysteresis loop plotted with a contracted H axis, and the inner hysteresis loop is a duplicate of the upper-right-hand curve plotted to this new scale. Examinations of the saturated loops indicate that the irradiation has caused H_C to increase and B_R to decrease. Although no constricted loop was found, the results suggest that ordering is occurring as a result of the irradiation.

The results shown in Figs. 8, 9, and 10, however, clearly indicate the possibility of ordering resulting from neutron irradiation. The sample illustrated in Fig. 8 is permalloy, 4% Mo-79% Ni-17% Fe (similar to Ni_3Fe, with added Mo), the sample illustrated in Fig. 9 is a silicon-nickel-iron alloy, 3% Si-43% Ni-54% Fe, (similar to NiFe, with added Si), and the sample illustrated in Figure 10 is a nickel-iron alloy, 48% Ni-52% Fe (similar



BEFORE IRRADIATION

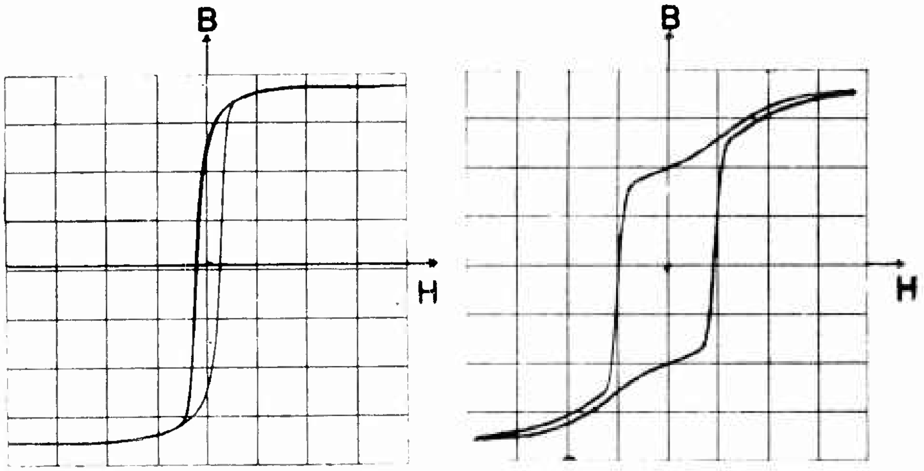
AFTER IRRADIATION



AFTER IRRADIATION
(CONTRACTED H AXIS)

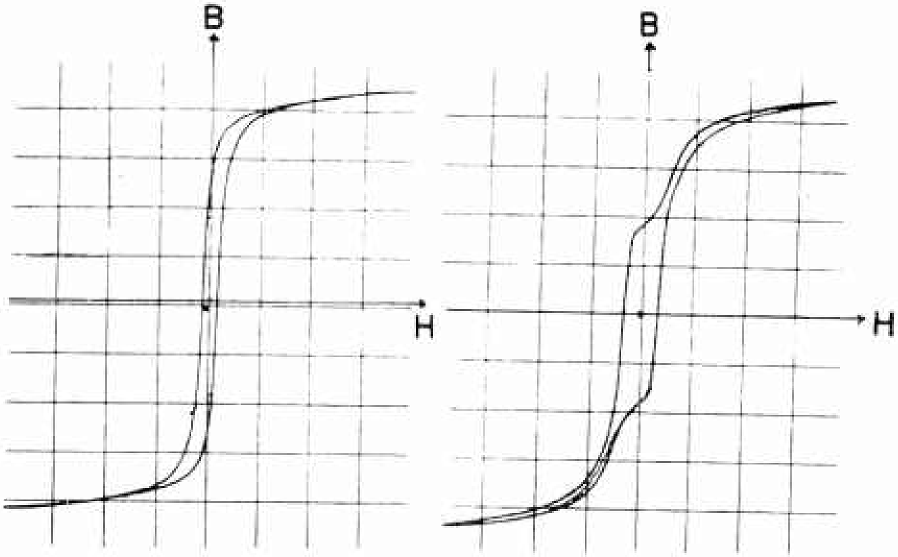
5-79 Mo PERMALLOY

Figure 7 - Effect of irradiation on the 60 cycle hysteresis loop of supermalloy.



BEFORE IRRADIATION AFTER IRRADIATION

Figure 8 - Effect of irradiation on the 60 cycle hysteresis loop of permalloy.



BEFORE IRRADIATION AFTER IRRADIATION

Figure 9 - Effect of irradiation on the 60 cycle hysteresis loop of 3% Si - 43% Ni - 54% Fe.

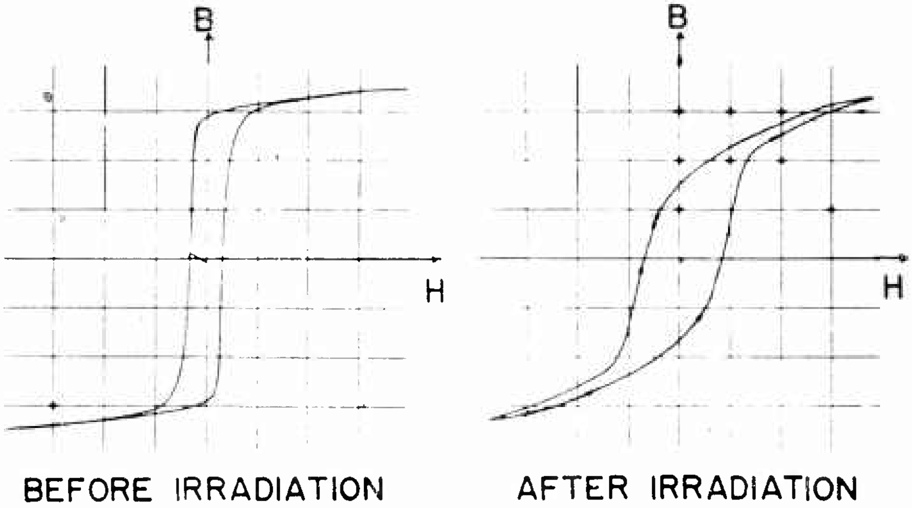


Figure 10 - Effect of irradiation on the 60 cycle hysteresis loop of 48% Ni - 52% Fe.

to NiFe). All three samples are known to undergo ordering transformations, and the pre-irradiation hysteresis loops are again characteristic of partially disordered samples. After 4 weeks of neutron irradiation the hysteresis loops shown in Figs. 8 and 9 are of the constricted type while that shown in Fig. 10 appears to be just in the initial stage of constricted loop formation. These curves seem to indicate that ordering results from the neutron irradiation. If this view is correct, irradiation is much more effective in producing order, even at room temperature, than thermal treatment is at elevated temperatures. This result is deduced from the post-irradiation curves shown in Figs. 9 and 10. Both samples are NiFe-type material, rather insensitive to thermal ordering and difficult to treat thermally to obtain constricted hysteresis loops.

To determine the relationship between thermal ordering and ordering resulting from neutron irradiation, examinations were made of the effect of irradiation on samples of material which had been ordered partially by thermal treatment. Four toroids of each of three types of 50% Ni-50% Fe alloys (47% Ni-50% Fe-3% Mo, square-loop 50% Fe-50% Ni, and 48% Ni-52% Fe) and three types of 75% Ni-25% Fe alloys (permalloy: 4% Mo-79% Ni-17% Fe, supermalloy: 5% Mo-79% Ni-16% Fe, and mumetal: 5% Cu-2% Cr-77% Ni-16% Fe) were heat treated to different degrees of order prior to irradiation. Although Ni_3Fe is known to have a critical temperature on cooling of 490°C, the ordering temperatures of the commercial materials are less well known. The heat treatments used were selected in an attempt to determine the ordering temperature and are as follows:

Treatment 1 - As received, presumably the material had been treated by the supplier to obtain optimum magnetic properties and consequently was mainly disordered.

Treatment 2 - Annealed for 138 hours at 600°C, then furnace cooled.

Treatment 3 - Annealed for 49 hours at 600°C, followed by cooling at 2°C/hr to 400°C, then furnace cooled.

Treatment 4 - Heated to 550°C, followed by cooling 1°C/hr to 400°C, then furnace cooled.

An examination of the resultant 60-cycle hysteresis loops indicated that the 50% Ni-50% Fe type of material was changed most by treatment 2 while the hysteresis loops of the 75% Ni-25% Fe type material were changed most by treatment 4. Since neither the coercive force nor the remanence is a linear function of the state of order, it was not possible to determine the degree of order produced by the thermal treatment, although some notion of the relative amount of ordering could be obtained. The samples were then irradiated at Brookhaven and the average value of the total integrated flux is as follows:

$\phi_{\text{thermal}} = 1.4 \times 10^{19}$ nvt
$\phi_{\text{intermediate}} = 7.0 \times 10^{17}$ nvt
$\phi_{\text{fast}} = 5.2 \times 10^{16}$ to 2.1×10^{17} nvt.

The pre- and post-irradiation hysteresis loops of the samples investigated are shown in Figs. 11 through 16, and the heat treatment used is indicated above each curve. In no case did the thermal treatment result in a constricted loop, so that all the pre-irradiation samples can be assumed to be in some state between total disorder and the critical ordering necessary for constricted loops. In this initial region, the coercive force increases and the remanence decreases monotonically as a function of increasing order. The pre-irradiation curves were therefore lined up in order of increasing H_C or decreasing B_R, whichever was most significantly changed by the heat treatment used. It is presumed that this is the direction of increasing order.

The post-irradiation curves were almost entirely of the constricted type, and for these, two general types of behavior were found. For the difficult to order, NiFe-type alloys (Figs. 11, 12, 13) the biggest changes occurred in the remanence, and this quantity was found to tend toward lower values (from left to right in each figure). It would appear, therefore, that if the sample was thermally more ordered before irradiation it would be more ordered after irradiation as well. This suggests that thermal ordering and radiation-induced ordering are similar processes and the results are additive.

In Figs. 14, 15, and 16, the more easily ordered Ni_3Fe-type samples, a somewhat different behavior was

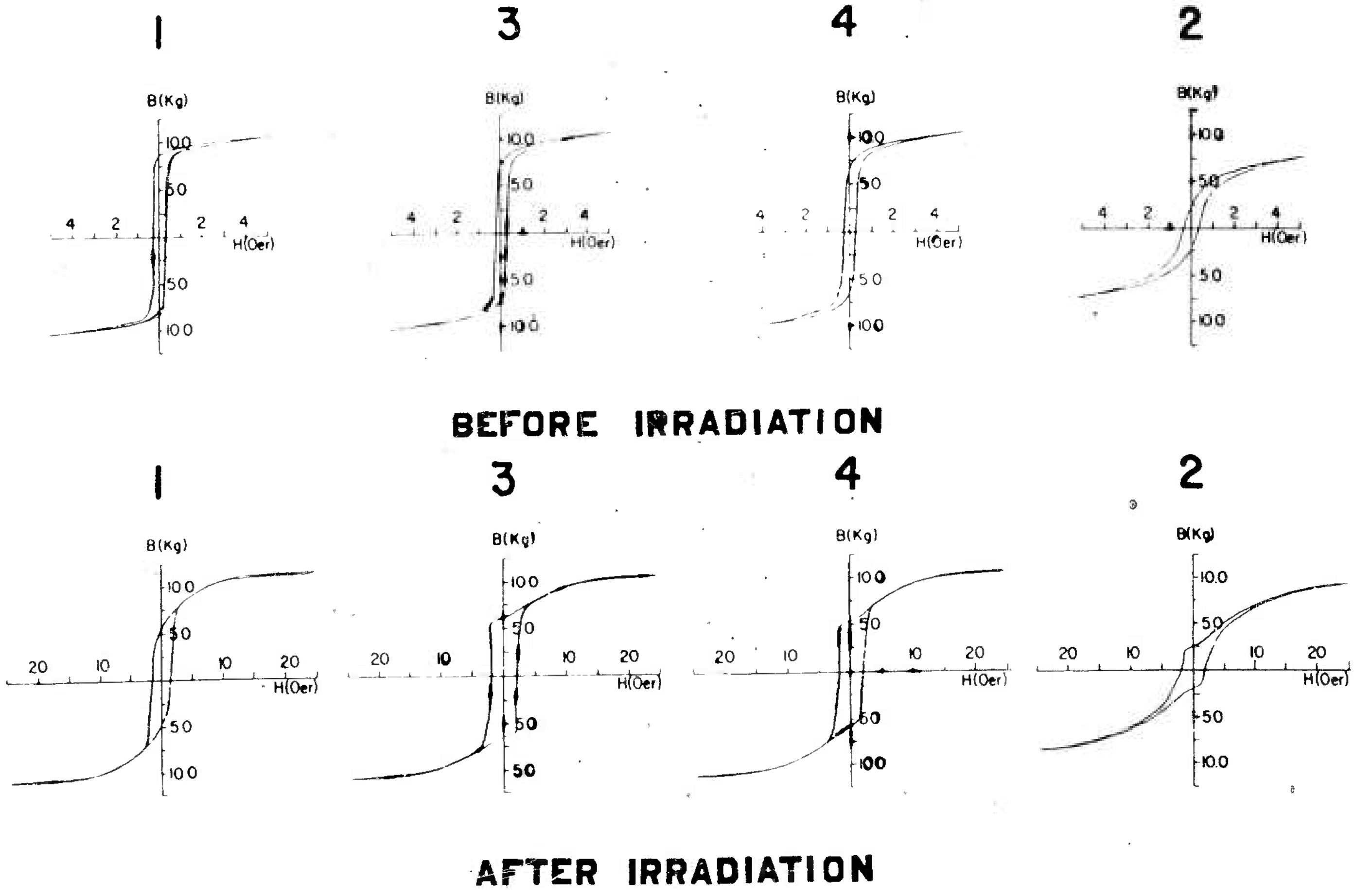


Figure 11 - Variation of the effect of irradiation with partial ordering on the 60 cycle hysteresis loops of 48% Ni - 52% Fe.

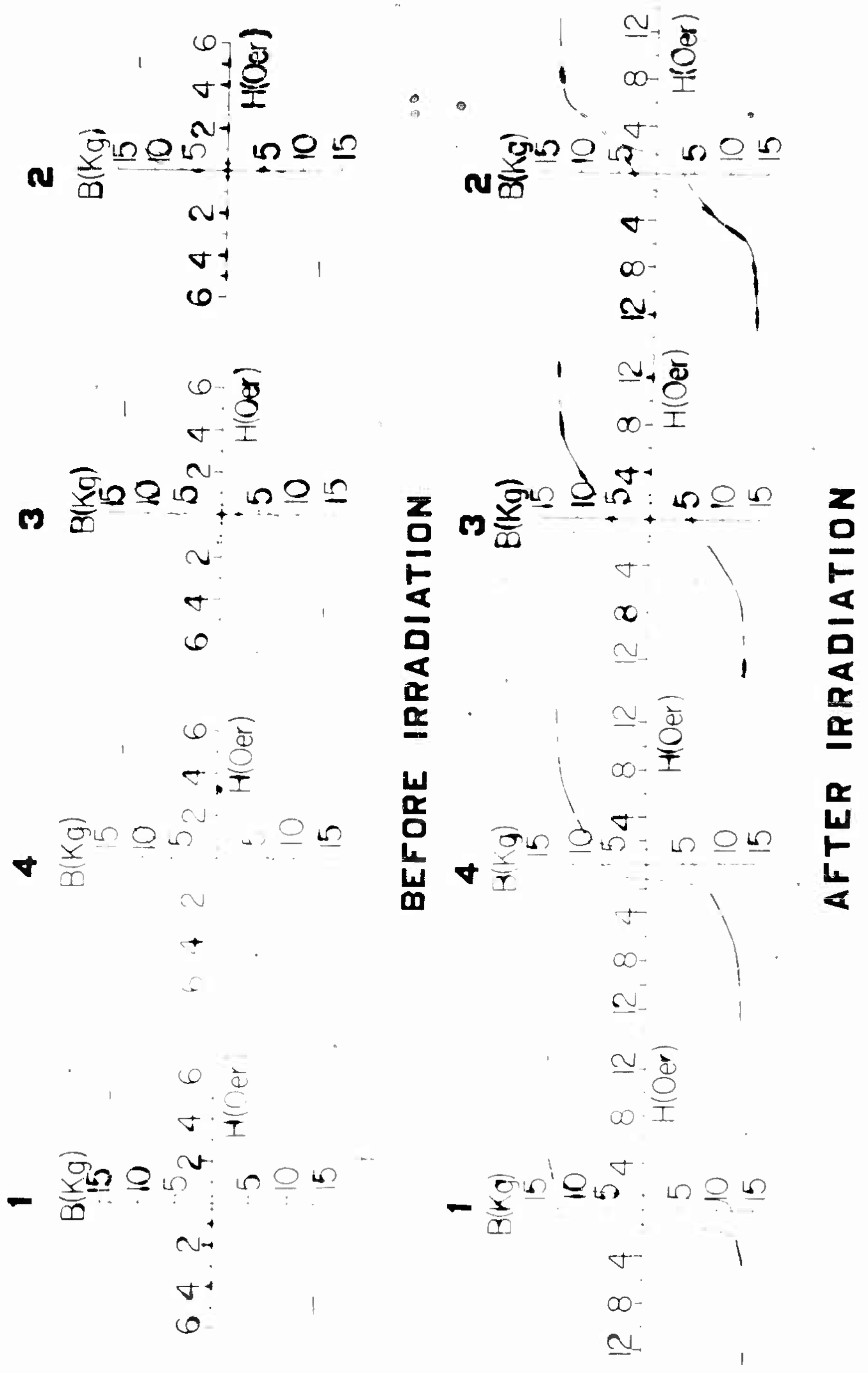


Figure 12 - Variation of the effect of irradiation with partial ordering on the 60 cycle hysteresis loops of square loop 50% Ni - 50% Fe.

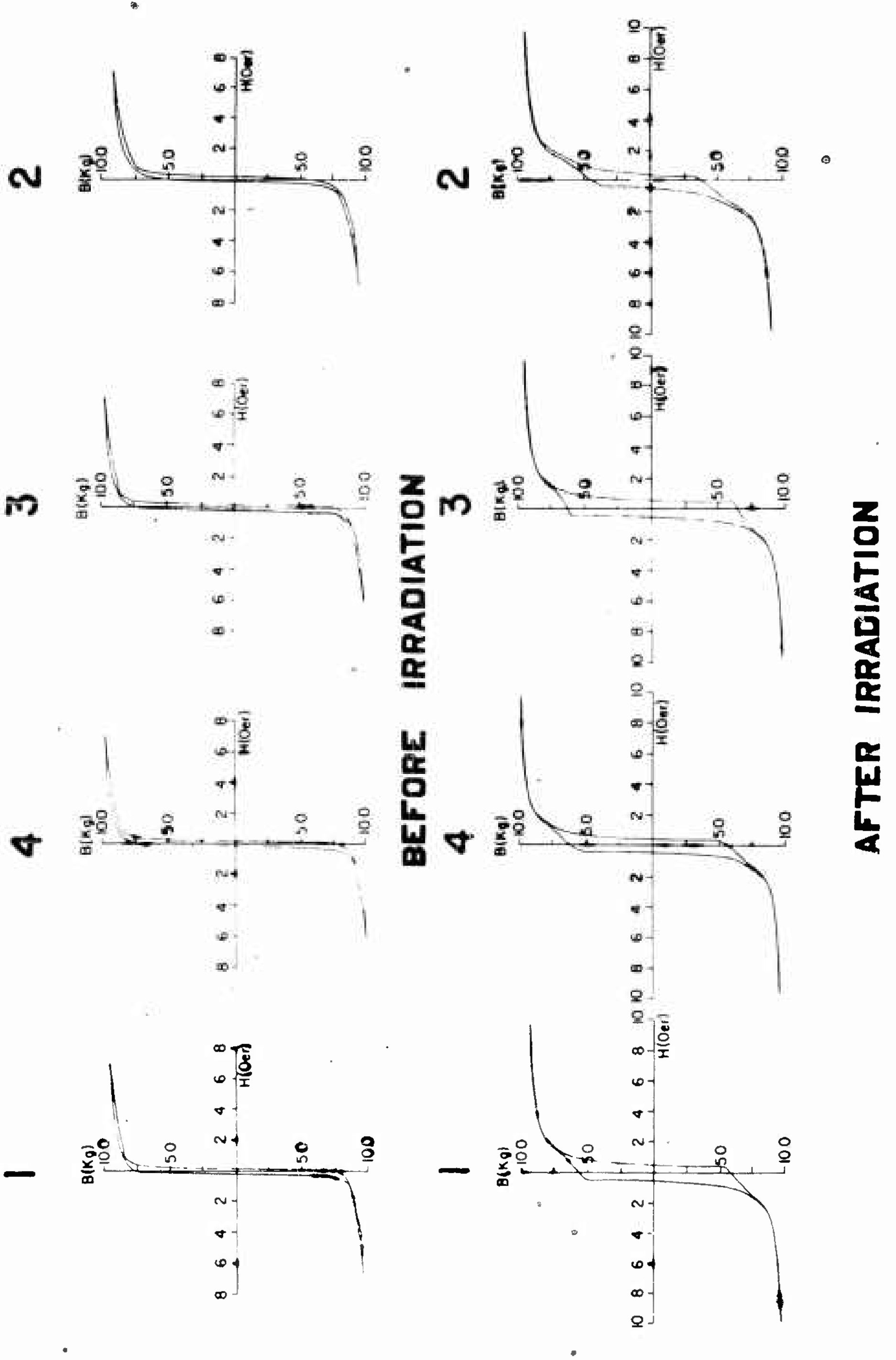


Figure 13 - Variation of the effect of irradiation with partial ordering on the 60 cycle hysteresis loops of 30% Mo - 47% Ni - 50% Fe.

found. In these cases, both H_C and B_R tended to larger values from left to right. If it is assumed that the minimum in the B_R and the maximum in H_C occur at slightly different degrees of order, the trend noted would indicate that we have passed through the critical stage of ordering characteristic of constricted loops, and into long-range order as a result of the neutron irradiation. The lower-right-hand curve in Fig. 14 is of the normal type and its position in the sequence seems to indicate that long-range ordering has occurred in this case.

The main result of the above experiments is the indication of the similarity between thermal and radiation-induced ordering. In order to substantiate further this conclusion, the following experiment was performed. Two untreated samples of an alloy of 48% Ni-52% Fe (NiFe type) were irradiated to approximately the same total integrated neutron flux. During the irradiation, the temperature of sample 1 was kept at 190°C while sample 2 was kept at 60°C. The result of this experiment is shown in Fig. 17. Prior to irradiation both samples exhibited similar hysteresis loops shown by the curve at the top, while the post-irradiation curves of samples 1 and 2 are shown below. The post-irradiation curves clearly indicate that the coercive force of sample 2 is smaller than that of sample 1 (note the change of H scale in sample 1), and the remanence of sample 2 is larger than that of sample 1. Again we would deduce from these results that sample 1 has been ordered to a greater extent than sample 2. If ordering is the sole cause of the magnetic changes due to irradiation, these measurements show that the radiation-induced ordering is enhanced by elevating the temperature. This would indicate that radiation-induced ordering may take place as a result of diffusion of vacancies which are in turn created by the neutron irradiation. This lends credence to the notion of the similarity between radiation-induced ordering and thermally-induced ordering in that both seem to result from increased atomic mobility.

To test further the similarity between thermally-induced ordering and radiation-induced ordering, another experiment was carried out in which the samples were irradiated in the presence of a saturating magnetic field. The samples used were again commercially available toroids of mainly iron-nickel alloys. Two samples of each material were wound with appropriate B and H coils and

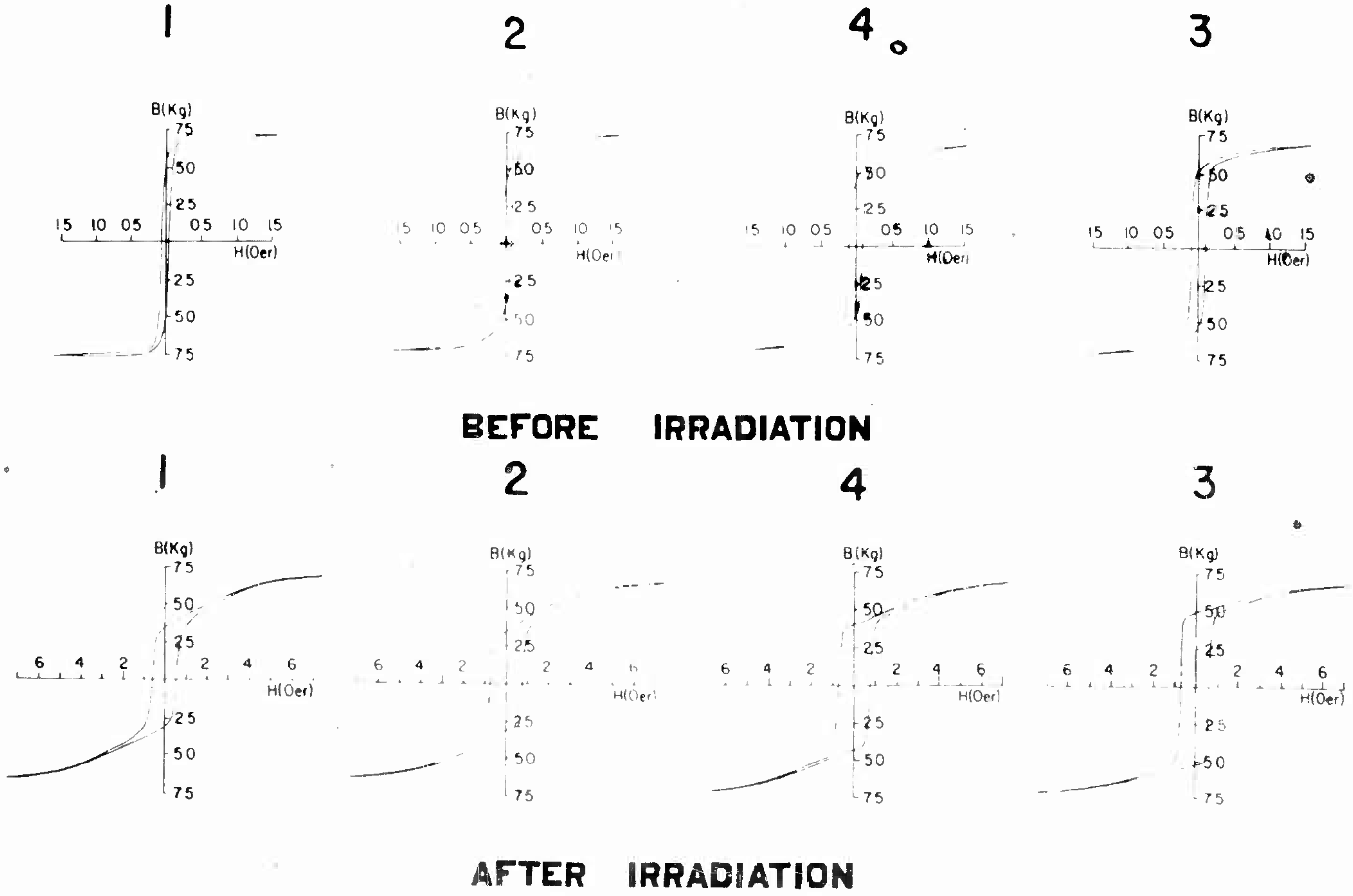


Figure 14 - Variation of the effect of irradiation with partial ordering on the 60 cycle hysteresis loops of Supermalloy.

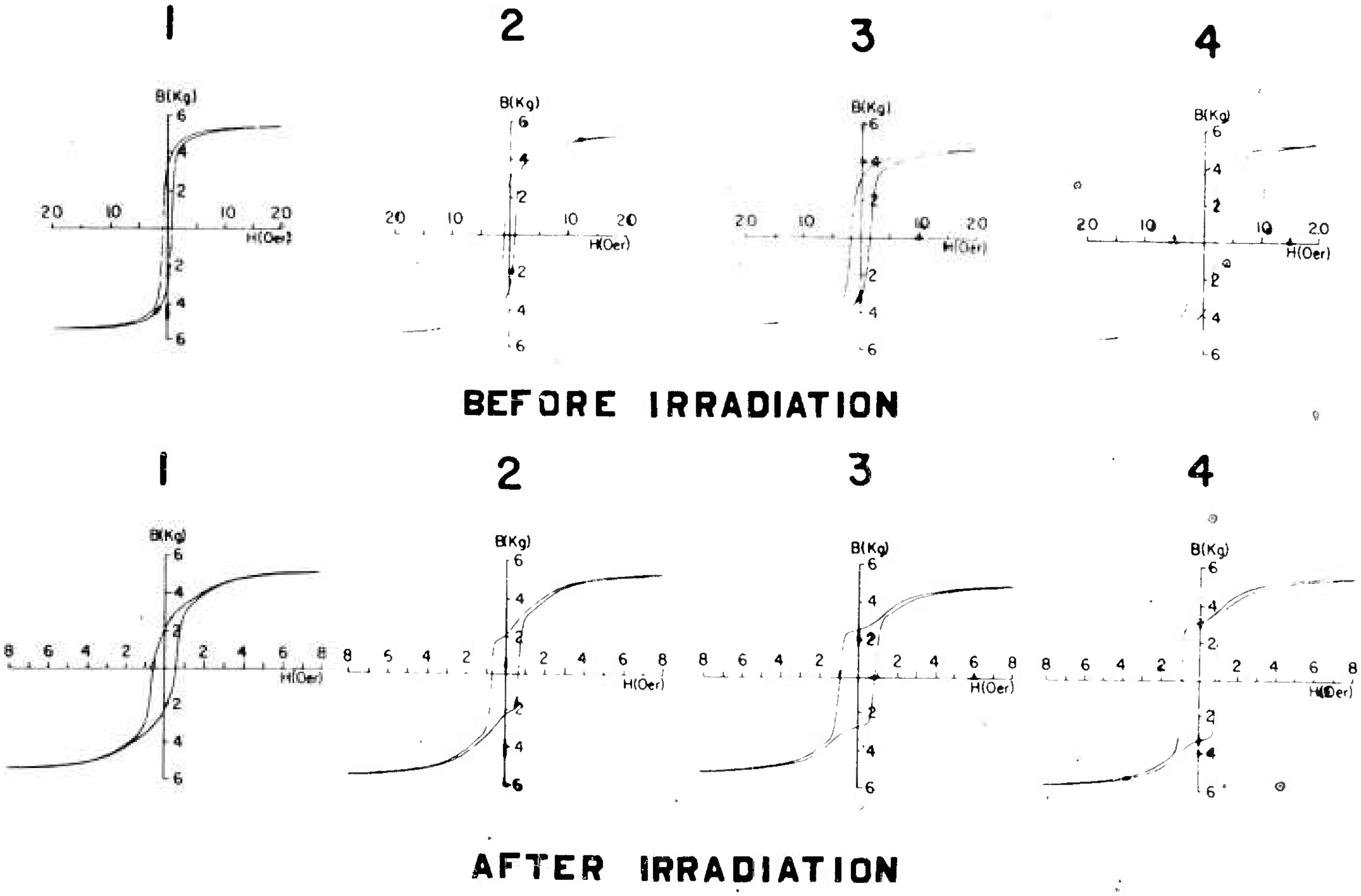


Figure 15 - Variation of the effect of irradiation with partial ordering on the 60 cycle hysteresis loops of Mumetal.

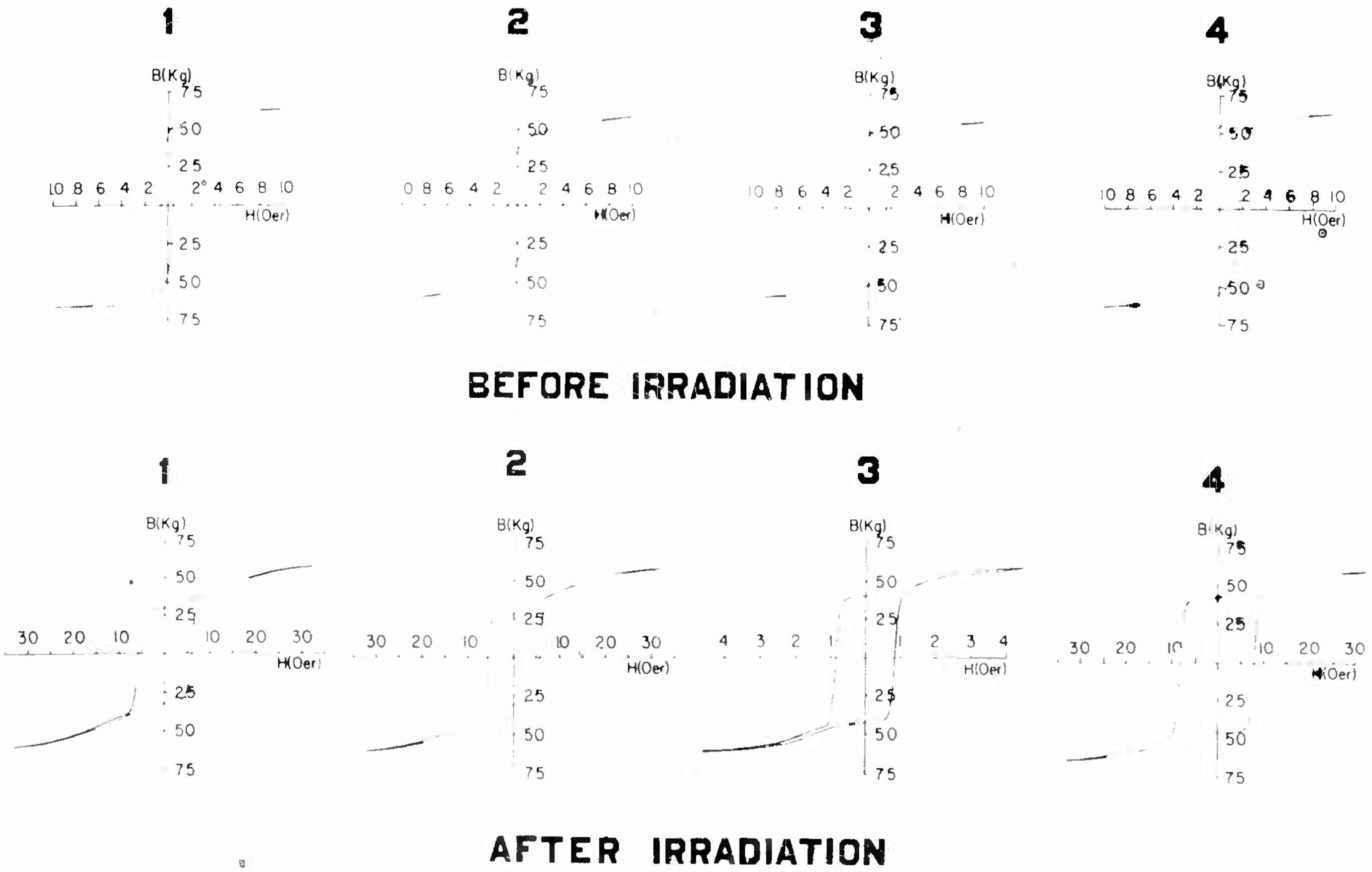


Figure 16 - Variation of the effect of irradiation with partial ordering on the 60 cycle hysteresis loops of Permalloy.

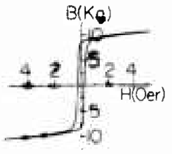
installed on the same sample holder. The samples were irradiated at the Brookhaven Reactor for two weeks. The total integrated fluxes for these samples were

ϕ fast = 1.25×10^{16} to 3.0×10^{16} nvt
ϕ epi = 2.6×10^{17} nvt
ϕ thermal = 5.9×10^{18} nvt,

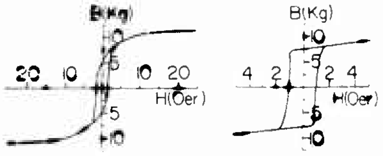
while the temperature during irradiation varied between 80° and 120°C. During irradiation a saturating magnetizing dc current was passed through the H coil of one sample of each pair. The field produced was, of course, in the same direction as the field applied during hysteresis loop measurements. After the irradiation, the hysteresis loops of each sample were again taken.

In Figure 18 is shown the pre- and post-irradiation hysteresis loops of permalloy and in Fig. 19 the loops for samples of Mumetal. The pre-irradiation loops are marked a, while the post-irradiation loops are the curves marked b and c. Loop b is the hysteresis loop of the sample irradiated in the absence of a magnetic field, while loop c is that for the sample irradiated in the presence of a saturating magnetic field applied to the toroid. In both cases loop b is similar to results previously observed, i.e., decreased remanence, increased coercive force, and decreased permeability. (Note that to saturate the sample a field several times that of the pre-irradiation saturating field is required.) In loop c a strikingly different effect was observed; namely, square loop characteristics were obtained with both the coercive force and remanence increasing.

In addition to aligning the magnetization vectors of the individual domains by annealing in a magnetic field, it is possible in some alloys to obtain partial alignment by a combination of mechanical and thermal treatments. 50% Fe-50% Ni is an alloy which responds to such treatment. In Fig. 20 is shown the dependence of the irradiation-modified hysteresis loop upon the domain distribution present during the irradiation. The sample whose pre- and post-irradiation hysteresis loops are shown on the left has been mechanically and thermally treated to have rectangular loop characteristics, i.e., preponderance of domains having aligned easy directions of magnetization. The hysteresis loops on the right are of a sample of the



BEFORE IRRADIATION



1 **2**

AFTER IRRADIATION

Figure 17 - Effect of temperature during irradiation on the 60 cycle hysteresis loops of 48% Ni - 52% Fe. T_1 = 190°C; T_2 = 60°C.

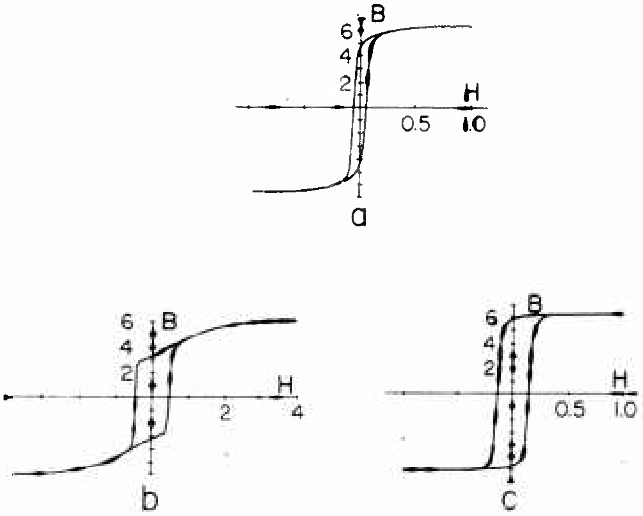


Figure 18 - The effect of neutron irradiation on the 60 cycle hysteresis loops of permalloy. (a) Pre-irradiation hysteresis loops. (b) Post-irradiation hysteresis loop (no field applied during irradiation). (c) Post-irradiation hysteresis loop (saturating magnetic field applied during irradiation). B in kilogauss, H in oersteds.

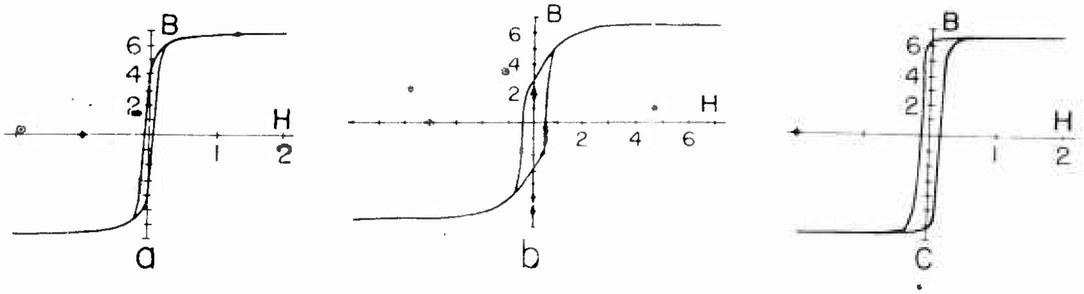
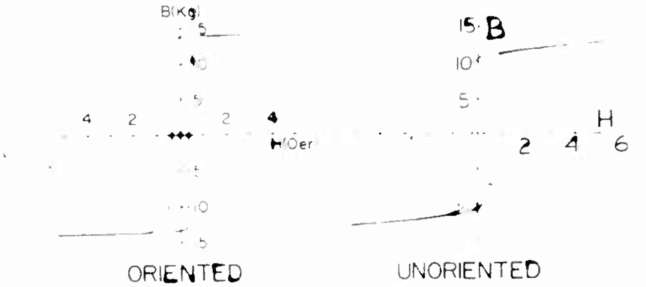
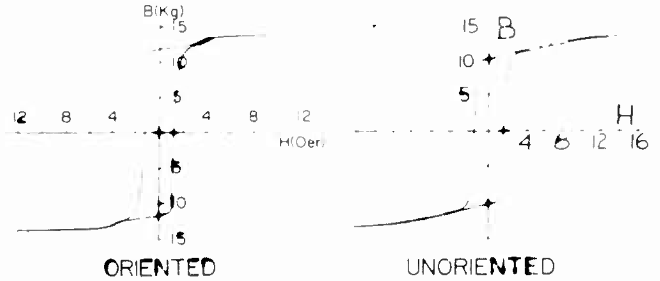


Figure 19 - The effect of neutron irradiation on the 60 cycle hysteresis loops of mumetal. (a) Pre-irradiation hysteresis loop. (b) Post-irradiation hysteresis loop(no field applied during irradiation). (c) Post-irratiation hysteresis loop (saturation magnetic field applied during irradiation). B in kilogauss, H in oersteds.



BEFORE IRRADIATION



AFTER IRRADIATION

Figure 20 - The dependence of the shape of the post-irradiation hysteresis loop upon the domain distribution present during irradiation for a 50% Ni - 50% Fe alloy.

same material in which the domains are oriented at random. Both samples were irradiated in the absence of an external magnetic field. The larger value of B_R and the strong constriction in the post-irradiation hysteresis loop of the sample on the left reflects the alignment of the domains during the irradiation. The mechanical alignment is similar to the magnetic alignment resulting from an applied magnetic field during irradiation except that the alignment is not as complete. After the irradiation, because of the stabilization of the domain direction by directional ordering, only a small field is necessary to saturate the majority of the domains. Much larger fields are required to rotate the magnetization vectors of the other domains into the field direction. This can be clearly seen in the sequence of post-irradiation hysteresis loops of a mechanically oriented sample of 50% Ni-50% Fe shown in Fig. 21. As the maximum field is increased a hysteresis loop is obtained that appears to be saturated and rectangular. In the figure this loop is that of $H_{max} = 1.23$ oer. The domains thus far aligned are those which were aligned in this field direction during the irradiation by the mechanical treatment prior to the irradiation. Further increasing the maximum field now aligns the domains whose magnetization vectors were unoriented during the irradiation and were consequently stabilized in directions out of the measuring field direction. A maximum applied field is finally reached for which true saturation occurs. The ratio of B_R to B_S is in fact a measure of the fraction of the total sample volume in which the domains were aligned in the measuring field direction during the irradiation. An adaptation of this effect can be used as a method of obtaining information relating to domain distribution present in metals and alloys.

All of the results that have been presented are consistent with the suggestion that neutron irradiation causes ordering in Ni-Fe alloys. The modified hysteresis loops reflect a change in the uniaxial anisotropy of the state of final order. In the case of samples irradiated for short times in the absence of an applied field, the resulting directional short-range ordering causes constricted hysteresis loops, while for samples irradiated in a magnetic field the resulting directional ordering causes square hysteresis loops.

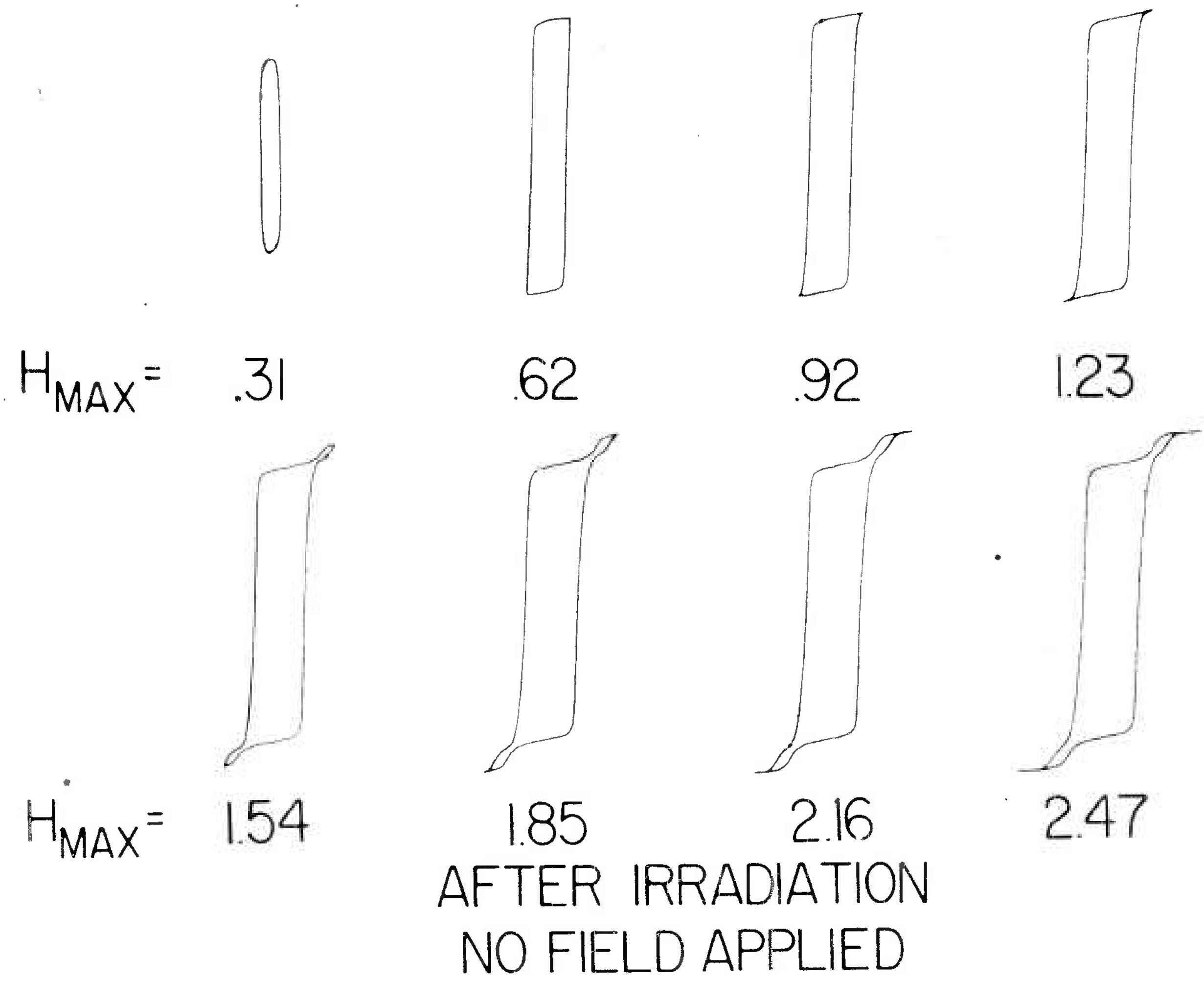


Figure 21 - Sequence of minor post-irradiation hysteresis loops of an oriented 50% Ni - 50% Fe alloy illustrating the stabliization of magnetization directions resulting from irradiation induced directional diffusion

There is, however, one significant difference between radiation-induced directional ordering and the magnetic annealing treatment. In the latter case, the samples must be heated to temperatures of the order of 600°C, which approaches the Curie temperatures of the alloys. Consequently the internal magnetic field, which imposes directionality to the diffusion, is relatively small. In our experiments, the diffusion is enhanced not by temperature but rather by the increased density of vacancies resulting from the neutron irradiation. This then results in much larger effects, since the sample temperatures during irradiation are low and, consequently, the internal magnetic fields which influence diffusion are larger than those present during the thermal treatment.

II. FERRITES

Experiments have been also carried out on the effect of neutron irradiation on the Curie temperature, hysteresis loop parameters, and magnetic moments of a series of pure and mixed ferrites. The mixed ferrites were commercially obtained for the sake of expediency. Very serious disadvantages exist with this approach, since the exact chemical composition and manufacturing procedures are complicated and are usually trade secrets. The samples are generally commercially prepared by mixing and compressing oxide powders and sintering at elevated temperatures. Such parameters as the coercive force and permeability of the resultant material depend not only upon the chemical composition, but also on various factors connected with the sintering process, such as porosity and the size and shape of the pores and of the crystallites. No information of these sintering parameters were available, and it must be assumed that they were unaffected by the irradiation process.

The mechanisms which would suggest that the intrinsic magnetic properties of certain ferrites are sensitive to neutron irradiation are identical to those pertaining to the metal alloys. Strains introduced in the vicinity of radiation-induced defects would also be expected to decrease the permeability of ferrites. However the magnitude of such changes should be considerably smaller than in the case of metals because of the porous nature of the commercial ferrite material. Also, constricted loops related to irradiation-induced directional ordering would be expected, since many ferrites are known to respond to magnetic-anneal treatments.

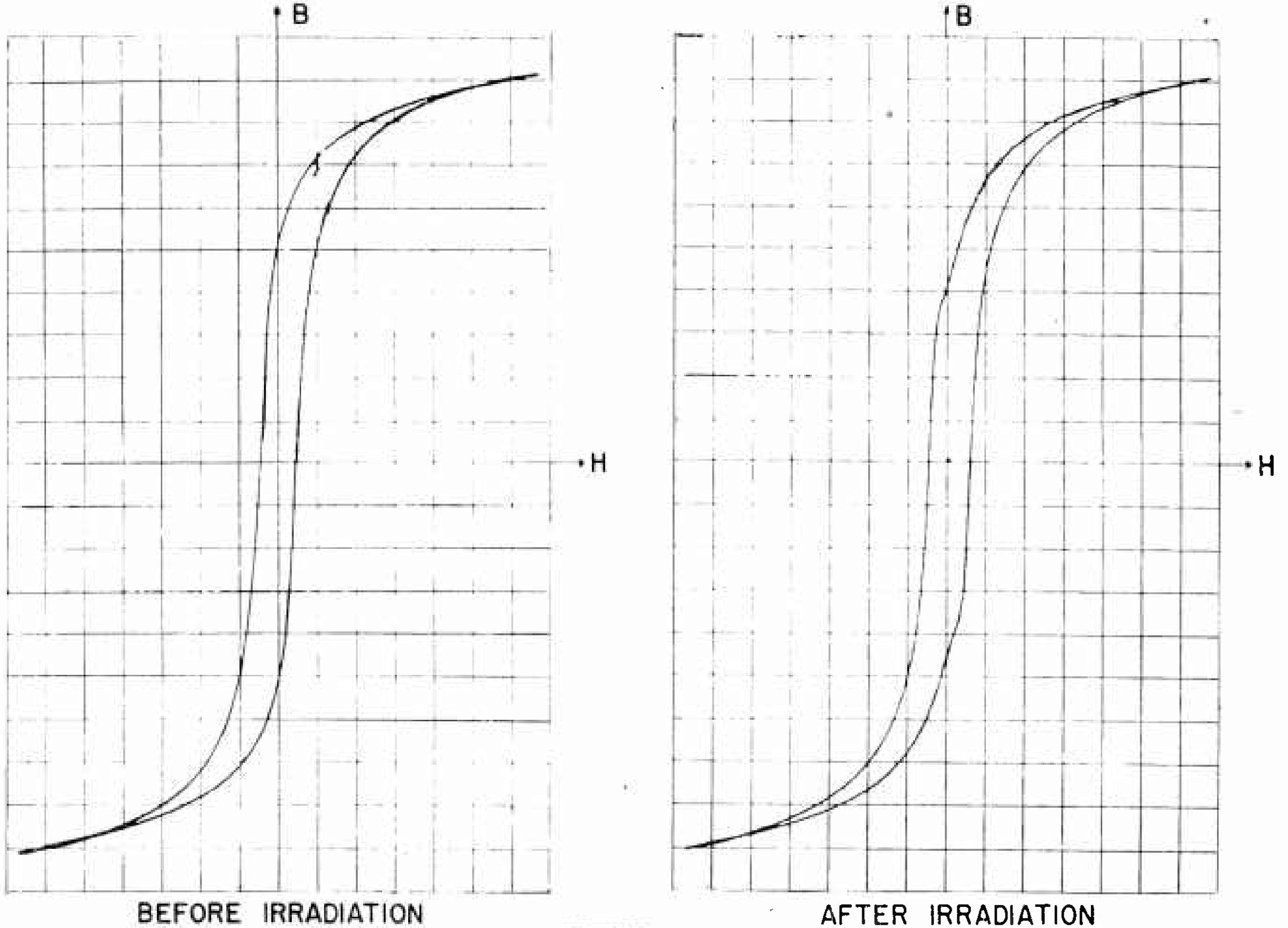
In addition, one might expect that the magnetic moment of ferrites should be sensitive to neutron irradiation. Ferrites in general have the spinel structure, and the magnetic moment is determined by the balancing of the moments of the ions located, respectively, upon the octahedral and tetrahedral sites. Both radiation-induced replacements of ions on these two types of sites and radiation-induced vacancies would be expected to cause sizable changes of the magnetic moments.

RESULTS

The resultant changes in B_R and H_C for a variety of commercial ferrites are compiled in Table 1. If less than a 10% change in either of these parameters is arbitrarily considered to be insignificant, then only samples II, IV, and VI may be assumed to have been unaffected. All other ferrites showed larger changes.

In addition constrictions were found in the post-irradiation hysteresis loops of the ferrites shown in Figs. 22 and 23. These samples are complex ferrites containing mixtures of oxides of Mn, Zn, and Fe (Fig. 22) and Ni, Zn, Cu, and Fe (Fig. 23). It is quite likely that the constrictions in the loops are related to the irradiation-induced directional ordering similar to that found in the Fe-Ni alloys.

Measurements were also made on the effect of irradiation on the Curie temperature of ferrites. An extremely simple method was employed in which a heated magnetized sample was located in close proximity to a sensitive ship's compass. As the temperature of the sample was raised through the Curie temperature, the compass heading changed, approaching the magnetic north direction. Pre-irradiation Curie temperature measurements were made; then the samples were irradiated at the Brookhaven Reactor for four weeks and post-irradiation Curie temperature measurements were obtained. During the irradiation, the average temperature was 50°C and the average total integrated flux was 6.3×10^{18} nvt. The results of these measurements are shown in Table 2. Samples 1 through 14 are soft magnetic materials while

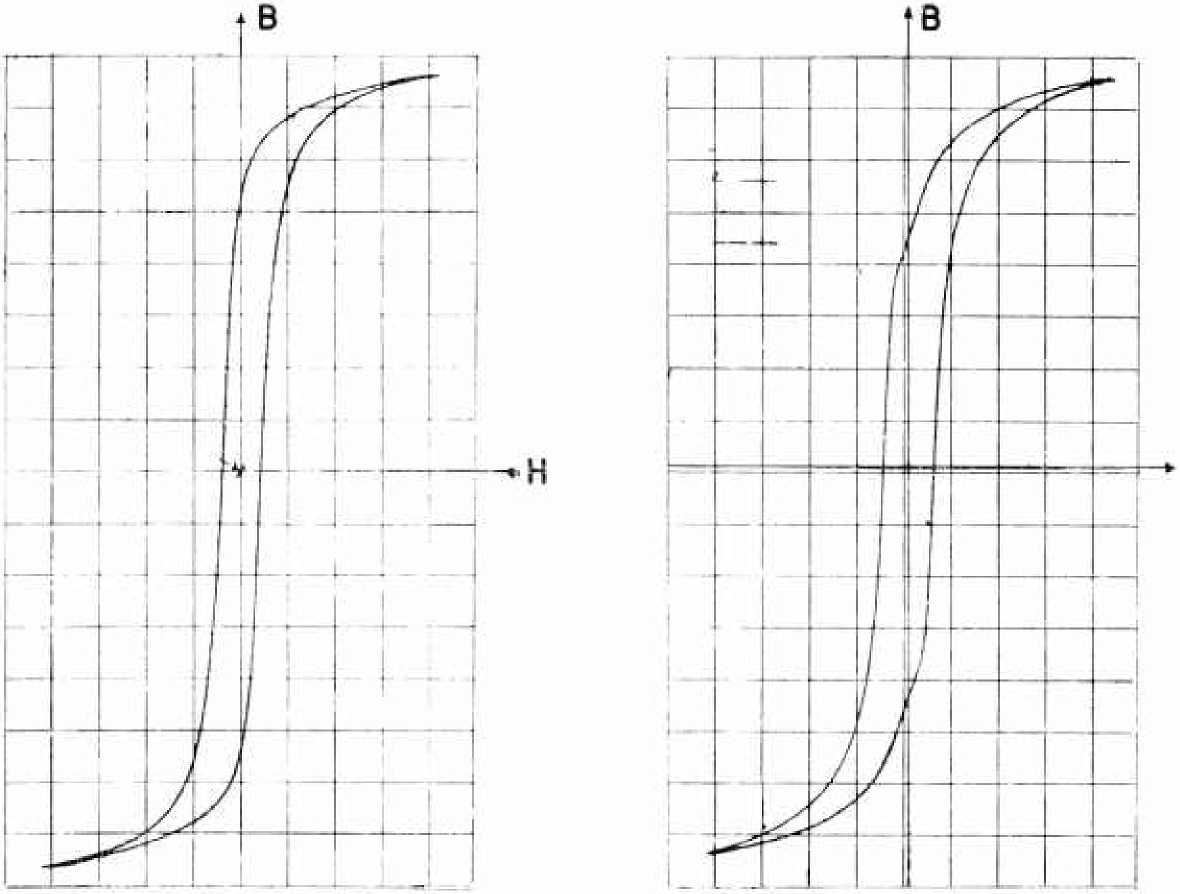


BEFORE IRRADIATION

AFTER IRRADIATION

VIII

Figure 22 - Effect of irradiation on the 60 cycle hysteresis loop of a commercial manganese zinc ferrite illustrating constriction



BEFORE IRRADIATION

AFTER IRRADIATION

IX

Figure 23 - Effect of irradiation on the 60 cycle hysteresis loop of a commercial nickel zinc copper ferrite illustrating constriction

Table 1

Effect of Irradiation on the Coercive Force and Remanence for Various Ferrites

Sample No.	Oxide Constituents	Applied H Max. (oer)	Δ H_C	Δ B_R
XII	NiO, Fe_2O_3	14	- 8%	-36%
X	NiO, ZnO, Fe_2O_3	7	+ 22%	- 9%
IV	NiO, ZnO, MnO, Fe_2O_3	20	0	+ 5%
II	NiO, ZnO, MnO, Fe_2O_3	25	0	+ 7%
V	NiO, ZnO, MnO, MgO, Fe_2O_3	14	+106%	-15%
III	NiO, ZnO, MnO, MgO, Fe_2O_3	6	+ 14%	-12%
IX	NiO, ZnO, CuO, Fe_2O_3	3	+ 30%	-17%
VII	NiO, ZnO, CuO, Fe_2O_3	25	+ 21%	-15%
XI	NiO, ZnO, MnO, MgO, Fe_2O_3	9	+ 40%	- 6%
XIII	MgO, MnO, Fe_2O_3	25	+ 48%	- 1%
VIII	MnO, ZnO, Fe_2O_3	5	+ 6%	-20%
VI	NiO, ZnO, MnO, MgO CuO, Fe_2O_3	2	+ 5%	0

sample 15 is a permanent magnet material. In general it was found that no significant changes were found in the Curie temperatures of the ferrite samples examined. If any redistributions of ions on the two types of sites occurred as a result of the irradiation, it may be inferred that the Curie temperature is not very sensitive to such a redistribution.

Perhaps the most significant part of the ferrite investigation is the determination of the effect of neutron irradiation upon the magnetic moment (6). Two types of ferrites were chosen: γFe_2O_3 and Fe_3O_4. It was not possible to obtain absolutely pure material and the analyses of the samples used are as follows:

γFe_2O_3		Fe_3O_4	
Fe_2O_3	90.90%	Fe_3O_4	77.22%
Fe_3O_4	6.52	Fe_2O_3	21.76
SiO_2	0.53	SiO_2	0.22
SnO_2	0.07	CaO	1.00
Al_2O_3	1.63	MgO	0.20
H_2O	0.65		

The samples were irradiated at the Brookhaven Graphite Reactor to a total integrated flux of 6.2×10^{18} nvt and pre- and post-irradiation moment measurements were obtained. Figure 24 shows plots of magnetization as a function of magnetic field for γFe_2O_3 and Fe_3O_4 before and after irradiation. It is seen that the absolute saturation magnetization for γFe_2O_3 is decreased by about 3%, while for Fe_3O_4 the decrease is 15%. This reduction can be related to an irradiation-induced redistribution of magnetic ions on opposing sites, or to an irradiation-induced oxidation process whereby the ferrimagnetic oxides are converted to antiferromagnetic αFe_2O_3. The change found for the γFe_2O_3 is probably a result of ion redistribution, while that for the Fe_3O_4 is probably primarily a result of oxidation. This latter possibility has been somewhat substantiated by neutron diffraction measurements made on the irradiated Fe_3O_4. Similar investigations were made of the effects of neutron irradiation on the magnetization of yttrium iron garnet, a commercial manganese ferrite, and a commercial nickel ferrite. No appreciable changes were found in the saturation magnetization in any of these materials due to the irradiation.

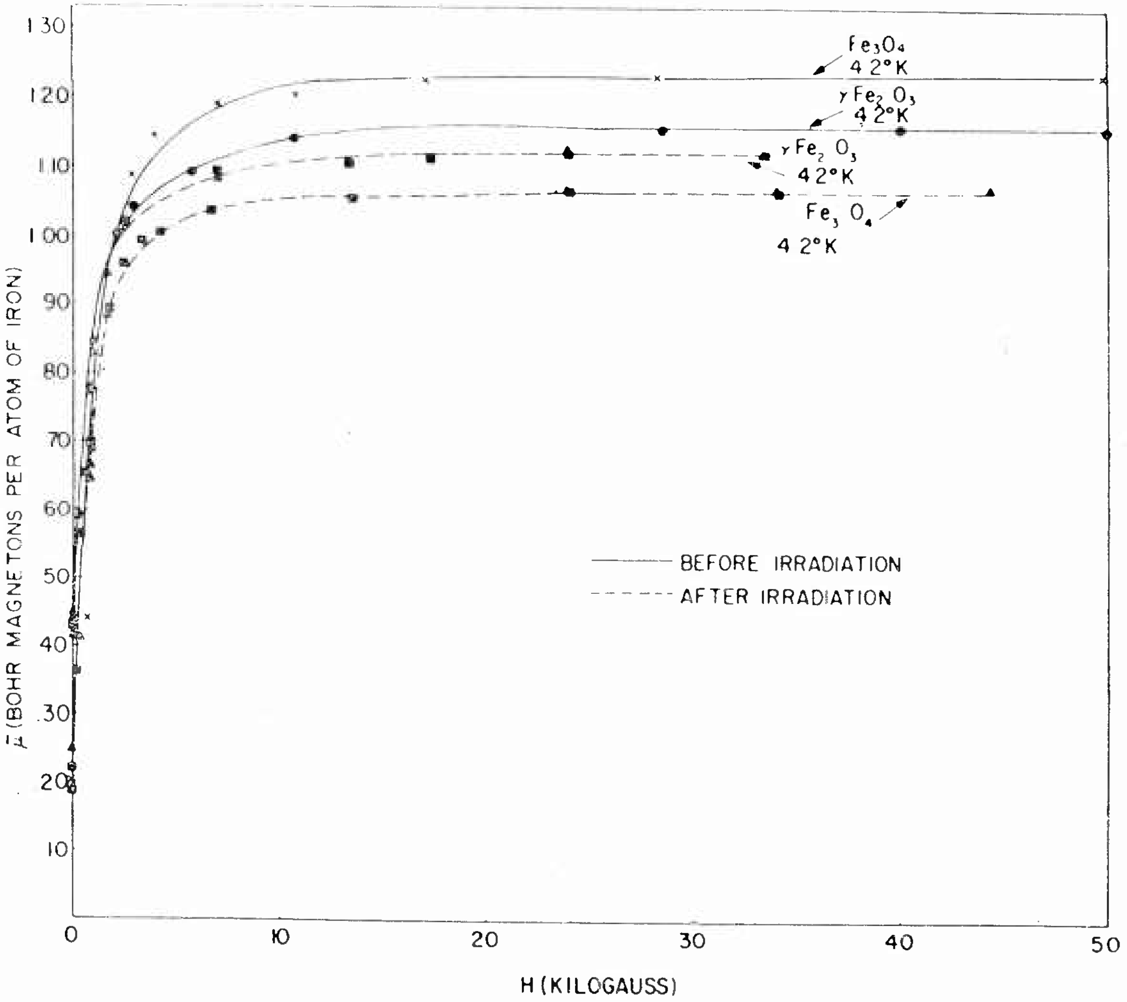


Figure 24 - Plot of average magnetization ($\bar{\mu}$) per atom of iron as a function of magnetic field at 4.2°K for γ-Fe_2O_3 and Fe_3O_4. The continuous curves represent magnetization before irradiation, while the broken lines represent magnetization after irradiation

Table 2

Curie Temperature of Soft Ferrite

Sample No.	Oxide Constituents	Curie Temperature (°C)	
		Before	After
1	NiO, MnO, MgO, ZnO, Fe_2O_3	341	337
2	MgO, MnO, Fe_2O_3	267	263
3	MgO, MnO, Fe_2O_3	271	275
4	NiO, Fe_2O_3	258	256
5	NiO, MnO, ZnO, Fe_2O_3	462	460
6	NiO, MgO, MnO, ZnO, Fe_2O_3	291	287
7	MnO, ZnO, Fe_2O_3	176	177
8	NiO, ZnO, Fe_2O_3	154	154
9	NiO, MnO, MgO, ZnO, Fe_2O_3	252	249
10	NiO, MnO, ZnO, Fe_2O_3	368	368
11	NiO, ZnO, Fe_2O_3	73	79
12	NiO, ZnO, CuO, Fe_2O_3	159	161
13	NiO, MnO, ZnO, Fe_2O_3	373	368
14	NiO, MnO, MgO, ZnO, Fe_2O_3	215	214
15	Lead-Ferrite	446	445

ACKNOWLEDGMENTS

The results presented represent the combined efforts of many people, in particular Dr. E. I. Salkovitz, Mr. G. C. Bailey, Dr. G. S. Ansell, and Dr. W. E. Henry. I am also grateful to the Radiation Operations Section under the supervision of Mr. L. E. Steele for furnishing considerable assistance related to the irradiation and post-irradiation operations.

REFERENCES

1. Dienes, G.J., and Vineyard, G.H., "Radiation Effects in Solids," New York: Interscience, 1957

2. Salkovitz, E.I., Schindler, A.I., Sakiotis, N.G., and Ansell, G.S., "Proceedings of an International Conference on Solid State Physics in Electronics and Telecommunications," Brussels, 1958, M. Desirant and J.L. Michels, editors, Vol. 4, New York: Academic Press, 1960

3. Schindler, A.I., Salkovitz, E.I., and Ansell, G.S., J. Appl. Phys. 30:282S (1959)

4. Schindler, A.I., and Salkovitz, E.I., J. Appl. Phys. 31:245S (1960)

5. Salkovitz, E.I., Bailey, G.C., and Schindler, A.I., J. Appl. Phys. 29:1747 (1958)

6. Henry, W.E., and Salkovitz, E.I., J. Appl. Phys. 30:286S (1959)

7. See R.M. Bozorth, "Ferromagnetism," New York: Van Nostrand, 1951, p.624

8. Ibid., p.109

9. Kaya, S., Rev. Mod. Phys., 25:49 (1951)

10. Néel, L., J. phys. radium 15:225 (1954)

11. Taniguchi, S., Sci. Rept. RITU A-7:269 (1955); A-8:173 (1956)

UNCLASSIFIED

Naval Research Laboratory. Report 5686.
THE EFFECTS OF IRRADIATION ON MAGNETIC PROPERTIES OF ALLOYS AND FERRITES by A. I. Schindler. 37 pp. & figs., August 11, 1961.

Studies have been conducted on the effects of neutron irradiation on the magnetic characteristics of metal alloys and ferrites. The magnetic properties examined include the coercive force, remanence, shape of the hysteresis loop, Curie temperature, and magnetic moment. Only the Curie temperature was found to be relatively insensitive to neutron irradiation; all the other properties were modified by varying amounts. The radiation sensitivity of metal alloys and selected ferrites appears to be primarily related to the dependence of magnetic properties on the short-range ordering of the atoms along certain crystallographic

UNCLASSIFIED (Over)

1. Alloys – Magnetic properties – Effects of radiation
2. Ferrites – Magnetic properties – Effects of radiation

I. Schindler, A. I.

UNCLASSIFIED

Naval Research Laboratory. Report 5686.
THE EFFECTS OF IRRADIATION ON MAGNETIC PROPERTIES OF ALLOYS AND FERRITES by A. I. Schindler. 37 pp. & figs., August 11, 1961.

Studies have been conducted on the effects of neutron irradiation on the magnetic characteristics of metal alloys and ferrites. The magnetic properties examined include the coercive force, remanence, shape of the hysteresis loop, Curie temperature, and magnetic moment. Only the Curie temperature was found to be relatively insensitive to neutron irradiation; all the other properties were modified by varying amounts. The radiation sensitivity of metal alloys and selected ferrites appears to be primarily related to the dependence of magnetic properties on the short-range ordering of the atoms along certain crystallographic

UNCLASSIFIED (Over)

1. Alloys – Magnetic properties – Effects of radiation
2. Ferrites – Magnetic properties – Effects of radiation

I. Schindler, A. I.

UNCLASSIFIED

Naval Research Laboratory. Report 5686.
THE EFFECTS OF IRRADIATION ON MAGNETIC PROPERTIES OF ALLOYS AND FERRITES by A. I. Schindler. 37 pp. & figs., August 11, 1961.

Studies have been conducted on the effects of neutron irradiation on the magnetic characteristics of metal alloys and ferrites. The magnetic properties examined include the coercive force, remanence, shape of the hysteresis loop, Curie temperature, and magnetic moment. Only the Curie temperature was found to be relatively insensitive to neutron irradiation; all the other properties were modified by varying amounts. The radiation sensitivity of metal alloys and selected ferrites appears to be primarily related to the dependence of magnetic properties on the short-range ordering of the atoms along certain crystallographic

UNCLASSIFIED (Over)

1. Alloys – Magnetic properties – Effects of radiation
2. Ferrites – Magnetic properties – Effects of radiation

I. Schindler, A. I.

UNCLASSIFIED

Naval Research Laboratory. Report 5686.
THE EFFECTS OF IRRADIATION ON MAGNETIC PROPERTIES OF ALLOYS AND FERRITES by A. I. Schindler. 37 pp. & figs., August 11, 1961.

Studies have been conducted on the effects of neutron irradiation on the magnetic characteristics of metal alloys and ferrites. The magnetic properties examined include the coercive force, remanence, shape of the hysteresis loop, Curie temperature, and magnetic moment. Only the Curie temperature was found to be relatively insensitive to neutron irradiation; all the other properties were modified by varying amounts. The radiation sensitivity of metal alloys and selected ferrites appears to be primarily related to the dependence of magnetic properties on the short-range ordering of the atoms along certain crystallographic

UNCLASSIFIED (Over)

1. Alloys – Magnetic properties – Effects of radiation
2. Ferrites – Magnetic properties – Effects of radiation

I. Schindler, A. I.

UNCLASSIFIED

directions. Materials which can be easily ordered show the greatest sensitivity. It has been found that the application of a magnetic field during irradiation can cause directional ordering to take place which in many cases produces more desirable magnetic characteristics. The studies of the ferrites are more complicated since the effects appear to be related to atomic displacements as well as possible radiation induced oxidation. This has been deduced from magnetic measurements of irradiated samples coupled with neutron diffraction data of the same material.

UNCLASSIFIED

UNCLASSIFIED

directions. Materials which can be easily ordered show the greatest sensitivity. It has been found that the application of a magnetic field during irradiation can cause directional ordering to take place which in many cases produces more desirable magnetic characteristics. The studies of the ferrites are more complicated since the effects appear to be related to atomic displacements as well as possible radiation induced oxidation. This has been deduced from magnetic measurements of irradiated samples coupled with neutron diffraction data of the same material.

UNCLASSIFIED

UNCLASSIFIED

directions. Materials which can be easily ordered show the greatest sensitivity. It has been found that the application of a magnetic field during irradiation can cause directional ordering to take place which in many cases produces more desirable magnetic characteristics. The studies of the ferrites are more complicated since the effects appear to be related to atomic displacements as well as possible radiation induced oxidation. This has been deduced from magnetic measurements of irradiated samples coupled with neutron diffraction data of the same material.

UNCLASSIFIED

UNCLASSIFIED

directions. Materials which can be easily ordered show the greatest sensitivity. It has been found that the application of a magnetic field during irradiation can cause directional ordering to take place which in many cases produces more desirable magnetic characteristics. The studies of the ferrites are more complicated since the effects appear to be related to atomic displacements as well as possible radiation induced oxidation. This has been deduced from magnetic measurements of irradiated samples coupled with neutron diffraction data of the same material.

UNCLASSIFIED