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**PRESSURE VESSEL STEEL EMBRITTLEMENT
MONITORING BY
MAGNETIC PROPERTIES MEASUREMENTS ***

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ABSTRACT

The magnetic properties of specimens of one heat of A533B nuclear pressure vessel grade steel have been examined in the as-received condition and after neutron irradiation to various fluence levels up to $4 \times 10^{18} \text{ cm}^{-2}$ ($E > 0.1 \text{ MeV}$) in the University of Illinois Advanced TRIGA reactor core at two temperatures, approximately 120 and 260°C. The effect of some heat treatments was also investigated. The magnetic properties were measured by an automated hysteresis curve tracing method using a miniature transformer which incorporated the specimens in its core. Changes in magnetic hysteresis energy loss were correlated with neutron fluence in the case of certain irradiated specimens, and with microhardness measurements in the case of heat treated specimens. At the higher irradiation temperatures, no significant changes in either the magnetic hysteresis properties or the microhardness were noted for the present fluences. The relationship between the observed magnetic properties response and irradiation-induced embrittlement is discussed.

I. INTRODUCTION

The potential for the use of magnetic properties as an indication of the degree of accumulated radiation-induced damage in ferromagnetic materials is virtually unexplored. While past irradiation damage studies [1,2] have established that some features of the magnetic hysteresis response of ferromagnetic materials are effected by irradiation, they did not attempt to correlate the observed magnetic properties changes with other materials properties changes. The understanding of irradiation-induced properties changes has progressed markedly since those studies, and a much better understanding of the microstructural response of materials to irradiation now exists. Furthermore, the earlier work centered on a few select materials, none of which are directly relevant to structural applications. In the present work, nuclear pressure vessel steels are studied to determine the effects of irradiation on both mechanical and magnetic properties. Radiation-induced magnetic properties changes in this class of materials have received scant attention [3-5], though there is obvious potential for the use of magnetic properties as an indication of bulk properties changes which would also influence mechanical performance.

Steels, including those grades employed for nuclear pressure vessels and support systems, are ferromagnetic. In fact, this ferromagnetism extends to temperature well in excess of those where they are employed for nuclear applications; the Curie temperature of Fe is 770°C. This may provide a means for recording and analyzing materials properties changes over a range of temperatures of interest to operating nuclear reactors. Furthermore, magnetic properties measurement techniques are relatively simple and amenable to automation, even in adverse environments, thus real-time monitoring may be possible.

The aim of the present work is to correlate changes in magnetic properties of nuclear pressure vessel steels with the onset and evolution of irradiation-induced embrittlement of those materials. As far as the authors are aware, there is no precedent for the application of magnetic properties such as remanence, coercivity, saturation magnetization or hysteresis loss for the monitoring of damage dosimetry and pressure vessel embrittlement. The basis for a correlation between magnetic and mechanical properties stems from the fact that the principal feature controlling magnetic hysteresis response is the pinning of magnetic domain walls. The microstructural features capable of pinning these walls are the same as those which influence the flow of dislocations. Thus the level of materials hardening should correlate directly to the energy to move magnetic domain walls and align magnetic domains.

While scant theory exists regarding the precise effects of fine microstructural features on magnetic hysteresis response, a few recent studies on hard magnetic materials show that magnetic remanence, at least, is dramatically affected by very low doses of neutrons. Cost and Brown [6-8] found that Fe-Nd-B and Co-Sm permanent magnets lose their field strength when subjected to (epithermal) neutron irradiation. The changes were very sensitive to dose, so sensitive that large changes in magnetic remanence were found after only 10^{15} or 10^{16} n/cm² ($E > 0.1$ MeV). No similar data are available for the response of low alloy steels, however, they should be amenable to similar magnetic properties measurements.

EXPERIMENTAL

Definitions

The magnetic hysteresis is a familiar property of ferromagnetic materials. Several features of the hysteresis response are possible means of judging the degree of domain wall pinning and the level of domain alignment under an applied magnetic field. In particular, the magnetic remanence, coercivity, saturation magnetization, and the hysteresis loss are readily quantifiable with standard magnetic properties measurement techniques. These are defined as follows (see Figure 1). Saturation magnetization, M_s , is the upper limit to the magnetization of the material that can be achieved by an externally applied field, H . The magnetic remanence, B_r , is level of magnetism remaining in the specimen after the applied field, H , is removed, that is, set to zero. The coercivity, H_c , is the further field which must be applied to remove the magnetic remanence, that is, set back to zero. The hysteresis loss is area of the hysteresis curve. It should be noted that it is often difficult to reach full magnetic saturation; values of B are found to increase with the increase of H even after the tips of the hysteresis loop are closed [9]. In the present work, it is found that only small changes in the hysteresis response are found by increasing the applied field beyond the point where the hysteresis loop closes at the tips even though the value of B continues to rise with H . Alternate definitions of effective or technical magnetic saturation have been proposed based on the width of the loop at $B = 0$, that is the coercivity, [10] or by an extrapolation of the closed part of the loop back to the $H = 0$ axis. [9]

Magnetic Properties Measurement Technique

A major point in the development of present measurement techniques is that magnetic properties are bulk properties and are amenable to measurement in very small

volumes of materials. In fact, the development of eddy currents during such measurement can be a major disadvantage to the use of large sample volumes. The small specimen sizes have additional advantages that only small volumes of material are activated during irradiation, and that self-shielding effects are minimized during irradiation. Low specimen activity allows magnetic property measurements to be performed after only short "cooling" periods, and also aids other handling processes.

While several specimen configurations are possible, the present techniques employs specimens in the shape of cylindrical pins with diameter 1.51 mm and length 12.8 mm. The specimen and yoke configuration for measurements is depicted in Figure 2, where the primary windings on the yoke and the secondary winding coil for properties measurement are also shown. The primary windings around the yoke, 40 turns, are to sustain the heavy current necessary to generate strong magnetic flux. The secondary loop coil, a plastic reel with 800 turns of fine copper wire, is designed with numerous turns of the fine wire to induce a large, more accurately measurable voltage from its interaction with the magnetic field generated in the specimen. The specimen is inserted through the yoke and reel arrangement for measurements. This specimen setup is linked between a primary driver circuit where the magnetic field is applied, and a secondary monitoring circuit which measures the hysteresis response of the specimen.

The circuit for magnetic properties measurement is depicted in Figure 3. The primary side current is provided by a programmable power supply in a current control operating mode. The current source amplifies a 60Hz-sinusoidal input signal from a signal generator. The maximum current intensity is set sufficiently high to extend the hysteresis loop beyond the point of closure at the tips. It is found that, while true magnetic saturation is not reached, the values of magnetic properties of interest are influenced to only a minor extent by increasing the applied field toward full saturation once the loop closes. The maximum and minimum values of the H field which meet the above criterion are then used for all subsequent measurements. The primary current is monitored using a current probe which is not directly connected to the circuit. This avoids any direct heating effect which might induce inaccuracy in measuring under high current conditions. The signal sensed by the current probe is out of phase with the primary current, however the phase shift is constant and can be adjusted numerically in the data acquisition and processing stage.

During measurement, it is necessary to apply the high current intensity for only a few seconds to allow the diagnosis of required signals on both primary and secondary sides of the circuit. Prior to and following measurement, the current source is disconnected to decrease potential heating of the yoke and specimen. Further cooling is

applied by a cooling fan directed across the yoke-specimen assembly. Measurements of the operating temperature of the yoke reveal that heating is minimal; temperature rises of 2 to 3°C above ambient are typical.

The secondary circuit is a simple system which monitors the potential (voltage) across the pick-up coil windings. The variation in voltage is a direct measure of the magnetic response of the specimen. The integration of the current produced in the pin, a necessary step for the determination of the hysteresis response, is performed by the data acquisition and analysis system. Repeated tests on a standard specimen showed acceptable standard deviations of all measured parameters and established the reliability and applicability of this numerical integration method. Variations in measurements were less than 2.5% in all cases.

In a previous description of magnetic properties monitoring techniques [5], a secondary integration circuit was employed to monitor magnetic hysteresis response. This circuit was found to have the useful feature that the hysteresis response could be monitored directly from the charging and discharging of the capacitor in the circuit. However, the components, principally the capacitor, on the secondary side were found to provide possible inaccuracies in the desired measurements. While the results were amenable to correction, a simpler means of avoiding such difficulties was available, that is, the elimination of the secondary side circuit. The change to measure only the voltage variation across the specimen coil also eliminated any possibility of mutual inductance in the secondary circuit, which could also have complicated the measurements.

Material and Irradiation

Specimens for magnetic properties measurements were cut from a plate of A533B nuclear pressure vessel steel. The composition of the plate and its heat treatment history are given in Table 1. These steel specimens were fabricated in the shape of cylindrical pins with diameter 1.51 mm and length 12.8 mm and blocks with dimensions of 2mm by 2mm by 6mm. The cylindrical specimens were designed to be inserted in the slot of a miniature yoke, Figure 2, to take magnetic properties determinations, while blocks were used to establish microhardness values of material.

Specimen were irradiated for varying numbers of cycles at full power (1.5 MW) in the University of Illinois Advanced TRIGA reactor. Irradiation periods were typically 8 hours and several irradiation periods were required to reach the desired fluences. To date, the maximum total accumulated fluence is 4×10^{18} n/cm²

($E > 0.1\text{MeV}$). Prior to irradiation, samples were vacuum-encapsulated. Samples were placed both in the central core region and in a cadmium lined location at the periphery of the core to obtain two flux and fluence levels. Nickel wires were employed to determine neutron fluences.

Besides the dose rate difference, the irradiation temperature was higher in the core central position than in the peripheral position. Thermal paint was applied to separate block specimens to estimate the irradiation temperature. From color changes of the paint, the temperature at the central core location was determined to fall between 250 and 280°C. Temperature in the core peripheral region was approximately 120°C.

In addition to the irradiations, specimens were heat treated either to intentionally alter the steel microstructure, or to establish whether the thermal cycles associated with the irradiations at 250 to 280°C had any influence on the materials hardness or magnetic properties in the absence of radiation. The former heat treatments were described previously. [5] The latter heat treatments were carried out on six batches of specimens, aged at 260°C for a number of 8hr aging cycles. Magnetic parameters and hardness values were determined following each of the cycles.

A complete listing of the irradiation and heat treatment parameters are provided in Table 2.

RESULTS AND DISCUSSION

Results of the magnetic properties measurement are shown in Figure 4 for two pins irradiated in the core peripheral region to doses of 5.22×10^{17} and 6.34×10^{17} n/cm² ($E > 0.1\text{MeV}$) where the irradiation temperature was on the order of 120°C. A loop from the as-received condition is also shown for comparison sake. The loops show that a notable increase in the effective magnetic saturation point is identifiable, however similar values for the remanence, B_r (value of B at $H=0$), and the coercivity, H_c (value of H at $B=0$), are nearly identical within the experimental error. The hysteresis losses, noted in Table 2, increase with dose for the irradiated specimens.

For specimens irradiated at higher temperatures, smaller changes in magnetic response following irradiation were observed. Figure 5 shows the hysteresis loops for two of ten specimens irradiated to doses up to 4×10^{18} n/cm² ($E > 0.1\text{MeV}$). Both specimens again exhibit higher saturation magnetization than the as-received pin, though in these cases, the differences are very small. Similar values of coercivities and remanences were found again regardless of specimen exposure. Furthermore, while the two irradiated conditions were very different neutron doses, the hysteresis loops

nearly overlap suggesting effects other than radiation damage have influenced magnetic properties. This is consistent with no notable changes in hardness measured at those conditions.

To separate temperature and irradiation effects, specimens were heated and cooled to simulate the thermal cycles which the pins irradiated at 260°C experienced. The results of two specimens from the set which received the heat treatments only are depicted in Figure 6. It is evident that changes in the hysteresis response due to heat treatment are similar to those found during irradiation at 260°C, and are small in all cases. This is again consistent with the small observed changes in specimen hardness in those cases.

Other heat treatments were carried out to alter the as received materials microstructure; the heat treatment conditions are noted in Table 2. [5] It can also be noted that these heat treatments resulted in substantial softening of the material. The hysteresis responses of these specimens are shown in Figure 7. The results indicate that observable changes were shown in most of the standard magnetic properties.

A comparison of the hysteresis curves from unirradiated, irradiated and thermally treated specimens in Figures 4 to 7 shows that the effective magnetic saturation rises with irradiation exposure and may be affected by heat treatment. Even at low neutron dose differences, deviations in the values are apparent in the figures. These changes are best reflected in the hysteresis loss, which correlates with increases in the effective magnetic saturation.

The magnetic properties of the A533B pressure vessel steel examined here were found fairly radiation-resistant at the higher irradiation temperature and more sensitive to irradiation at the lower temperature. Earlier work by Gordon and Sery [1] show small irradiation-induced effects in high purity Fe. However, the lack of pronounced changes in the higher irradiation temperature tests here may be more directly associated with the low fluences. The lower temperature irradiations show a greater degree of sensitivity which is consistent with observed temperature dependences of post irradiation mechanical properties changes. Further work is underway to clarify the temperature and dose effects.

CONCLUSIONS

A nondestructive method for monitoring irradiation-induced changes in magnetic properties of nuclear pressure vessel steels was described, and typical measurements on small specimen were carried out. The technique is able to employ

very small volumes of material and has been shown to be sensitive to magnetic properties changes following irradiation to less than $1 \times 10^{18} \text{ n/cm}^2$ ($E > 0.1 \text{ MeV}$) at approximately 120°C . To date, irradiations to higher doses at approximately 250°C have not produced changes in either microhardness or magnetic properties. Changes in both microhardness and magnetic properties are found after certain thermal aging studies. In all cases where changes have been seen, increases in the hysteresis loss correlates with increased microhardness. In cases where no irradiation-induced magnetic properties changes are found, the microhardness is also found not to change.

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REFERENCES

1. D. I. Gordon and R. S. Sery, "Irradiation of Iron and 7-59 Mo Permalloy with 2 MeV Electrons," J. Appl. Phys., **35**, No. 3 (1964) 879-880.
2. C. W. Chen, Magnetism and Metallurgy of Soft Magnetic Materials, Vol. XV in Selected Topics in Solid State Physics, North Holland Publ. Co., Amsterdam, 1977, Chapter 7, Part 1.
3. E. H. El-Kalla, F. A. Rahman, and A. A. Maaty, "The Effect of Neutron Irradiation on the Magnetic Properties of Low Carbon Steels," Appl. Radiat. Isot., **40**, No. 2 (1989) 159-162.
4. O-Y. Kwon, K. Ono, G.E. Lucas and G.R. Oddette, "Detection of Irradiation Effects on Reactor Pressure Vessel Steels by Magneto-Acoustic Emission," in Proceedings of the Ninth International Conf. on NDE in the Nuclear Industry, Tokyo, Japan April 24-28, 1988.
5. J.F. Stubbins, J.G. Williams, A.M. Ougouag, J.U. Patel, and W-J. Shong, "Measurement of Irradiation Damage in Nuclear Pressure Vessel Steels by Magnetic Properties Changes," in Proceeding of the Fifth International Symposium on Environmental Degradation of Materials in Nuclear Power Systems -- Water Reactors, ed. E.P. Simonen, in press.
6. J. R. Cost, R. D. Brown, A. L. Giorgi, and J. T. Stanley, "Radiation Effects in Rare-Earth Permanent Magnets," IEEE Trans on Magnetics, **24** (1988) 2016-2019.

7. R. D. Brown and J. R. Cost, "Radiation-Induced Changes in Magnetic Properties of Fe-Nd-B Permanent Magnets," IEEE Trans on Magnetics, **25** (1989) 3117-3120.
8. J. R. Cost and R. D. Brown, "Sm-Co Permanent Magnets: Effects of Fast Neutron Irradiation," Met. Trans. A, **21A** (1990) 1817-1821.
9. M. Takahashi, "Physics and Engineering Applications of Magnetism", ed. Y. Ishikawa and N. Miura, Springer-Verlag, 1991, Vol.92, p.164
10. D. Jiles, Introduction to Magnetism and Magnetic Materials, Chapman and Hall, London, 1991.

LIST OF FIGURES

1. Typical hysteresis loop of A533B pressure vessel steel. The positions of the remanence, coercivity, and effective magnetic saturation are indicated. The hysteresis loss is the area of the curve.
2. Magnetic properties measurement specimen, yoke, and sensing reel arrangement. For magnetic properties measurement, the specimen is inserted into the yoke/reel apparatus on the right hand side.
3. The primary circuit and the secondary or integrating circuit for making magnetic property measurements. The inputs to an oscilloscope are indicated.
4. The hysteresis loops for specimens irradiated at approximately 120°C: AR: as received (unirradiated) condition; 1: 5.22×10^{17} n/cm² (E>0.1MeV) and 2: 6.34×10^{17} n/cm² (E>0.1MeV).
5. The hysteresis loops for specimens irradiated at approximately 260°C: AR: as received (unirradiated) condition; 1: 6.5×10^{17} n/cm² (E>0.1MeV) and 2: 2.28×10^{18} n/cm² (E>0.1MeV).
6. The hysteresis loops for specimens heat treated with thermal cycles to simulate the reactor exposures at 260°C; AR: as received, HT1: 260°C for 8 hr and HT2: cycled to 260°C for 8 hr twice, for 3 hr once, then for 8 hr three times. These thermal cycles were the same as the specimens (1 and 2) received during their irradiation cycles, see Figure 5 for the comparable irradiation results.
7. The hysteresis loops for specimens heat treated to various annealing conditions: AR: as received, 1: 910°C for 30 min, furnace cool; 2: 900°C for 20 min, furnace cool to 800°C, air cool; and 3: 900°C for 20 min, furnace cool to 800°C, air cool, 700°C for 48 hr, air cool.

Table 1
Chemical Composition (wt. %) of A533B Cl. I Plate*

C	Mn	P	S	Si	Mo	Ni	Cu	Cr
0.19	1.32	0.013	0.013	0.22	0.54	<.05	0.12	<.05

* Plate donated by Combustion Engineering, the plate had received a post-weld heat treatment cycle to simulate typical nuclear pressure vessel fabrication practices

Table 2

Properties of Irradiated and Heat Treated A533B Steel Specimens*

Condition	Fluence (10E18 n/cm ²)	Hysteresis Loss (10E6 erg/cm ³)		Coercivity (Oe)		Remanence (KGu)		DPH ⁺
		pre	post	ratio	pre	post	ratio	
Control		1.81	1.81	1.00	17.47	17.35	1.00	291
<u>Irradiation Temperature 260°C</u>								
	0.65	1.70	1.80	1.06	16.51	17.46	1.06	-
	0.65	1.79	1.83	1.02	17.22	17.95	1.04	-
	1.32	1.82	1.82	1.00	17.73	17.42	0.98	334
	1.32	1.64	1.70	1.03	16.49	17.32	1.05	341
	2.28	1.72	1.88	1.09	16.12	17.54	1.09	328
	2.28	1.80	1.82	1.01	17.17	18.01	1.05	319
	2.98	1.82	1.83	1.01	17.56	17.63	1.00	258
	2.98	1.62	1.68	1.03	16.57	16.96	1.02	276
	3.64	1.84	1.82	0.99	17.55	17.76	1.01	291
	3.64	1.83	1.82	0.99	17.74	17.27	0.97	291
<u>Irradiation Temperature 120°C</u>								
	0.52	-	1.86	-	-	17.03	-	-
	0.63	-	1.93	-	-	17.15	-	-
<u>Heat Treatments</u>								
HT1	-	-	1.70	-	-	17.22	-	85
HT2	-	-	1.70	-	-	17.15	-	81
HT3	-	-	1.74	-	-	16.92	-	83
HT4	-	1.79	1.81	1.01	17.56	17.90	1.02	271
HT5	-	1.83	1.81	0.99	17.65	17.05	0.97	234

* To avoid oxidation or loss of carbon, each specimen was vacuum encapsulated for heat treatment

+ Measured with a 100 gm load

HT1: 910°C for 30 min, furnace cool

HT2: 900°C for 20 min, furnace cool to 800°C, air cool

HT3: 900°C for 20 min, furnace cool to 800°C, air cool, 700°C for 48 hr, air cool.

HT4: 260°C for 8 hr, air cool

HT5: 260°C for 43 hr (8+8+3+8+8), air cooled between each interval

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