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Monitoring neutron embrittlement in nuclear pressure vessel steels using micromagnetic Barkhausen emissions

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Abstract
In nuclear power plants, neutron embrittlement of pressure vessel steels has been one of the main concerns. The use of micromagnetic Barkhausen emissions is a promising method to monitor the variations in microstructural and subsurface stress states due to their influence on these emissions. Measurements of these emissions can reveal neutron irradiation degradation in nuclear power plant components. Samples which were irradiated at different neutron fluences and annealed at different temperatures were obtained from three reactor surveillance programs. The results of different neutron fluences and annealing procedures showed noticeable fractional changes in the magnetic Barkhausen effect signal parameter, ΔMBE/MBE, and in the mechanical properties of these specimens. For example, increased intensity of neutron fluence decreased the ΔMBE/MBE as well as impact energy and upper-shelf energy, but increased Rockwell hardness and yield strength. Typical changes in this parameter were in the range from −20% to −45% for fluences of up to 25×10^{18} n cm^{-2}.

Keywords
Neutrons, Barkhausen effects, Annealing, Molecular beam epitaxy, Neutron radiation effects

Disciplines
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Comments
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In nuclear power plants, neutron embrittlement of pressure vessel steels has been one of the main concerns. The use of micromagnetic Barkhausen emissions is a promising method to monitor the variations in microstructural and subsurface stress states due to their influence on these emissions. Measurements of these emissions can reveal neutron irradiation degradation in nuclear power plant components. Samples which were irradiated at different neutron fluences and annealed at different temperatures were obtained from three reactor surveillance programs. The results of different neutron fluences and annealing procedures showed noticeable fractional changes in the magnetic Barkhausen effect signal parameter, AMBE/MBE, and in the mechanical properties of these specimens. For example, increased intensity of neutron fluence decreased the AMBE/MBE as well as impact energy and upper-shelf energy, but increased Rockwell hardness and yield strength. Typical changes in this parameter were in the range from −20% to −45% for fluences of up to 25×10¹⁸ n cm⁻².

INTRODUCTION

The nuclear industry has been investigating inspection methods to ensure safe operation of nuclear reactors beyond their projected life span. Previous work has concentrated not only on destructive techniques such as Charpy tests but also on noninvasive methods such as x-ray, eddy current, hysteresis, positron annihilation, and magnetoacoustic emission measurements. Neutron bombardment creates point defects distributed throughout the irradiated material. Magnetic properties such as remanence, coercivity, maximum differential permeability, and hysteresis loss are sensitive to the presence of these defects, as are Barkhausen emission spectra which were employed to characterize microstructure and subsurface stress states of the materials.

EXPERIMENTAL

The samples investigated were mainly broken Charpy specimens of A533 grade B class 1 pressure steels. Some measurements also were taken on unbroken Charpy samples (27.5×10×10 mm³). Classification of the samples was made on the basis of the original surveillance program data at three different nuclear reactor plants. All the magnetic hysteresis measurements and magnetic Barkhausen measurements on base or weldment portions of the irradiated samples were taken in the radioactive “hot cell” using a commercial Barkhausen effect device, which has been described elsewhere.

RESULTS AND DISCUSSION

Measurements at Westinghouse Electric Corp., Pittsburgh, PA were conducted on the specimens from the first surveillance program where they had been irradiated to three different levels of neutron fluences: 3.9×10¹⁸, 17.7×10¹⁸, and 23.7 n cm⁻². The analysis showed that magnetic Barkhausen signals at each measurement depth were influenced by neutrons.

To investigate the response of Barkhausen emissions to the changes in the mechanical properties of the samples as a result of neutron embrittlement, the fractional change in the magnetic Barkhausen signal was compared to change in Rockwell hardness, upper-shelf energy, yield strength, and impact energy over the range of neutron irradiation fluence. On these same specimens, the effect of sample orientation with respect to rolling direction was also studied. The samples were denoted as transverse and longitudinal. Weldment specimens from the first surveillance program were also measured.

Similar measurements were carried out on the samples from the second surveillance program where fluence of neutrons was 13.3×10¹⁸ n cm⁻². As summarized in Table I, the Barkhausen signal as well as the impact energy and upper-shelf energy decreased with neutron influence, whereas Rockwell hardness and yield strength were found to increase with neutron fluence over the range of 0–25×10¹⁸ n cm⁻². It was expected that higher hardness would result in lower magnetic Barkhausen emissions due to the increased number of defect pinning centers which impede both the movement of dislocations and magnetic domain walls.

The second part of investigation was to study the influence of successive neutron irradiation and annealing processes on magnetic Barkhausen signals from the samples from the third surveillance program. Barkhausen voltage wave forms from the unirradiated sample, [Fig. 1(a)], from the sample irradiated at 8.4×10¹⁸ n cm⁻², [Fig. 1(b)], and from the sample annealed at 850 F for 168 h, [Fig. 1(c)] correspond to changes in micromagnetic emissions in the materials of interest. Before irradiation, the rms Barkhausen voltage was 6 V while after irradiation it was 2.5 V. Annel-
TABLE I. The changes in magnetic Barkhausen signal parameter and mechanical properties.

<table>
<thead>
<tr>
<th>Change in magnetic Barkhausen signal</th>
<th>Change in Rockwell hardness</th>
<th>Change in yield strength</th>
<th>Change in impact energy</th>
<th>Change in upper-shelf energy</th>
</tr>
</thead>
<tbody>
<tr>
<td>Transverse samples from the 1st surveillance program</td>
<td>-45%</td>
<td>48%</td>
<td>18.5%</td>
<td>-26%</td>
</tr>
<tr>
<td>Longitudinal samples from the 1st surveillance program</td>
<td>-19%</td>
<td>41%</td>
<td>19.5%</td>
<td>37%</td>
</tr>
<tr>
<td>Weldment samples from the 1st surveillance program</td>
<td>-42%</td>
<td>40%</td>
<td>9.5%</td>
<td>-52%</td>
</tr>
<tr>
<td>Samples from the 2nd surveillance program</td>
<td>-21%</td>
<td>37%</td>
<td>7%</td>
<td>-64%</td>
</tr>
</tbody>
</table>

ing resulted in a signal of 5 V. It was also found that the magnetic Barkhausen signal parameter decreased as a result of the first irradiation at $8.4 \times 10^{18}$ n cm$^{-2}$ at all measurement frequencies and hence at all depths. The first anneal, 168 h at 850 °F, restored the Barkhausen parameter. On the other hand, the second irradiation, an additional $6.6 \times 10^{18}$ n/cm$^2$, did not cause noticeable change in the Barkhausen signal. However, the second annealing step with some experimental conditions, as in the first annealing, further restored the magnetic Barkhausen signal as seen in Fig. 2. The magnetic Barkhausen signal together with the Rockwell hardness, impact energy, and upper-shelf energy have been plotted for each case in Fig. 2. As can be seen in Fig. 2(a), the first irradiation led to a decrease in magnetic Barkhausen signal while Rockwell hardness increased. On the other hand, the first annealing process decreased the Rockwell hardness but increased the magnetic Barkhausen signal. The impact energy, Fig. 2(b), and the upper-shelf energy, Fig. 2(c), in every case.
CONCLUSIONS

This investigation revealed the existence of a relationship between the extent of radiation damage by neutrons and the Barkhausen emission signals. There were significant correlations between mechanical properties of these materials and magnetic Barkhausen signals and these have been interpreted in terms of the effects of defects on both magnetic domain wall motion and dislocation movement. The results indicated that irradiation with neutrons caused damage throughout the volume of the material. It was anticipated that increased hardness would give rise to lower Barkhausen emissions due to increased pinning of domain walls. Successive neutron irradiation and annealing procedures also revealed a relationship between Rockwell hardness and the magnetic Barkhausen signal which resulted from neutron embrittlement.

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