IV Conferencia Panamericana de END
Buenos Aires – Octubre 2007

AAENDE
Asociación Argentina de Ensayos No Destructivos y Estructurales

Nondestructive Evaluation of Ageing Steel Structures

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Abstract

Usually, materials are inspected by nondestructive methods to detect and evaluate defects that may cause failure under the designed operation conditions. However, the structure may also fail due to uncertainties of material properties like strength or hardness, and may also suffer from unexpected degradation during operation. A reliable NDE technique for detecting the inception of failure during early stages of embrittlement is presently not available. For the characterization of material degradation phenomena, nondestructive methods that are sensitive to the microstructure of the material are required and useful. However, they must be applied in the field. The difference between Microscopy and NDE poses a challenge when dealing with the scanning of macroscopic objects. The presented results were tested under laboratory conditions only and have to be certified through comprehensive field trials. Appropriate techniques for the assessment of material degradation should be able to detect and identify micromagnetic properties of ferritic steel, for example; magnetization phenomena are affected by microstructure and stress states. Other techniques with similar potential are Eddy Current, Ultrasonic and Thermal testing methods.
We present results of a research project funded within the German Nuclear Safety Program. Using different micromagnetic methods we were able to analyze the neutron induced embrittlement of a 20 MnMoNi 55 steel material. Magnetic Barkhausen noise data, the upper harmonics analysis of magnetization, and eddy current data allowed a nondestructive prediction of Vickers hardness and the shift of the ductile-to-brittle transition temperature (DBTT) that characterizes embrittlement. As a second example, we present results that allow the assessment of the precipitation induced embrittlement in WB 36 (15 NiCuMoNb 5) steel material. Both examples demonstrate the complexity of the problem and the viability of the NDT approach.

Keywords: Thermal ageing, neutrons embrittlement, nondestructive, micromagnetic

1. Introduction

Nondestructive material characterization techniques have traditionally been employed to detect, classify and size defects in materials. However in the last two decades a significant amount of effort has been invested to develop NDT techniques which can reliably characterize materials in terms of properties describing the fitness for use. In the case of power plant components, such as pressure vessels and pipes, the fitness for use under mechanical loads is characterized in terms of the determination of mechanical properties like mechanical hardness, yield and tensile strength, toughness, ductile-to-brittle transition temperature, fatigue strength or usage factor. With the exception of hardness tests, which are weakly invasive, all of these parameters can be determined by using destructive tests on special standardized samples. Such a procedure cannot be performed on components in service and is therefore restricted to quality checks during manufacturing where sufficient representative material is available. Procedures based on less destructive material sampling and weakly influencing the integrity of the component are not yet validated or standardized and are still the object of investigations. Therefore, there is a need for the development of such nondestructive testing techniques.

2. Metallurgical Aspects of Embrittlement

When irradiating metals by neutrons, two basic processes occur, core transformations and generation of vacancies. In technical materials such as steels, further processes can appear, particularly as a consequence of the generation of vacancies. This is the reason for the irradiation or neutron embrittlement. The irradiation (neutron) embrittlement of a reactor pressure vessel (RPV) is an essential factor used in evaluating the lifetime of nuclear power plants. Irradiation embrittlement appears either as a shift of the ductile-to-brittle transition temperature to higher temperatures, or as a reduction of the upper shelf toughness. The process depends on environmental influences such as radiation dose energy and temperature. Metallurgical characteristics (alloy composition, microstructure of the initial state) also play an important role. The embrittlement of typical (Mn-Mo-Ni-) pressure vessel-steels, which is manifested as a shift of the ductile-to-brittle temperature, accompanies an increase in strength and the yield strength. Therefore, in the English scientific literature this effect is often referred to as irradiation hardening instead of irradiation embrittlement.

Based on current knowledge, radiation induced diffusion and generation of defect clustering are the basic mechanisms of irradiation embrittlement. The formation of so called freely migrating defects is essential for the diffusion amplified by irradiation. This irradiation leads to the generation of different very fine (typical 1 to 2 nm large) phases at service temperatures of approximately 290 °C, causing enhancement of the hardness of the material by dislocation hindrance. It is particularly important that particles rich in copper (Cu) are considered as well as particles rich of manganese and nickel, which are generated in the presence of Cu. Matrix defects, generated independent from the Cu content, may also contribute to the embrittlement.

3. Assessment of the Embrittlement

In addition to the mechanical properties, the embrittlement phenomenon also influences the magnetic properties. Smallest changes in the materials state can affect the magnetic domain structure. Micromagnetic test methods have a high potential to detect the change of microstructure defects because they sensitively react to changes of the domain wall (which separate the magnetic domains) configuration. Therefore, electromagnetic procedures could potentially be used to assess the material embrittlement. Due to that fact electromagnetic technologies are suitable for the acquisition and follow-up of various parameters linked to the characterization of the embrittlement and to deliver them to a failure assessment system for extraction.

Using several electromagnetic measurements at the same time, a variety of measuring quantities is derived for each measurement cycle. When combined they achieve the desired result (e.g. material property) more effectively than when they are each considered individually. By using the calibration function the desired quantity of an unknown set of samples investigated by that method can be detected nondestructively.

3.1 NDT Characterization of the Thermal Induced Embrittlement

Smallest changes in the materials state, e.g. the change of the precipitation of coherent Cu particles induced by thermal ageing, affect the magnetic domain structure.

In the typical “as delivered” state of WB 36 (15 NiCuMoNb 5), half of the contained Cu is already precipitated, while the other half remains in solid state. After long term service exposure above 320 °C, damage was observed due to further precipitation of Cu; an increase in yield strength $\Delta\sigma y$ = +150 MPa and a shift of the ductile-to-brittle transition ΔFATT = 70 °C can be measured. Small angle neutron scattering measurements revealed the fact that changes in the mechanical properties are caused by Cu precipitates ranging from 1 to 3 nm in size[(1)]. The particles are coherent, have a bcc structure that induces a high level of compressive residual stress in their vicinity, balanced by tensile stresses in the environmental matrix. On a set of approximately 70 cylindrical samples (80 mm in length, diameter 6 mm) of WB 36, thermal service exposure was simulated in an accelerated manner through long-term annealing at 400 °C. A U-shaped electromagnet was used to excite an alternating magnetic field along the longitudinal axis of the sample. A disc-shaped pickup coil and a temperature stabilized hall probe were used to record Barkhausen noise events and magnetic field strength, respectively. The Barkhausen noise signal was amplified by 60 dB and band-pass filtered to a range of 5 to 200 kHz. All signals were digitized using common data acquisition hardware. The Barkhausen noise signal was then digitally re-filtered for separate analysis of its different frequency components. Characteristic scalar quantities[(2)] were extracted from the envelope of the Barkhausen noise signal as a function of the applied magnetic field strength. Moreover, an upper harmonics analysis[(3)] of the magnetic field strength signal was performed and characteristic quantities derived. As changes in conductivity may be expected due to Cu precipitation, a simplified eddy current analysis procedure was performed based on the relationship between magnetic field strength and exciting voltage of the electromagnetic coil. The measuring quantities of all three methods (Barkhausen noise, upper harmonics and eddy current analysis) were combined to a vector which characterized the material condition.

In the case of WB 36 steel, only a few of the measured quantities seemed to correlate well with the Cu precipitation state. Therefore, a preliminary experiment was performed in order to identify the optimum measurement parameters and most significant quantities for the detection of Cu precipitation in WB 36. The electromagnetic properties of single samples were recorded in several stages of the simulated service exposure. The annealing was therefore interrupted in regular intervals during which electromagnetic tests were performed. All measurements were done using a fixed set of different magnetization frequencies and magnetic field amplitudes in order to create a comprehensive database of material behavior. In total 6 samples underwent this procedure for statistical significance coverage, and reference samples were kept for verification.

As a result of these experiments, it was found that eddy current measurements allowed the detection of Cu precipitates whilst remaining insensitive to most disturbing influences. Figure 1 shows how the coil impedance (expressed in terms of its relative magnitude and phase) performed as a function of the simulated service exposure duration for initially recovery-annealed WB 36. Changes in the eddy current impedance represent changes in the conductivity and the permeability of the material. The electrical conductivity is proportional to the concentration of the free electrons and their velocity, which decreases with rising defect density. Therefore the electrical conductivity decreases. It was also shown that the increase of density of the Cu precipitates causes a decrease of the relative magnetic permeability.

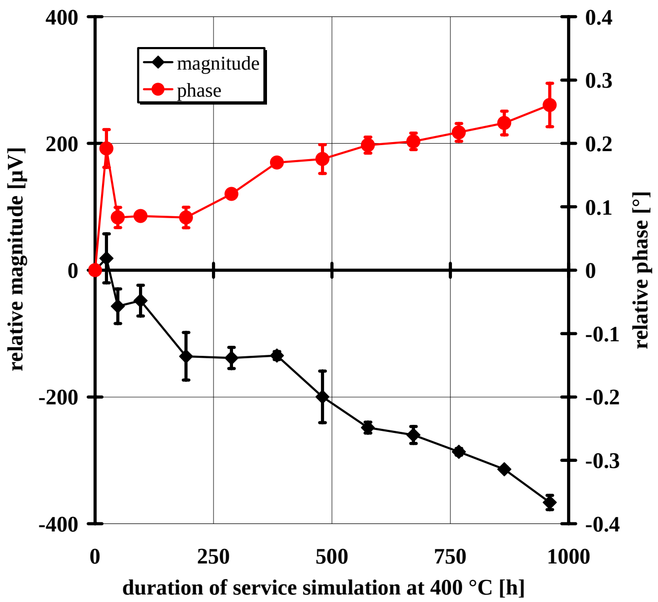

Figure 1. Eddy current quantities magnitude and phase as a function of service exposure duration for initially recovery-annealed (3 h / 600 °C) WB 36 steel

Fraunhofer Institute for Non-Destructive Testing (IZFP) uses the so-called micromagnetic 3MA-method (**M**icromagnetic **M**ulti-Parameter **M**icrostructure and Stress **A**nalysis) in order to solve the inverse problem of target quantity prediction from a limited set of calibration data[(4)]. In this case, a specialized pattern recognition algorithm[(5)] based on nearest neighbor search was used to obtain approximate values of the Vickers hardness (HV 5) from several quantities, including the eddy current quantities mentioned above. The optimum parameters which were found in the preliminary experiments were used throughout this measurement. Figure 2 shows the resulting correlation of actual and predicted hardness values[(6)].

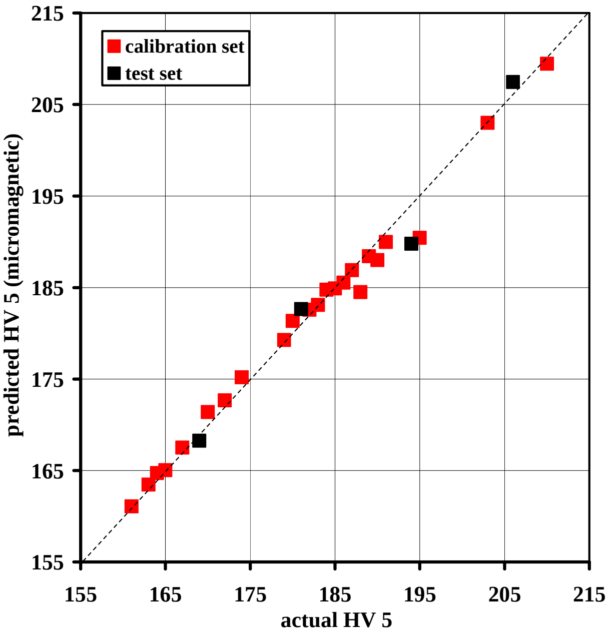

Figure 2. Predicted Vickers hardness for WB 36 versus actual Vickers hardness

3.2 NDT Characterization of the Neutron Irradiation Induced Embrittlement

Depending on the specific design of a pressure vessel – which varies in different countries – the pressure vessel material in nuclear power plants (NPP) exposed to neutron flux is in a range between $5*10^{18}$ n/cm^2 (in 32 years at 288 °C) in Germany and $8*10^{19}$ n/cm^2 (in 14 years at 254 °C) in France. The energy input of the neutrons is producing lattice defects like vacancies both directly and indirectly by stimulating the precipitation of coherent Cu-rich particles. As in the case of the thermally-induced embrittlement of the steel WB 36 their diameter is approximately 3 mm and they are coherently embedded in the bcc lattice. Both the vacancies and the precipitates reduce the toughness of the material, which can be characterized by a reduction of Charpy energy and a shift in the fracture appearance transition temperature to higher temperatures. In practice the material degradation is characterized in surveillance programs by using standardized Charpy V-notch specimen and tensile test specimen made of the pressure vessel steel and its welds. The specimens are exposed in special radiation chambers near the NPP core at a higher neutron flow than at the surface of the pressure vessel wall. From time to time these specimens are removed from the chambers and used for destructive tests. In order to assume higher nuclear safety between two subsequent destructive tests one would like to have many nondestructive tests and therefore NDT technology should also be developed as an in service inspection method to be applied at the pressure vessel inner surface. In order to characterize the neutron irradiation-induced embrittlement, samples (10*10*55 mm) made of 20 MnMoNi 55 steel used in the German reactor pressure vessel in non-irradiated state, as well as after irradiation of $3.78*10^{18}$ n/cm^2, $7.66*10^{18}$ n/cm^2 and $1.05*10^{19}$ n/cm^2 neutron flux have been investigated. As in the case of the thermally-induced embrittlement of the steel WB 36, it was observed that a suitable measuring quantity for the characterization of the neutron irradiation-induced embrittlement is the eddy current impedance magnitude which decreases with the neutron flow (Figure 3). This fact suggests that, similar to the steel WB 36, where the decrease of the eddy current impedance magnitude occurred due to the thermal ageing-induced Cu precipitation (Figure 1), in case of the samples made of Cu containing steel 20 MnMoNi 55 the decrease of the eddy current impedance magnitude was caused by the precipitation of Cu but induced during the neutron irradiation.

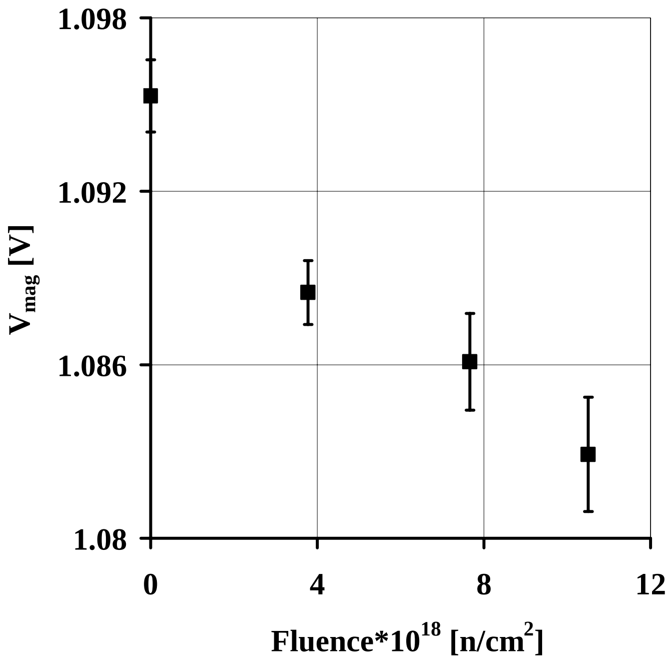

Figure 3. Correlation between the eddy current impedance magnitude and the neutron flow

IZFP has applied micromagnetic approaches to calibrate regression models. The measurements were performed in the same way as in the case of the steel WB 36. One part of each specimen set was used to calibrate and the other part – independently selected – was taken to test the model. In this case the pattern recognition algorithm mentioned above(5) was used to obtain approximate values of the shift of ductile-to-brittle transition temperature, which is a measure of the embrittlement.

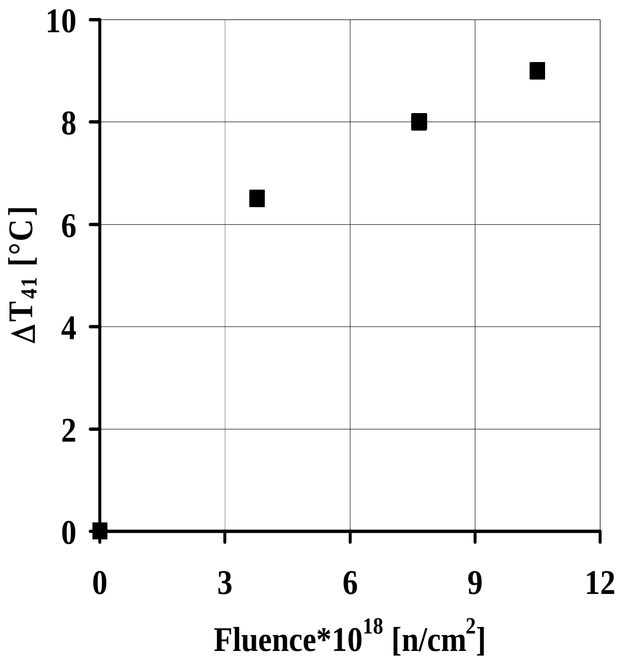

Figure 4. Dependency of the ductile-to-brittle transition temperature on the neutron flow

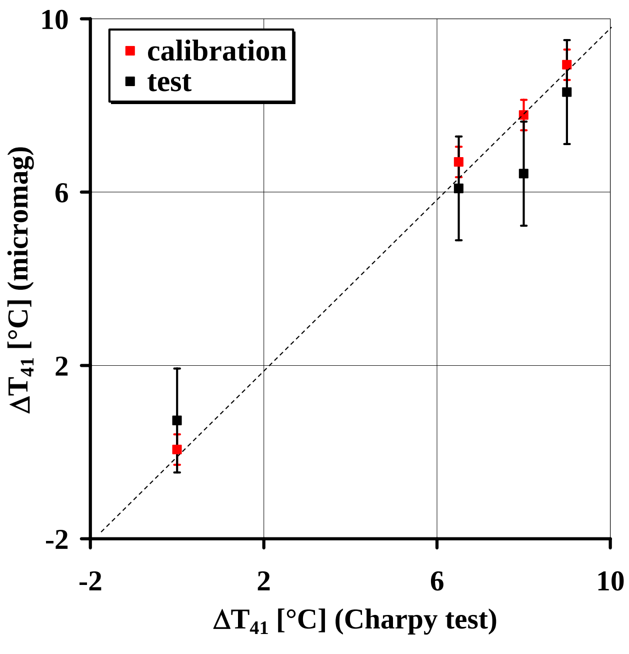

Figure 5. Prediction of the shift of the ductile-to-brittle transition temperature by the micromagnetic method

It was demonstrated that by means of the micro-magnetic procedure the prediction of the shift of the ductile-to-brittle transition temperature is possible (Figure 5). By using a regression analysis algorithm a correlation coefficient of 98.3 % and a residual standard deviation of 0.35 °C is reached. By testing the calibration with independently selected test specimens a standard error of 1.2 °C (residual standard deviation to the destructive test values) was obtained.

First micro-magnetic measurements showed that a nondestructive detection of the neutron irradiation on the component is possible, even under the presence of practical disturbances like the austenitic cladding[(7)].

In order to ensure deep penetration of the electromagnetic fields, low-frequency EMATs were used, and hence it was possible to examine the material condition in the ferritic base material directly below the austenitic cladding from the clad side. For that purpose low-frequency (< 10 kHz) electromagnetic ultrasonic transducers should be developed, which are working mainly magnetostrictively.

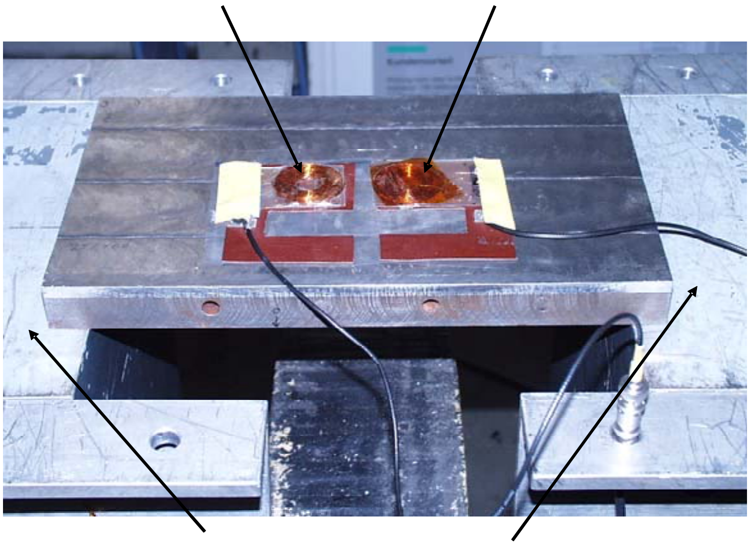

Figure 6. EMAT configuration with HF-coils and laboratory electromagnet

The possibility of exciting and receiving ultrasonic waves in the ferritic base material through the cladding was tested with the help of HF-coil-configurations on differently clad test objects both by means of laboratory electromagnets and by means of a permanent magnet system. Signal-to-noise ratios of 23 dB were reached (Figure 7) on an austenitic clad test object with a wall thickness of 30 mm and with austenitic cladding thicknesses of 8 mm at an excitation on the clad side with magnetizing field strength of 250 A/cm (use of laboratory magnet) and frequencies between 40 and 50 kHz.

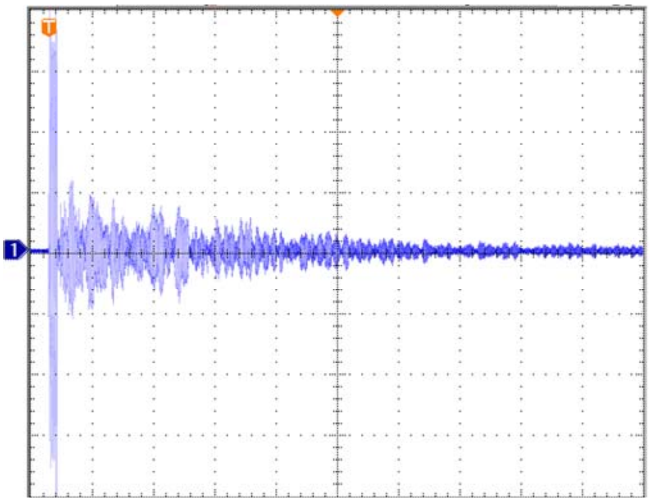

Figure 7. Ultrasound signals by an excitation on the austenitic clad side (8 mm thickness), magnetic field strength: 260 A/cm, frequency: 50 kHz

A second experiment was conducted to clarify if ultrasonic signals can still be produced on the ferritic side even with a coil lift-off of 8 mm. For this purpose the lift-off was achieved by using an 8 mm thick PVC plate. The obtained echo signals are presented in Figure 8. It can be observed that the signals obtained on the austenitic clad side are smaller. The smaller excitation effectiveness on the clad side (Figure 7) is caused by the shield effect of the austenitic cladding compared to the PVC plate. The experiments proved that it is possible to transmit and receive ultrasonic signals magnetostrictively through the cladding of the ferritic base material.

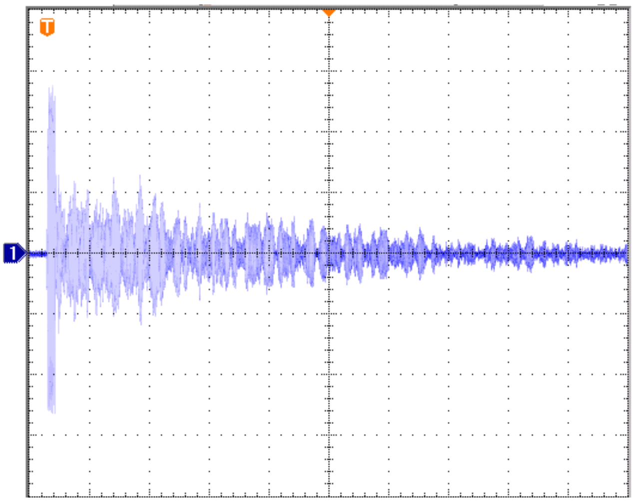

Figure 8. Ultrasound signals generated by an excitation through a PVC plate (8 mm thickness), magnetic field strength: 260 A/cm, frequency: 50 kHz

4. Conclusion

In the present research work the NDT characterization of the material degradation was discussed on the basis of experiences with precipitation-induced embrittlement in WB 36 steel and neutron-irradiation induced embrittlement in the 20 MnMoNi 55 steel used in German reactor pressure vessels. The study also shows that eddy current impedance measurements represent a suitable method to characterize thermally-induced as well as the neutron irradiation-induced embrittlement. It was shown that the micro-magnetic approach using pattern recognition or regression analysis methods based on magnetic Barkhausen noise, upper harmonics analysis, incremental permeability and eddy current analysis data allows with high accuracy a nondestructive prediction of the Vickers hardness and of the shift of the ductile-to-brittle transition temperature, which are measures to characterize embrittlement. A future application of the micro-magnetic and EMAT testing methods on the component appears feasible based on research observations. Further investigation is necessary into electromagnetic characterization of fatigue behavior in comparison to embrittlement behavior by superposition of thermal and mechanical material load.

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