Early Detection of Critical Material Degradation by Means of Electromagnetic Multi-Parametric NDE

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Abstract. With an increasing number of power plants operated in excess of their original design service life an early recognition of critical material degradation in components will gain importance. Many years of reactor safety research allowed for the identification and development of electromagnetic NDE methods which detect precursors of imminent damage with high sensitivity, at elevated temperatures and in a radiation environment. Regarding low-alloy heat-resistant steel grade WB 36 (1.6368, 15NiCuMoNb5), effects of thermal and thermo-mechanical aging on mechanical-technological properties and several micromagnetic parameters have been thoroughly studied. In particular knowledge regarding the process of copper precipitation and its acceleration under thermo-mechanical load has been enhanced. Whilst the Cu-rich WB 36 steel is an excellent model material to study and understand aging effects related to neutron radiation without the challenge of handling radioactive specimens in a hot cell, actually neutron-irradiated reactor pressure vessel materials were investigated as well. The neutronfluence experienced and the resulting shift of the ductile-brittle transition temperature were determined electromagnetically, and it was shown that weld and base material can be distinguished from the cladded side of the RPV wall. Low-cycle fatigue of the austenitic stainless steel AISI 347 (1.4550, X6CrNiNb18-10) has been characterized with electromagnetic acoustic transducers (EMATs) at temperatures of up to 300 °C. Time-of-flight and amplitude of the transmitted ultrasound signal were evaluated against the number of load cycles applied and observed as an indication of the imminent material failure significantly earlier than monitoring stresses or strains.

Keywords: electromagnetic, micromagnetic, 3MA, EMAT, ultrasound, time-of-flight, magnetostriction

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INTRODUCTION

An increasing number of power plants operates in excess of or near the original design service life. Particularly in the field of nuclear power generation, high safety margins in the component design parameters allow for a certain amount of material degradation to occur without having to discontinue operation. However, there is a need to understand and monitor aging effects in the individual materials. In this context, many components, e.g. feed water pipes, surge lines and reactor pressure vessels are exposed to thermal, mechanical and (partially) radiation loads which promote aging over different, material-dependent mechanisms [1]. Most of the relevant effects take place on the micro- and nanometer scale, e.g. the occurrence of dislocation development, precipitates as well as micro cracks, pores and voids, accompanied by the respective micro residual stresses.

A long-lasting history of projects in the field of reactor safety research has identified non-destructive testing methods suitable for the characterization of changes in these properties [2–5]. A well-known example is magnetic Barkhausen noise analysis, where the physical interaction of magnetic domain walls with microstructure and residual stress fields allows the scientist to investigate microscopic properties of ferromagnetic steel with rather macroscopic, practical sensor elements (coils) [6]. Most magnetic properties reflect those of the microstructure and are affected by stress. This is the basis of micromagnetic materials characterization.

In the case of austenitic steel, where micromagnetic effects only occur if the chemical composition and the service temperature allows a phase transformation from the austenitic γ-fcc lattice to the α’ martensite with bcc-lattice, ultrasound may be used as a means for retrieving information on the microstructure. Small lattice defects,
such as dislocations and micro voids (affecting the mass density), increase in number as fatigue proceeds. Depending on the frequency, they scatter the ultrasonic wave (dispersion) and/or slow down its propagation. Therefore, effects of material degradation reflect in ultrasonic attenuation and time-of-flight (TOF), respectively in phase velocity.

This paper provides an overview of recent findings concerning the application of micromagnetic and ultrasonic methods for characterizing the aging state of several power plant component steels.

**EXPERIMENTAL METHODS**

**3MA**

Since both microstructure and stress affect the magnetic behavior of ferromagnetic materials, the well-known challenge of micromagnetic NDE is the separation of these different superimposed influences in the measured quantities. Therefore, IZFP developed the multi-parametric approach 3MA (Micromagnetic Multi-Parameter Microstructure and Stress Analysis) in order to better deal with superimposed disturbances and increase the range of applications [7]. 3MA comprises the analysis of magnetic Barkhausen noise, incremental permeability, upper harmonics in the magnetic tangential field strength, and eddy current impedance (fig. 1, left hand side). Approximately 40 measuring quantities span a feature space in which different material states can be separated by means of pattern recognition or multivariate regression analysis using a set of calibration data obtained on samples of well-defined, quantitatively known target properties (e.g. Vickers hardness, yield strength, residual stress). Although these quantities are not perfectly uncorrelated to each other, they represent the individual material properties to different extent, e.g. the stress dependence of magnetic Barkhausen noise is much more pronounced than that of incremental permeability [8]. This allows for the implicit suppression of disturbances during calibration.

![FIGURE 1](image)

**FIGURE 1.** The four sub-methods of 3MA and their relation to the magnetic hysteresis loop; photo of device and PC

The technical implementation of 3MA requires a sensor containing a U-shaped electromagnet which is driven with a sinusoidal voltage signal and excites an alternating magnetic field in the material it is placed on. Magnetic Barkhausen noise is picked up by a small pancake-shaped coil between the poles of the electromagnet. Eddy current impedance and incremental permeability are measured by analyzing the impedance of either the Barkhausen noise pickup coil or an additional coil. Harmonics analysis is performed using the magnetic tangential field signal detected by a Hall probe. All signals are preprocessed by means of analog and digital filters in the 3MA device (fig. 1, photo to the right) and finally analyzed by a PC which controls it.

**Ultrasonic Time-of-Flight and Dynamic Magnetostriction Measurement with EMATs**

Uncooled ElectroMagnetic Acoustic Transducers (EMATs) withstand temperatures up to 300 °C and can be applied in a radiation-exposed environment. EMATs generate and receive ultrasound in conductive and/or magnetic materials without couplant. Fig. 2 illustrates the principle of operation which requires a permanent magnet that provides a bias field in the material and a coil that is driven with short high-frequency burst signals. The ultrasonic
wave is excited directly in the material by means of magnetic, magnetostrictive and Lorentz forces [9]. An EMAT receiver is of same construction and uses the corresponding inverse effects.

The ultrasonic time-of-flight (TOF) is the time the ultrasonic wave requires for travelling from transmitter to receiver. For a given wave mode, material properties and stress are of influence on the effective ultrasound velocity and therefore affect the TOF. The influence of microstructure and texture can be expected in the order of a few percent, whereas the influence of stress lies in the order of one-tenth of a percent [10]. The TOF is determined by fitting a peak of the received waveform with a parabolic function and determining the peak position analytically. The achievable accuracy lies in the single-digit nanosecond range.

![Principle of ultrasound generation with EMATs](Image1)

**FIGURE 2.** Principle of ultrasound generation with EMATs (to the left); large, uncooled high-temperature EMAT (to the right, looking at the HF coil), suitable for temperatures up to 300 °C

Depending on the design and orientation of EMAT coil and bias field, magnetostrictive ultrasound generation becomes the predominant mechanism at low bias field strengths. This effect can be used to indirectly determine magnetostrictive properties of the material, which possess a very high sensitivity for stress [8, 11]. The so-called dynamic magnetostriction (DM) curve of the material requires the strength of the magnetic bias field to be slowly varied while observing the received ultrasound amplitude. This is accomplished by using an electromagnet instead of a permanent magnet in the EMAT. The DM curve E(H) is obtained by plotting the amplitude E of a received ultrasound signal as a function of the applied field strength H. Characteristic quantities derived from the DM curve (e.g. possible peaks and amplitudes at different field strengths) can be used as complementary information in an extended 3MA approach, where DM is used as a fifth sub-method. It has been shown in the past that this additional information can significantly increase the calibration accuracy [12].

**EXPERIMENTS AND RESULTS**

Many power plant components are permanently exposed to varying thermal and thermo-mechanical loads as well as radiation. Besides long-term exposure to temperatures typically between 200 and 350 °C, additional loads occur during startup and shutdown of the plant. The last-mentioned effect can be expected to gain importance as the role of renewable energy grows, since their non-constant availability (wind, sunlight) leads to higher and faster load variations in fossil and nuclear power plants. Even under constant operating conditions, elbows and T-joints of pipes are particularly concerned because of steep thermal gradients caused by inhomogeneous flow velocity or different media temperatures. Components exposed to neutron radiation (reactor pressure vessels) may show an increasing number of dislocations and point defects that build up over time (neutron embrittlement), but also pronounced Cu precipitates [13].

The experiments described in the following sections demonstrate that electromagnetic and ultrasonic NDE can provide the information required to quantify and better understand aging of component materials and detect imminent damage in time.

**Characterization of Aged Reactor Pressure Vessel Materials**

A set of neutron-irradiated reactor pressure vessel (RPV) base and weld materials of western and eastern design was provided by the project partners Areva NP GmbH and Helmholtz-Zentrum Dresden-Rossendorf e.V, Germany. In collaboration with these partners, a set of Charpy V-notch samples was characterized in a hot cell using 3MA and EMAT-based dynamic magnetostriction analysis. The neutron fluence of each sample was known, and the shift of
the ductile-brittle transition temperature (DBTT) relative to the pristine condition was determined by the partners in Charpy tests. Table 1 lists the materials investigated.

The samples were split up into calibration and validation samples. 3MA and dynamic magnetostriction data were recorded for all samples. Using a pattern recognition approach and a genetic optimization algorithm specifically developed for this task [14], it was shown that neutron fluence and DBTT shift of all samples could be quantitatively determined with a standard error comparable to the one of the reference data. Separate calibrations had to be performed for base and weld materials. Fig. 3 shows the results for the base material calibration only. The calibration with weld materials achieved a comparable accuracy.

**TABLE 1.** List of neutron-irradiated base and weld materials

<table>
<thead>
<tr>
<th>Material</th>
<th>Base (B) / Weld (W)</th>
<th>Acronym</th>
<th>Fluence [n/cm²]</th>
<th>DBTT shift ΔT₄¹ [K]</th>
</tr>
</thead>
<tbody>
<tr>
<td>22NiMoCr3-7</td>
<td>B</td>
<td>P7 GW*¹</td>
<td>0 – 53.8 x 10¹⁸</td>
<td>0 – 32</td>
</tr>
<tr>
<td>20MnMoNi5-5</td>
<td>B</td>
<td>P141 GW*¹</td>
<td>0 – 10.8 x 10¹⁸</td>
<td>0 – 9</td>
</tr>
<tr>
<td>22NiMoCr3-7</td>
<td>B</td>
<td>P147 GW*¹</td>
<td>0 – 46.2 x 10¹⁸</td>
<td>0 – 23</td>
</tr>
<tr>
<td>ASTM A508 C1.3</td>
<td>B</td>
<td>JFL*²</td>
<td>0 – 86.7 x 10¹⁸</td>
<td>0 – 78</td>
</tr>
<tr>
<td>ASTM A533B C1.1</td>
<td>B</td>
<td>JRQ*²</td>
<td>0 – 98.2 x 10¹⁸</td>
<td>0 – 222</td>
</tr>
<tr>
<td>S3 NiMo 3 / OP 41 TT</td>
<td>W</td>
<td>P16 SG*¹</td>
<td>0 – 11.8 x 10¹⁸</td>
<td>0 – 67</td>
</tr>
<tr>
<td>S3 NiMo 1 / OP 41 TT</td>
<td>W</td>
<td>P141 SG*¹</td>
<td>0 – 14.0 x 10¹⁸</td>
<td>n/a</td>
</tr>
</tbody>
</table>

*¹ western design  *² eastern design

**FIGURE 3.** Correlation between nondestructively determined and reference values of the shift in the ductile-brittle transition temperature (ΔT₄¹) and the experienced neutron fluence of different reactor pressure vessel base materials

In order to ensure that the application of 3MA and EMAT is feasible under realistic conditions, i.e. for determining the state of the base material or the weld of an RPV from the inside through an 8 mm austenitic cladding layer. The measurements were performed at MPA Stuttgart, Germany, inside an actual RPV that was left over from the planned and started but finally aborted construction of block C of the Biblis nuclear power plant.

Regarding the high penetration depth required for the measurement through the cladding, a large 3MA/EMAT combination probe for low-frequency inspection was developed and tested. The pole pieces of the electromagnet had a cross-section of 25 x 25 mm² and 25 mm spacing (fig. 4, to the left). For magnetic Barkhausen noise analysis, four pancake-shaped coils were connected in series and placed between the poles, where a Hall probe was mounted as well. The electromagnet had a dual use in this probe, since it also acted as EMAT besides being the magnetic field source for 3MA. The magnetization frequencies were in the order of 10 – 100 Hz, and typical burst frequencies were a few kHz. A Crown I-T6000 stage amplifier connected to a National Instruments data acquisition board was used in order to drive the electromagnet with a superimposed waveform for magnetization and sound generation at the same time. In order to achieve high reception efficiency in this (audible) frequency range, a low-frequency (0.5 Hz – 30
kHz) piezo transducer was integrated into the sensor and pressed on the material surface during the measurement. In this frequency range, a coupling agent is not required.

The first task was to detect the location of the weld. This was accomplished by means of 3MA measurements along the RPV wall. It was observed that the coercivity of the weld had an apparent coercivity $H_C$ approximately 10 A/cm higher than the one of the base material. Since the cladding separated the Hall probe from the base material, the actual $H_C$ difference might be different. The locations of weld and base material were marked on the RPV wall, and dynamic magnetostriction measurements were performed on two spots over the weld and two spots over the base material. Figure 4 (right hand side) shows the $E(H)$ plots obtained at a magnetization frequency of 57 Hz and a burst frequency of 3.36 kHz. The base material showed a significantly higher dynamic magnetostriction compared to the weld. The S/N was sufficient to distinguish both materials clearly.

**FIGURE 4.** RPV sensor prototype (photo to the left), dynamic magnetostriction curves of weld and base material, measured through an approximately 8 mm thick austenitic cladding layer. The error bars indicate the standard deviation of the samples and were only shown once for improved visibility.

**Low-Cycle Fatigue of WB 36**

The Cu-rich ferritic steel WB 36 (1.6368, 15NiCuMoNb5) is an excellent model material to study and understand aging effects related to neutron radiation without the challenge of handling radioactive specimens in a hot cell. It contains 0.65 wt.% Cu, approximately half of which is in solid solution in the pristine state. Under long-term thermal service exposure, Cu is precipitated from the solution. The fine-disperse precipitates lead to increased hardness and DBTT as well as decreased upper shelf energy. This effect and the applicability of micromagnetic NDE for its characterization have been thoroughly studied [2–4].

Under practical conditions, both thermal and mechanical loads act upon the material. In order to better understand the role of copper precipitation in a thermo-mechanical aging scenario, low-cycle fatigue (LCF) experiments were conducted on samples of WB 36 at different temperatures from RT to 350 °C. A strain amplitude of $\varepsilon = 1.05\%$ and a cycle duration of $T = 2400$ s were used. The samples were machined and fatigued at MPA Stuttgart, Germany. Each LCF series was interrupted after having reached predefined numbers of load cycles. The samples were then characterized with a compact 3MA device at IZFP [15] (fig. 5). After each micromagnetic test, the LCF loading continued. This procedure was repeated up to fracture appearance.

Figure 6 shows the harmonic distortion factor $K$ as a function of the number load cycles for a recovery-annealed material (E59 EG) and heat-treatment stabilized material (E59 S4). The stabilization was achieved by annealing the sample with a special temperature profile which leads to maximal Cu precipitation in form of large agglomerates which have negligible effect on the mechanical properties. Almost no additional Cu precipitation is possible in this state. $K$ is a result quantity of harmonics analysis and is sensitive for Cu precipitation, showing a decrease as the Cu precipitation proceeds.
In case of E59 EG, most of the total effect is already reached at 160 load cycles. It was found that the additional mechanical fatigue accelerates the precipitation process significantly and increases the amplitude of the effects, compared to thermal aging alone. The strongest effect was observed at 300 °C. At RT, nearly no effect was observed. The stabilized material E59 S4, however, shows a small, steady decrease of $K$ throughout the full experiment. Since the amount of precipitable Cu is the only difference here, this proves that the steep decrease of $K$ in the beginning of the E59 EG experiments occur due to Cu precipitation. This was also confirmed by reference measurements at MPA Stuttgart. Moreover, the results suggest that Cu precipitation is the predominant aging mechanism in WB 36.

**FIGURE 5.** Compact 3MA device variant with LCF sample. The device is included in the sensor enclosure and is directly connected to the PC via USB connection. An integrated power amplifier requires an external 48 V power supply.

**FIGURE 6.** Harmonic distortion factor $K$ of WB 36 as a function of LCF cycles experienced at a strain amplitude of $\varepsilon = 1.05\%$. The graph to the left shows the results for initially recovery-annealed material (E59 EG). The graph to the right shows the results for material stabilized by promoted precipitation through heat-treatment (E59 S4).
Low-Cycle Fatigue of X6CrNiNb18-10

Metastable austenitic steels with reduced carbon and enhanced Ni and Nb content cannot be characterized with 3MA due to the absence of ferromagnetic behavior at service temperatures (300 °C). In such cases, EMAT-based ultrasonic analysis can obtain useful information on the material state. The LCF behavior of AISI 347 steel (1.4550, X6CrNiNb18-10) was characterized in a conventional LCF setup, but with EMATs attached to both sample faces (fig. 7, sketch and photo). The samples were machined and fatigued at the WKK chair, Technical University of Kaiserslautern, Germany.

The experiment was performed using an LCF cycle duration of $T = 100\, \text{s}$ and strain-controlled conditions with strain amplitudes of 0.8%, 1.0%, 1.2% and 1.6%. An EMAT burst frequency of 800 kHz was used. The samples were heated to a temperature of up to 300 °C using an inductive heater. A radially polarized transverse wave was generated by the EMAT transmitter. After travelling through the sample, the ultrasonic wave was converted to a voltage signal by the EMAT receiver at the other end of the sample. The time-of-flight (TOF) was measured in a selected peak of the received ultrasonic signal using a precision peak detector and tracking algorithm. TOF variations due to applied load stress were filtered out by means of averaging. Fig. 7 shows the results for 300 °C.

An overall increase in TOF was observed in all conditions. This can be explained with an increasing number of micro-defects which impede the propagation of the ultrasonic wave. Since the experiments were strain-controlled, the average length of the sample remained constant throughout the experiment and varied only within each cycle, so any long-term TOF changes must be due to microstructure effects alone. TOF shows a final steep increase after having gone through a plateau and a slight decrease over most of the experiment. Most remarkable and important for an early recognition of imminent damage is that this final increase in TOF starts significantly before the final drop in stress amplitude recorded by the testing machine, thus indicating the nearing failure in time. The observation is explained with a build-up of micro-cracks in the final phase and their interaction with the ultrasonic wave.

FIGURE 7. Schematic representation of transmitter (T) and receiver (R) positions on the sample faces, direction of load stress $\sigma$ and ultrasonic wave (to the left), photo of the mounted sample, including an inductive heater, a clip gauge extensometer and electrical contacts for resistometry (in the center, EMATs not visible, image: WKK, TU Kaiserslautern), and comparison of TOF change and stress amplitude for experiments performed at 300 °C and different strain amplitudes from 0.8 to 1.6 % (to the right)

CONCLUSION

Aging effects in power plant component steels were characterized using micromagnetic and dynamic magnetostriction analysis as well as ultrasonic time-of-flight measurements with EMATs. The materials investigated
were exposed to different kinds of aging, including low-cycle fatigue and neutron irradiation. It was shown that the applied NDE methods are valuable tools for understanding the aging processes and determining their progression. Micromagnetic 3MA analysis helped prove and understand the significant role of copper precipitation in thermo-mechanical fatigue of 15NiCuMoNb5 steel (WB 36). Reactor pressure vessel (RPV) materials representing the globally different standards were investigated with a combination of 3MA and dynamic magnetostriiction analysis in order to indirectly determine the change of the ductile-brittle transition temperature and the experienced neutron fluence. In this context, experiments were also conducted in an unused reactor pressure vessel in order to collect proof that information on the base material can be obtained from the cladded inner surface with a cladding thickness of 8 mm. Finally, the fatigue behavior of austenitic X6CrNiNb18-10 steel was investigated with ultrasonic time-of-flight (TOF) measurements in-situ during conventional LCF tests. It was found that the TOF indicates imminent material failure significantly earlier than the stress amplitude measured by the testing machine does.

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