

**Nondestructive Characterization of Neutron Induced Embrittlement in Nuclear Pressure
Vessel Steel Microstructure by using Electromagnetic Testing**

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ABSTRACT

By using nuclear power for energy generation the pressure vessel wall is exposed to neutron fluences of different levels depending on the distance to the core. Hence materials undergo a change in their microstructure in terms of embrittlement, to be measured as toughness reduction and shift of the Ductile-to-Brittle Transition Temperature (DBTT) to higher temperatures.

Normally plant safety concerning this change in microstructure is assured by destructively testing surveillance samples. These are standard tensile and ISO V-specimen which consist of exactly the same material as the pressure vessel and its weld metal, being exposed to accelerated irradiation rates within special irradiation channels allowing a pronounced ageing.

It is demonstrated that electromagnetic parameters allow to characterize the changes in the microstructure generated through neutron irradiation. After a defined calibration process a quantitative characterization of the embrittlement especially in terms of the shift of the DBTT is possible. This has been demonstrated for reactor pressure vessel steels used in nuclear power plants of eastern and western designs. As testing methods 3MA (Micromagnetic, Multiparameter, Microstructure and stress Analysis) [1] and the dynamic magnetostriction using EMAT (Electromagnetic Acoustic Transducers) have been applied in a nondestructive combination measurement system.

Further experiments show the possibility to measure 3MA and dynamic magnetostriction quantities through an 8 mm thick austenitic stainless steel cladding.

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 such as mechanical hardness, yield and tensile strength, toughness, shift of Ductile-to-Brittle Transition Temperature (DBTT), fatigue strength. With the exception of hardness tests which are weakly invasive, all of these parameters can be determined within surveillance programs by using destructive tests on special standardized samples (Charpy V samples). The specimens are exposed in special radiation chambers near the core of the Nuclear Power Plant (NPP) to 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. The number of the Charpy samples is limited and in the future it will be very important that nondestructive methods are available to determine the mechanical material parameters on these Charpy samples. Furthermore in situ characterization of the reactor pressure vessel inner wall through the cladding is of interest additionally to the measurements on Charpy samples.

To solve this task a combination testing method based on 3MA (Micromagnetic, Multiparameter, Microstructure and stress Analysis) [2] and dynamic magnetostriction by using an EMAT (Electromagnetic Acoustic Transducers) [3] was developed. Electromagnetic methods have a high potential to characterize neutron induced microstructure states.

Assessment of the Embrittlement

In addition to the mechanical properties, the embrittlement phenomenon also influences the magnetic properties. Even the 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, e.g. precipitations, since 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. The neutron induced embrittlement results in microstructure changes. These microstructure changes lead to the generation of vacancies and precipitations of Cu-rich coherent particles (radius: 1-1.5 nm). This results in an increase of yield strength and tensile strength, a decrease of Charpy energy upper shelf value and an increase of DBTT. The potential of electromagnetic testing methods for detection of microstructure states is based on the analogy between the interaction of dislocations with microstructure states and of Bloch-walls with microstructure states. Similar to the movements of dislocations under mechanical load Bloch-walls move under magnetization. Therefore an analogy exists between mechanical technological values (e.g. hardness values) and magnetic values. So the interaction between dislocations and copper particles leads to an increase of mechanical hardness and the interaction of the copper particles and Bloch-walls leads to an increase of magnetic hardness. The potential of electromagnetic testing methods for the detection of copper precipitations was demonstrated on the Cu-rich pressure vessel and piping steel 15 NiCuMoNb 5 (1.65 weight % Cu) [4].

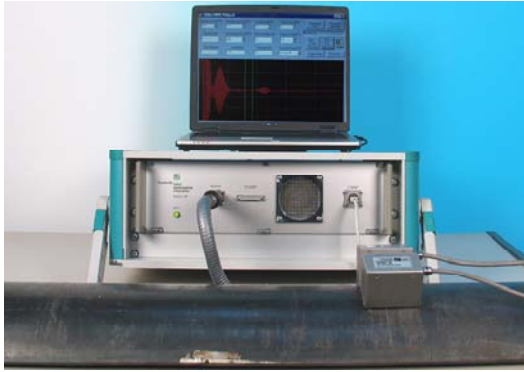
Since the dynamic magnetostriction is sensitive for lattice defects it was assumed that a magnetostrictively excited standing wave in the pressure vessel wall also reflects the neutron embrittlement and first experiments were performed with a special designed magnetostrictive transducer at Charpy specimens in the hot cell in order to principally demonstrate the potential.

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 efficiently compared to individual measurement. By using a calibration function or pattern recognition the desired quantity of an unknown set of samples investigated by that method can be detected nondestructively.

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 is exposed to neutron fluences in the range between 5.6×10^{18} n/cm² and 86.0×10^{18} n/cm². In order to characterize the neutron irradiation-induced embrittlement, Charpy samples exposed to neutron fluence in the above mentioned range have been investigated in a hot cell at AREVA NP GmbH whereby only the 3MA and EMAT sensors were arranged within the hot cell and the electronic equipment (3MA and EMAT device) was outside (see Figure 1). These Charpy samples (base material and weld material) of eastern and western design have been provided by AREVA NP GmbH and Research Centre Dresden-Rossendorf e.V. (see Table 1).

EMAT testing device



3MA testing device

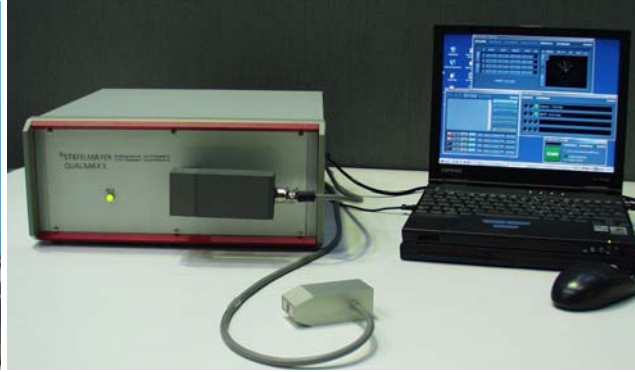


Figure 1 - Used equipment for electromagnetic measurements in the hot cell

Sample set of neutron irradiated base material

Material	Short term	Neutron-fluence [n/cm ²]	Range ΔT_{41} [K]
22 NiMoCr 3 7	P7	0 – 5.38 x 10 ¹⁸	0 - 32
20 MnMoNi 5 5	P141	0 – 10.7 x 10 ¹⁸	0 - 9
22 NiMoCr 3 7	P147	0 – 44.0 x 10 ¹⁸	0 - 23
ASTM A508 C1.3 (22 NiMoCr 3 7)	JFL	0 – 86.0 x 10 ¹⁸	0 - 78
ASTM A533B C1.1 (20 MnMoNi 5 5)	JRQ	0 – 98.0 x 10 ¹⁸	0 - 221

Sample set of neutron irradiated weld material

Material	Short term	Neutron-fluence [n/cm ²]	Range ΔT_{41} [K]
S3NiMo3-UP	P16	0 – 11.6 x 10 ¹⁸	0 - 67
S3NiMo1-UP	P141	0 – 13.9 x 10 ¹⁸	0 - 38
S3NiMo1	P140	0 – 10.4 x 10 ¹⁸	0 - 21

Table 1 - Overview of the set of neutron induced Charpy specimens (base and weld material)

Furthermore Charpy tests were carried out for determination of the DBTT (reference values).

Based on the data base consisting of EMAT measuring quantities and the reference values, model functions were developed using pattern recognition and regression functions. One part of each Charpy specimen set was used to calibrate and the other part – independently selected – was taken to test the model. The pattern recognition algorithm, mentioned above, was used to obtain approximation values of the shift of DBTT, which is a measure of the embrittlement (see Figure 2 and Figure 3).

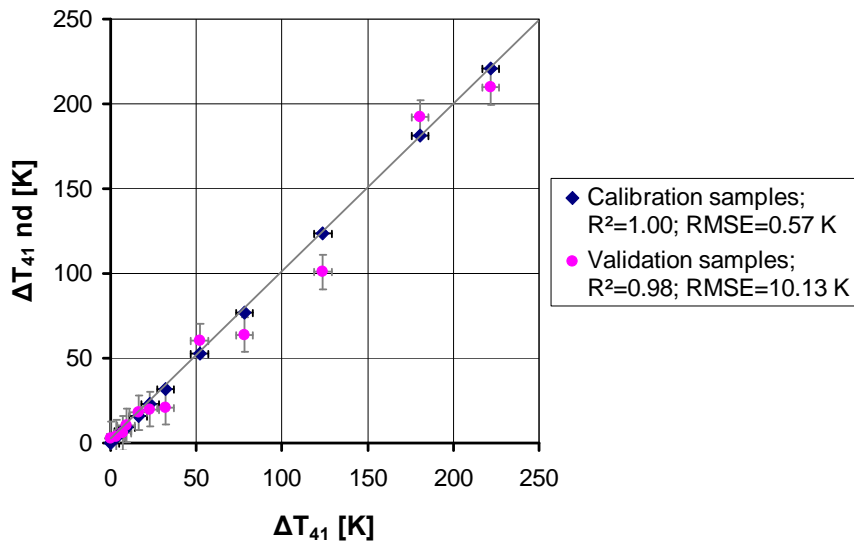


Figure 2 - Predicted shift of the DBTT for Ractor Pressure Vessel (RPV) -base material versus shift of DBTT determined by the Charpy test

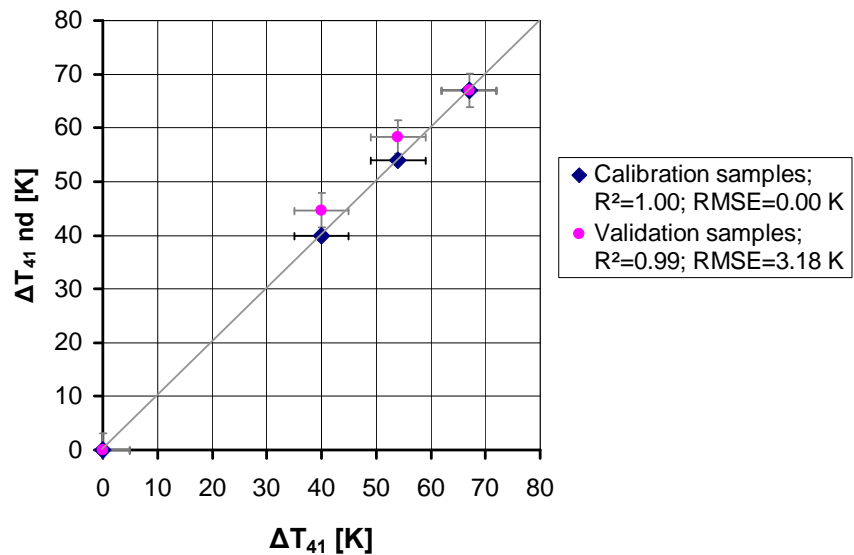


Figure 3 - Predicted shift of the DBTT for RPV- weldments versus shift of DBTT determined by the Charpy test

It was demonstrated that the prediction of the shift of the DBTT can be performed due to the micro-magnetic procedure. For the validation samples (base material) a correlation coefficient of $R^2=0.98$ and a RMSE (root mean square error) of 10.13 K was obtained. In the case of the weld material the correlation coefficient was $R^2=0.99$ and the RMSE value was 3.18 K.

With regard to in situ measurements on the reactor pressure vessel inner wall through the austenitic cladding, test measurements with 3MA and the dynamic magnetostriction-technique were carried out at a ferritic test piece with 8 mm thick austenitic cladding.

In order to ensure deep penetration of the electromagnetic fields, low-frequency EMATs (≤ 40 kHz) were applied, and hence it was possible to characterize the material condition in the ferritic base material directly below the austenitic cladding from the clad side.

Figure 4 shows the principle of such an inspection method [5] which up to now is not yet realized for in-service inspection.

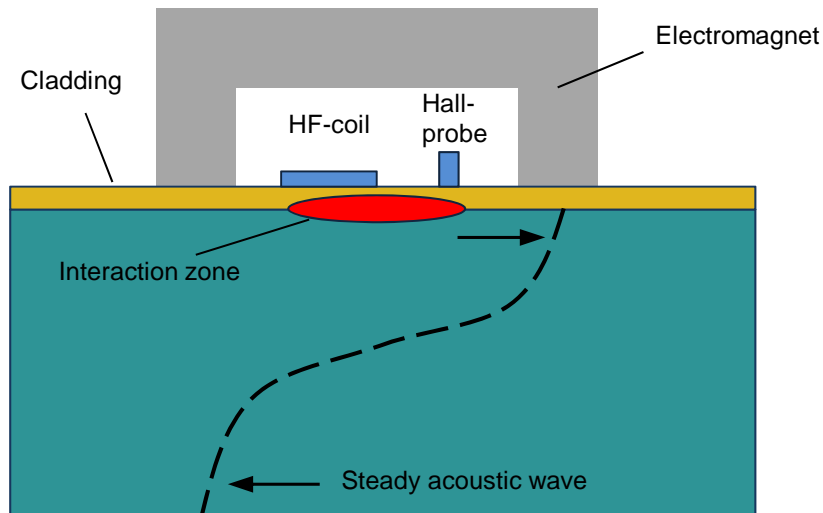


Figure 4 - Principle of the magnetostrictive excitation of a standing wave in thickness direction of the pressure vessel wall by using an EMAT

The possibility of exciting and receiving ultrasonic waves in the ferritic base material through the cladding was documented by using HF-coil-configurations on clad test pieces by superimposing magnetic fields excited by u-shaped electromagnets. Values of signal-to-noise ratio of 23 dB were obtained on a ferritic test object with a wall thickness of 30 mm and with an austenitic cladding thickness of 8 mm. The magnetizing field strength was 260 A/cm and the selected frequency of the EMAT was 40 kHz.

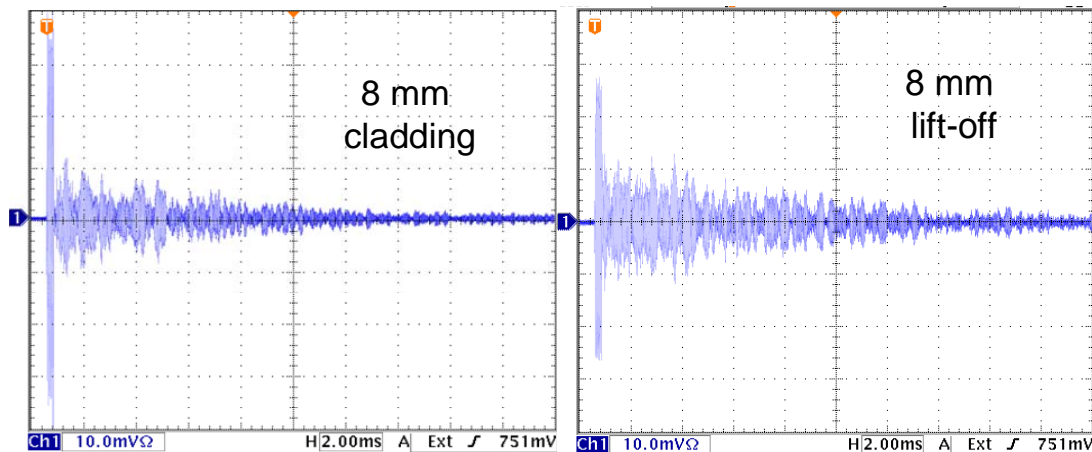


Figure 5 - Ultrasound signals by an excitation on the austenitic clad side (left); Ultrasound signals by an excitation on the side without cladding with 8 mm lift-off (right)

In a second experiment it was demonstrated that ultrasonic signals can still be produced even with a coil lift-off of 8 mm which was achieved by using an 8 mm thick PVC plate. The obtained EMAT-echo signals are presented in Figure 5. Comparing the two acoustic signals it is obvious that the cladding is influencing the excitation of the magnetostrictively generated ultrasonic wave mainly by lift-off and not by eddy-current damping.

Furthermore it also was shown that magnetic Barkhausen noise signals can also be received through the austenitic cladding with a good signal to noise ratio.

Therefore the non-destructive detection of the neutron irradiation by using 3MA techniques and the dynamic magnetostriction in a combination should be realized by developing an optimized transducer in the near future. The presence of the austenitic cladding is not a strong disturbance effect.

Furthermore it was demonstrated that the δ -ferrite changes in the cladding as well as a butt weld beneath the cladding do not disturb the ultrasonic amplitude very much (Figure 6).

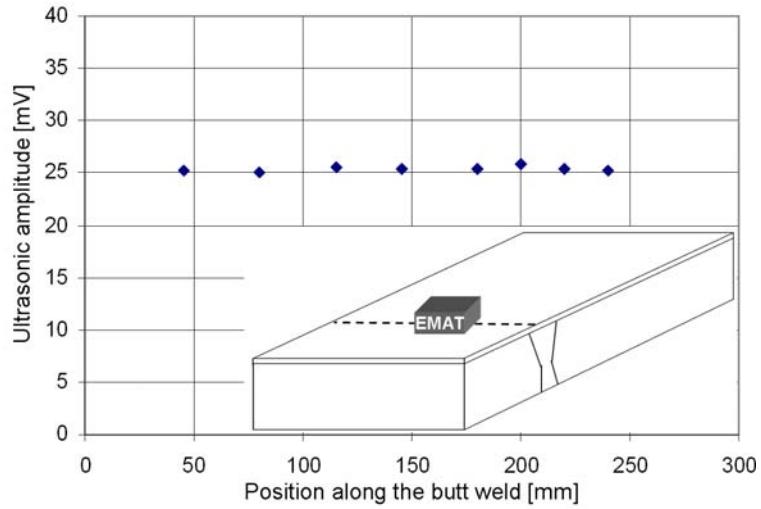


Figure 6 - Variation of the ultrasonic signal amplitude along a butt weld

Additionally typical dynamic magnetostriction curves have been recorded at a sound frequency of 4 kHz at the clad specimen (wall thickness: 150 mm, austenitic cladding thickness: 12 mm) of Figure 6. There exist characteristic differences between the curves measured at the butt weld (left side Figure 7) and the curves measured at the ferritic base material (right side Figure 7). Thanks to these curve differences it is possible, through the austenitic cladding, to differentiate the base material from the weldment by means of nondestructive testing.

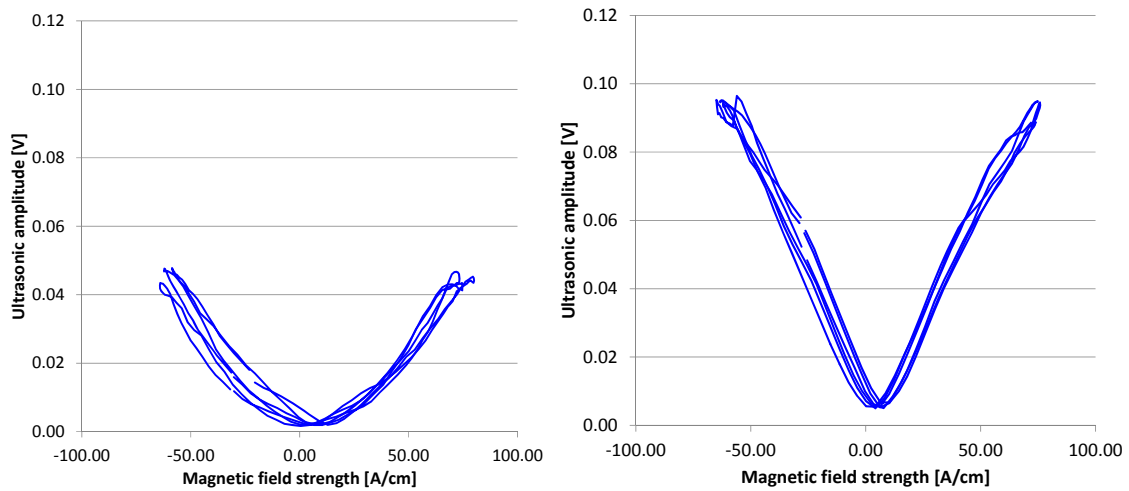


Figure 7 - Three Typical dynamic magnetostriction curves for RPV - weldment (left side) and base material (right side)

CONCLUSION

By combining 3MA measured quantities with those derived from dynamic magnetostriction measurements using an EMAT the shift of DBTT on irradiated Charpy samples of base and weld metal materials of reactor pressure vessels of western and eastern design can be determined nondestructively.

3MA- and EMAT-signals can be measured through an austenitic cladding. On the basis of these results the development of an in-service inspection technique is possible which allows to determine the shift of the DBTT nondestructively directly on the inner diameter surface of the reactor pressure vessel.

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REFERENCES

- 1) Dobmann G, Altpeter I, Becker R, Kern R, Laub U, Theiner W A, “Barkhausen Noise Measurements and related Measurements in Ferromagnetic Materials”, Topics on Non-destructive Evaluation Series Volume 1, Sensing for Materials Characterization, Processing, and Manufacturing (Birnbauer G, Auld B), American Society for Non-destructive Testing Inc., 1998, 233-251.
- 2) Altpeter I, Becker R, Dobmann G, Kern R, Theiner W A, Yashan A, “Robust solutions of inverse problems in electromagnetic non-destructive evaluation”, Inverse Problems, 2002, 18, 1907-1921.
- 3) Wolter B, Dobmann G, “Micromagnetic testing for rolled steel”, 9th European NDT Conference ECNDT 2006, Berlin, 2006.
- 4) Altpeter I, Dobmann G, Katerbau K H, Schick M, Binkele P, Kizler P, Schmauder S, “Copper Precipitates in 15 NiCuMoNb 5 (WB 36) Steel: Material Properties and Microstructure, Atomistic Simulation and Micromagnetic NDE Techniques” Nuclear Engineering and Design, 2001, 206, 337-350.
- 5) Altpeter I, Dobmann G, Hübschen G, Kröning M, “Verfahren zur Messung der Neutronenversprödung unterhalb der Plattierung von Druckbehältern“, patent-no.: WO 2008/135054.