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Non-destructive evaluation of material degradation of nuclear reactor pressure vessels

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Abstract

This paper is introducing a new method for measuring the irradiation-induced embrittlement of low alloy steels. The method is based on highly accurate resistivity measurements using the direct current potential drop (DCPD) method. The method was tested on Charpy specimens made from four different steels: 18MND5, 22NiMoCr37, A533b and A508Cl3. These steels are used for nuclear reactor pressure vessels in Eastern and Western Europe. The ductile-brittle transition temperature difference (ΔDBTT) was measured using the Charpy impact test.

The results show a correlation between measured resistivity and ΔDBTT for three out of four materials. Scatter and number of specimens can to limit the accuracy in predicting ΔDBTT using resistivity measurements.

1. Introduction

The nuclear reactor pressure vessel (RPV) is a large thick-walled container made of low alloy steel. In service, he RPV wall is exposed to high levels of neutron and gamma radiation from the reactor core. The neutron radiation impacts with atoms of the RPV, causing microstructural damage and embrittlement of the steel.

Currently, the embrittlement of RPVs is monitored with surveillance programs. The specimens of the RPV steel are put inside the RPV and exposed with similar irradiation condition. During the periodic safety review, some of these specimens are taken out from the reactor surveillance capsule position and tested destructively e.g. with Charpy impact tests [1,2]. These destructive testing methods are well-known and reliable. The nuclear power plant (NPP) has an assumed lifetime, and this is the basis for the RPV surveillance program [3]. Now it has been realized that after the end of the planned lifetime, extension is possible when the plant is still in a good condition and safe to use. This has led to the need for new test specimens, which can be prepared by reconstitution and miniaturization of old test specimens. Those specimens are then re-irradiated to a higher dose for prolonged safety analysis. If it is possible to find accurate, validatable and verifiable correlation between a non-destructively measureable material parameter and fracture mechanical properties, one could use it instead of destructive testing. For this reason, the development of new non-destructive methods has been started to monitor the embrittlement of RPV.

Ductile-brittle transient temperature difference (ΔDBTT) is one of the most important parameters to be determined during periodic safety reviews. The fracture mechanical properties of steels depend on temperature. In temperatures lower than DBTT, steel structures have much greater tendency to shatter on impact

instead of bending or deforming. Neutron irradiation tends to increase DBTT and therefore make steel structures brittle at higher temperatures [4].

There are no methods to measure the relevant microstructural properties of RPV material directly and non-destructively. However, there are several physical measurable material properties, which are related to the microstructure. According to microstructural models for resistivity (e.g. Drude model or Drude-Sommerfeld model), resistivity of metal is a function of the mean distance between atoms in the lattice [5]. This distance is changed during irradiation. Therefore, to the extent it is related to the associated shift in the applied measure of toughness, the change in resistivity can be used as an indicator of irradiation-induced embrittlement.

2. Direct current potential drop

Potential drop (PD) methods provide techniques developed for non-destructive evaluation of electrically conductive materials. The working principle of PD is to drive electrical current through a specimen or component and to measure the electrical potential difference between two points. For example, a higher than expected potential difference could indicate a defect between the two measurement points.

The two basic variations of the method are the direct current potential drop (DCPD) and alternating current potential drop (ACPD). ACPD is more sensitive than DCPD to detect near surface or surface defects. DCPD is more suitable for detecting embedded flaws. Because resistivity is a volumetric material property, DCPD is more suitable for measuring it than ACPD.

The measured electrical potential difference depends on three factors: electrical conductivity of the material, geometry of the specimen and location of the two measurement points. If the geometry of the specimen and location of the two measurement points are known, and electrical conductivity is homogeneous, conductivity can be calculated if the electrical potential difference is measured with high accuracy [6].

2.1 Measurement method

The measurement method was based on the same traditional four-point resistance measurement as DCPD. A well-defined constant current was conducted to the measured specimens using spring-loaded needle probes. The same type of needle probes were used for measuring the potential difference of the two measurement points.

Keithley Model 6621 AC+DC source was used as a current source. The output impedance of the current source was higher than 10^{14} Ω. This ensures well-defined constant current for the measurement. The electrical potential difference was measured using Keithley Model 2182A Nanovoltmeter. The noise amplitude of the device was 5 nV. The measured potential differences were in the order of 10 µV, therefore providing a four-decade difference between noise and signal levels.
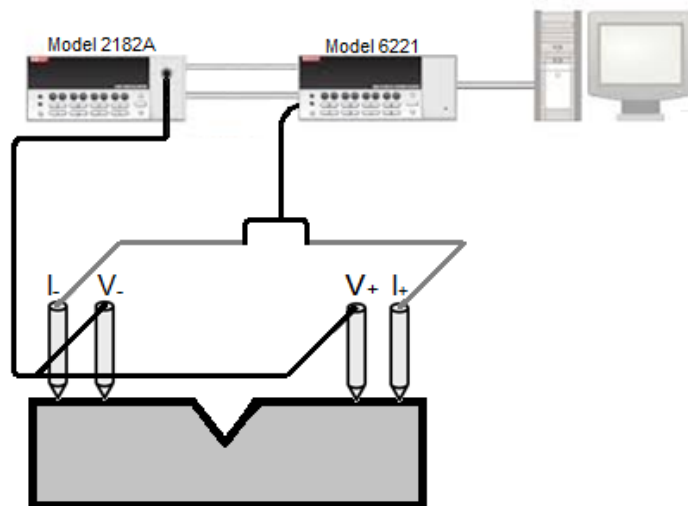
Figure 1. Measurement setup. The current feed into the specimen was through spring-loaded needle probes. The potential difference was measured using similar needle probes. The distance between two measurement points was 40.64 mm.

Since the electric current was stable with low drift, it was possible to calculate the resistance directly using the Ohm's law as a ratio of measured potential difference and predefined electric current. The measurement was repeated 10 000 times for each specimen. The relative accuracy uncertainty? of the indicated resistance was of the order of 0.1‰.

Each specimen was assumed to be homogenous. Thus, it was assumed that resistivity is given by multiplying the resistance (R) with a ratio of cross-section area (A = 10 mm x 10 mm) and the distance between two measurement points (l = 40.64 mm) using equation (1).

$$\rho = R\frac{A}{l} \tag{1}$$

Because the specimens were transformed radioactive due to neutron irradiation, the measurements were conducted in a hot-cell environment. Instead of measuring the distance between two measurement points in hot-cell for each specimen, a special specimen holder was designed. This specimen holder ensured the dimensional repeatability and accuracy of the measurements.

3. Specimens and irradiation conditions

Four different steel alloys used in nuclear reactors both in Eastern and Western Europe were studied: 18MND5 (Eastern), 22NiMoCr37 (Eastern), A533b (Western) and A508Cl3 (Western) (Table 1). The specimens were divided into groups for irradiation. Each group was irradiated under specific conditions regarding temperature and neutron fluence that were monitored during the irradiation process. The specimens were irradiated in the BR-2 reactor at the Belgian Nuclear Research Centre SCK•CEN. One group of each material remained unirradiated for reference. All specimens were ISO-standard based Charpy impact test specimens [2].

Table 1. Irradiation conditions of the specimens in different groups and corresponding ∆DBTT values.

| Material	Group	Temperature (°C)	Fluence (1/cm²)	∆DBTT (°C)
18MND5	Group 1	150	3.6E+19	183
	Group 2	260	4.8E+19	136
	Group 3	260	9.4E+19	218
	Reference	-	-	-
22NiMoCr37	Group 1	260	3.4E+19	52
	Group 2	260	5.7E+19	85
	Reference	-	-	-
A533b	Group 1	150	4.1E+19	196
	Group 2	305	4.2E+19	42
	Reference	-	-	-
A508Cl3	Group 1	150	2.7E+19	157
	Group 2	305	4.1E+19	34
	Reference	-	-	-

4. Results

The resistivity and ∆DBTT values have a clear correlation in three materials out of four. A clear drop in resistivity can be seen for materials 18MND5, A533B, and A508Cl3 (Figures 2, 4 and 5). However, the scatter of the results is still rather strong.

Unlike in all the other materials, no significant change in resistivity can be seen for material 22NiMoCr37 (Figure 3). The other notable difference between 22NiMoCr37 and the other materials is the relatively small ∆DBTT even with high fluence. The rest of the materials suffer twice as large irradiation embrittlement in terms of ∆DBTT.
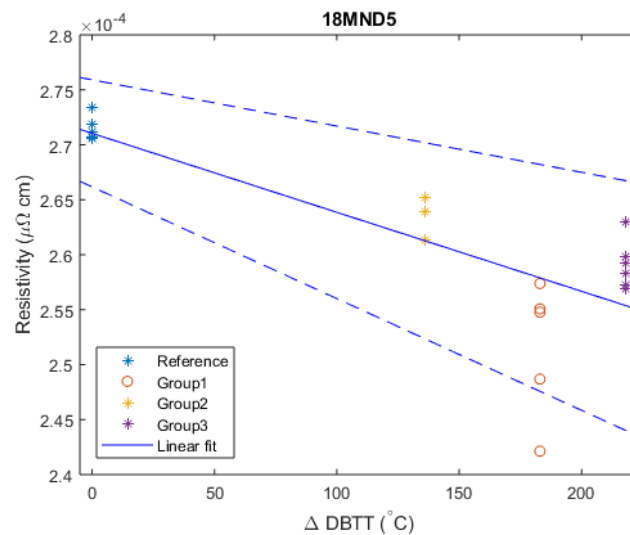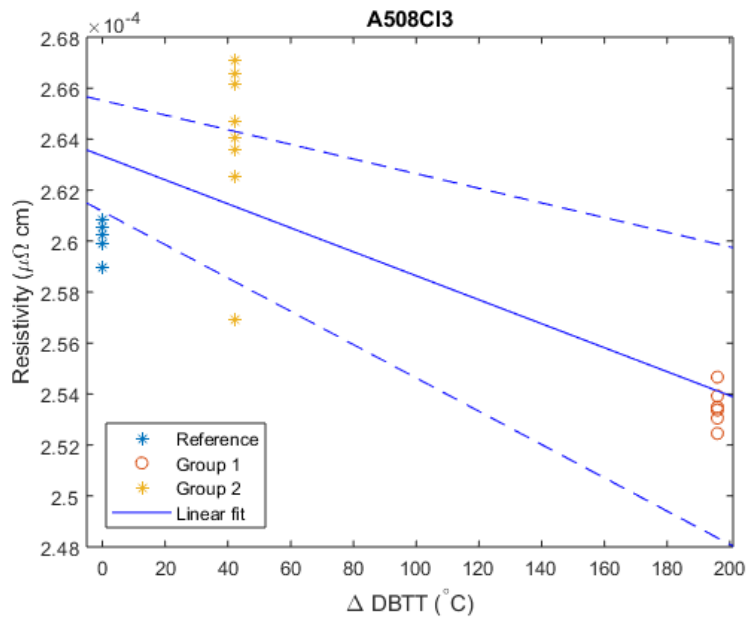

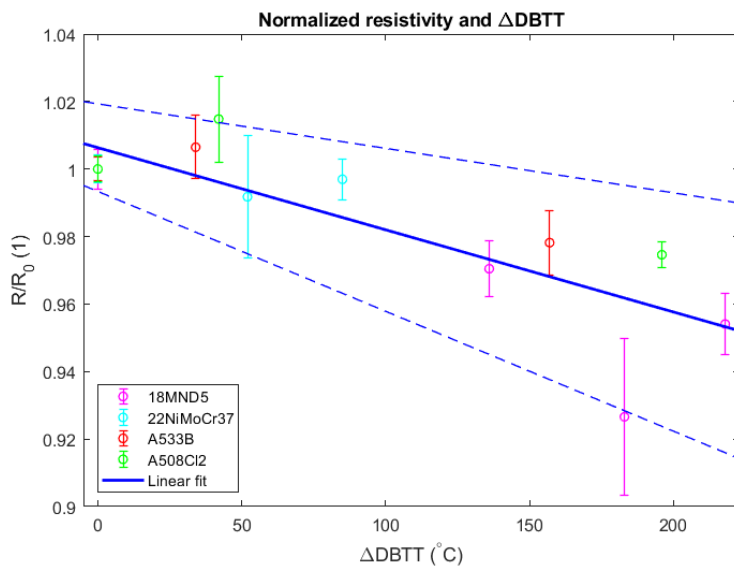**Figure 2.** Resistivity and ∆DBTT for 18MND5. A clear correlation between resistivity and ∆DBTT can be seen.
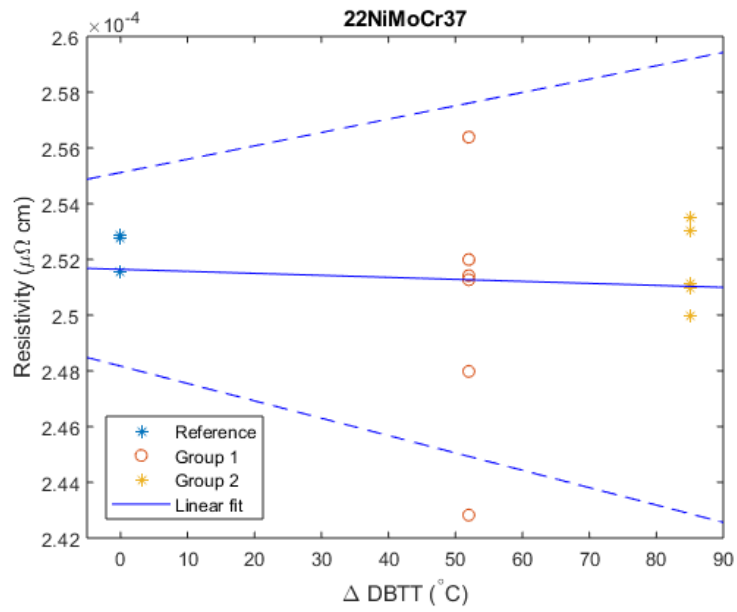
Figure 3. Resistivity and ΔDBTT. For 22NiMoCr37 no correlation between resistivity and ΔDBTT can be seen. However, the ΔDBTT for the highest level of irradiation is significantly smaller than for other materials studied.
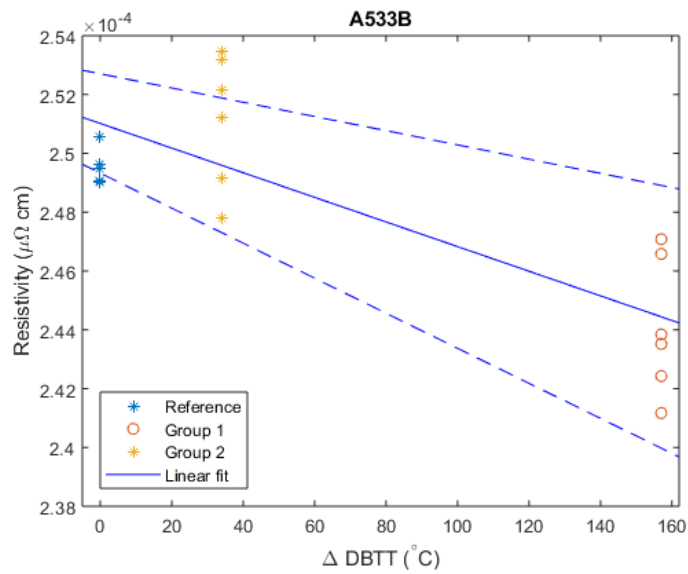
Figure 4. Resistivity and ΔDBTT for A533b. A clear correlation between resistivity and ΔDBTT can be seen.

Figure 5. Resistivity and ΔDBTT for A508Cl3. A clear correlation between resistivity and ΔDBTT can be seen.

Figure 6. Normalized resistivity and ΔDBTT for each each material and sample group. Clear drop in resistivity can be seen as a function of DBTT.

5. Discussion

The proposed method is a promising new candidate for non-destructive monitoring irradiation-induced embrittlement of RPV steels. In spite of the associated scatter, the relative resistivity drop appears to indicate significant ΔDBTT for those three materials out of four that showed largest embrittlement.

The correlation between resistivity and ΔDBTT was weakest for the material 22NiMoCr37, apparently because for this material the ΔDBTT value for highly irradiated specimens remained modest (approx. 90 °C) compared to the other three materials (170 - 200 °C).

The proposed method can be utilized in parallel with the existing surveillance programs. It is suggested that additional information be collected by measuring the resistivity of CVN surveillance specimens before

mechanical tests. The measurement is relatively fast: In hot-cell conditions, the measurement of one Charpy specimen took approximately 5 minutes. Therefore, it is possible to measure the surveillance specimens and return them back during an annual maintenance shutdown.

In practice, there are no physical limitations for using the method for measuring the condition of an actual RPV. However, more research related to the effects caused by reactor cladding is required. In addition, a new probe needs to be designed for that purpose.

Considering other limitations, low number of specimens could also constrain the accuracy in predicting ∆DBTT using resistivity measurements.

6. Conclusions

The results showed that the proposed new method is a promising approach for non-destructive evaluation of the RPV surveillance specimens. It is possible to develop this method further to assess the embrittlement of the actual RVP wall.

7. Acknowledgments

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