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Monitoring the embrittlement of reactor pressure vessel steels by using the Seebeck coefficient

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ABSTRACT

The degree of embrittlement of the reactor pressure vessel (RPV) limits the lifetime of nuclear power plants. Therefore, neutron irradiation-induced embrittlement of RPV steels demands accurate monitoring. Current federal legislation requires a surveillance program in which specimens are placed inside the RPV for several years before their fracture toughness is determined by destructive Charpy impact testing. Measuring the changes in the thermoelectric properties of the material due to irradiation, is an alternative and non-destructive method for the diagnostics of material embrittlement. In this paper, the measurement of the Seebeck coefficient ($\bar{K}$) of several Charpy specimens, made from two different grades of 22 NiMoCr 37 low-alloy steels, irradiated by neutrons with energies greater than 1 MeV, and fluencies ranging from 0 up to 4.5×10^{19} neutrons per cm^2, are presented. Within this range, it was observed that $\bar{K}$ increased by ≈500 nV/°C and a linear dependency was noted between $\bar{K}$ and the temperature shift $\Delta T_{41\,\mathrm{J}}$ of the Charpy energy vs. temperature curve, which is a measure for the embrittlement. We conclude that the change of the Seebeck coefficient has the potential for non-destructive monitoring of the neutron embrittlement of RPV steels if very precise measurements of the Seebeck coefficient are possible.

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1. Introduction

During the operation of a nuclear power plant (NPP) several safety relevant reactor components may undergo loads leading to degradation of the structural material. Important degradation phenomena are thermo-mechanical fatigue due to varying temperature and mechanical load and the embrittlement of the reactor pressure vessel (RPV) due to neutron irradiation. The importance of these topics is revived by the current desire for lifetime extension of nuclear power plants. In order to guarantee safe operation, monitoring of such degradations is highly desirable. The RPV made from low-alloy ferritic (LAS) steel is one of the most important safety barriers between core and the environment of a nuclear reactor, therefore its integrity is of utmost importance. In this paper, we concentrate on the embrittlement of the RPV, especially on its determination by a non-destructive method called the thermoelectric power method (TEP).

1.1. Embrittlement of the reactor pressure vessel (RPV) due to neutron irradiation

Irradiation of RPV steels with a neutron fluence $\varphi \geqslant 10^{17}$ n/cm^2 and energies above 1 MeV may result in lattice defects, which are the origin for an increase of the yield strength and a decrease of fracture toughness (embrittlement). Influencing parameters for this effect are, e.g., the chemical composition, operating temperature, segregations of Cu and P, grain size, fluence and energy spectrum of the neutrons and the operation temperature.

Brittle behaviour is characterized by an abruptly cracking without preceding plastic deformation, whereas ductile material shows plastic deformation before failure. The brittleness and ductility are also functions of the temperature. Some materials show a distinct change of the ductility within a certain temperature range, the so-called ductile-to-brittle (DBT) transition zone. A measure for these properties is the energy needed to crack special V-notched specimens by beating them with a pendulum hammer. This fracture energy is also called Charpy energy corresponding to the inventor of this testing method. Since a brittle failure of the RPV would result in a catastrophic accident, the lifetime of a NPP is limited by the achievement of the minimal allowed fracture toughness.

In the mandatory national surveillance program, Charpy V-notched specimens made from original RPV steel are irradiated in a NPP followed by Charpy impact testing. The fracture energy is measured as a function of the temperature, yielding a Charpy energy vs. temperature curve. Material embrittlement due to neutron irradiation can be characterized by the temperature shift $\Delta T_{41\,\mathrm{J}}$ of the DBT-zone, as shown in Fig. 1. The index 41 J indicates that the temperature shift of the DBT-zone is measured at the energy of 41 J.

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Beside the embrittlement, neutron irradiation leads to changes of several material properties. These changes can be used as indicators for the state of the degradation, i.e., the decrease of fracture toughness. One physical effect that might be used for the detection of material degradation is the Seebeck effect. It is one of several thermoelectric effects, mainly used for measuring temperatures with thermocouples [1]. Thomas Seebeck discovered in 1821 that a heat flow is accompanied with a small electric current. Within a certain temperature range, the generated electric field is proportional to the temperature gradient whereby the proportionality factor is called Seebeck coefficient ($\bar{K}$). We shall summarize the main equations coming from the theory of thermoelectricity, which are essential to understand the origin of the thermoelectric power and to interpret the measured results. A more detailed derivation of the theory of thermoelectricity based on quantum mechanics is given in Ref. [2].

Neutron irradiation, heat treatments and plastic material deformations are leading to a drift of the $\bar{K}$ [3–6]. However, if the change of $\bar{K}$ is a well-defined function of the neutron fluence and if the effect is large enough compared with that of other influencing parameters, it could be used for monitoring of material embrittlement, which in the case of the RPV is correlated to the neutron fluence. The application of the $\bar{K}$ for measuring the embrittlement has been investigated in the past decade by a small community. Some investigations of the TEP-method were performed within the framework of the European network Ageing Materials Evaluation and Studies (AMES) by Electricité de France (EDF) [7] and Joint Research Centre (JRC) in Petten [8]. At the Paul Scherrer Institut (PSI) the application of the TEP-method for material diagnostic is under investigation since 2001 [9]. At PSI measurements were performed on both, irradiated and fatigued specimens by using a TEP-device developed by the Institut National des Sciences Appliquées de Lyon (INSA).

In this paper, we present the $\bar{K}$ of two investigated LAS RPV steels of the type 22 NiMoCr 37 corresponding to ASTM 533-B CI.1 and ASTM 508 Cl. 2, respectively. One grade is the well characterized Japanese RPV-reference material JRQ [10] made by Kawasaki Steel Corporation at Mizushima Works, whereas the second grade stems from a Swiss nuclear power plant. The chemical compositions of the two materials are given in Tables 1a and 1b, respectively. Charpy specimens were made from both materials. The specimens made from JRQ-steel were irradiated at the PSI research reactor Saphir, whereas the second set of samples stem from the surveillance program and were therefore irradiated in a NPP.

We further compare the measured Seebeck coefficients with those measured on Charpy samples made from un-irradiated Incoloy 800. This comparison shall support the thesis, that the scatter of $\bar{K}$, which was observed for the RPV material, can be explained by inhomogeneous material properties and by the inherent uncertainties in the DBT-zone. Technical reasons (small gauge volume) of the measuring method may be an additional origin for the scatter of $\bar{K}$.

2. The Seebeck effect and its measurement

In solids, the heat is transported by phonons and by free electrons. For metals, the main contribution to the heat transport stems from the electrons. Thus, electrons are carriers of both, thermal energy and electric charge. That means: thermal and electric currents are coupled phenomena with the consequence, that an electric current accompanies a heat flow. This is the origin of the Seebeck effect whose manifestation is a thermoelectric voltage, in the following called thermoelectric power. In this paragraph we only emphasize the relevant results of the theory of thermoelectricity, some of its practical aspects and interpretations.

The important results of the thermoelectric theory are the two coupled Eqs. (1) and (2) for the electrical j_E and thermal current j_Q as a function of their origins, electric field E_x and temperature gradient dT/dx. The suffix x indicates that we only consider the x-direction

$$j_E = L^{11}E_x + L^{12}\left(\frac{-dT}{dx}\right), \tag{1}$$

$$j_Q = L^{21}E_x + L^{22}\left(\frac{-dT}{dx}\right). \tag{2}$$

In the above equations, the so-called transport coefficient L^{12} expresses that a temperature gradient is associated with an electrical current, whereas L^{21} represent the heat flow as a consequence of an electrical field. L^{11} and L^{22} are the electrical and heat conductivity, respectively. Eqs. (1) and (2) describe both, the Seebeck and its inversion, the Peltier effect. With the assumption $j_E = 0$, i.e., if we measure the electric potential using a high resistance voltmeter, we get from Eq. (1) the electric field inside the metal:

$$E_x = \left(L^{11}\right)^{-1}(L^{12})\frac{\partial T}{\partial x} = K(T)\frac{\partial T}{\partial x}. \tag{3}$$

The appearance of an electric field as a consequence of a temperature gradient is known as the Seebeck effect. For an applied electric field and a vanishing temperature gradient we get from Eqs. (1) and (2) the equations for the thermal and electrical current:

$$j_Q = L^{21}E_x \quad \text{and} \quad j_E = L^{11}E_x, \tag{4}$$

that can be summarized to

$$j_Q = L^{21}(L^{11})^{-1}j_E \quad \text{or} \quad j_E = L^{11}(L^{21})^{-1}j_Q, \tag{5}$$

where the coefficient $L^{21}(L^{11})^{-1}$ is the so-called Peltier coefficient. The thermoelectrical potential difference U (called thermoelectric voltage or thermoelectric power) between two points 0 and 1 of a specimen is the line integral of E_x evaluated from point 0 to 1

Table 1a
Chemical composition of JRQ-steel in wt%.

C	Si	Mn	P	S	Mo	Ni	Cr	Cu	V	Co	Al
0.19	0.25	1.39	0.019	0.0040	0.50	0.83	0.12	0.140	0.003	0.000	0.012

Table 1b
Chemical composition of 22 NiMoCr 37-steel from NPP in wt%.

C	Si	Mn	P	S	Mo	Ni	Cr	Cu	V	Co	Al
0.18	0.15	0.82	0.005	0.008	0.54	0.96	0.39	0.08	<0.01	0.014	0.016

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