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Quantification and local distribution of hydrogen within Zircaloy-4 PWR nuclear fuel cladding tubes at the nuclear microprobe of the Pierre Süe Laboratory from μ-ERDA

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Abstract

Hydrogen content and its distribution in in-core materials of nuclear plants are known to have a strong influence on their behaviour, especially on their mechanical properties but also on their corrosion resistance. This point has to be largely investigated in the case of the nuclear fuel cladding (Zr based alloys) of pressurized water reactors (PWR).

Two situations have been considered here, with regards to the hydrogen content and its spatial distribution within the thickness of the tubes:

(1) Irradiated fuel cladding tubes after a nominal period under working conditions in a PWR core.
(2) Non-irradiated fuel cladding previously exposed to conditions representative of an hypothetical "loss of coolant accident" scenario (LOCA).

As far as micrometric distributions of H were required, μ-ERDA has been performed at the nuclear microprobe of the Pierre Süe Laboratory. This facility is fitted with two beam lines. In the first one, used for non-active sample analysis, the μ-ERDA configuration has been improved to reduce the limits of detection and the reliability of the results. The second one offers the unique feature of being dedicated to radioactive samples. We will present the nuclear microprobe and emphasize on the μ-ERDA configuration of the two beam lines. We will illustrate the performance of the setup by describing the results obtained for Zircaloy-4 cladding both on non-irradiated and irradiated samples.

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1. Introduction

The mechanical behaviour and also the corrosion resistance of in-core structural materials are strongly influenced by the presence of hydrogen and its localisation regarding the microstructure of the materials considered. This point is of main importance in the case of the nuclear fuel cladding tubes of the pressurized water reactors (PWR), which represent the first confinement barrier, preventing the release of radioactive elements in the water of the primary circuit. Zr alloy cladding tubes (Zircaloy-4 composition in wt%: Fe 0.2%, Cr 0.1%, Sn 1.3%, Zr balance) are submitted to corrosion by the primary water circuit and a significant amount of hydrogen is picked up into the bulk. Two main situations have been studied in order to understand the

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involvement of hydrogen in the mechanisms governing corrosion and mechanical behaviour: (i) irradiated fuel cladding tubes after a nominal period under normal in-core working conditions and (ii) evolution of the non-irradiated cladding properties related to simulated conditions of a hypothetical loss of coolant accident (LOCA) scenario.

In both cases, the hydrogen quantification and local distribution at the microscopic scale have been measured using elastic recoil detection analysis (μ-ERDA) at the nuclear microprobe of the Pierre Süe Laboratory (LPS). The microprobe is equipped with two beam lines, the first one dedicated to non-active samples, the second one presenting the unique feature of allowing radioactive sample measurements.

After describing the facility and the ERDA detection configuration of the two beam lines, we will illustrate the performance of the setup with the presentation of the results obtained for the measurements performed on non-irradiated and irradiated samples.

2. The ERDA facility of the Pierre Süe Laboratory

Nuclear methods, more especially ERDA, have been used for a long time to determine the bulk hydrogen content of various kinds of matrix materials [1]. In our study, a 3 MeV $^4He^+$ incident beam with an intensity of 450 pA is impinging on the target to be measured, at a glancing angle of 15° with respect to the incident beam direction. By elastic scattering collision, the incident particles can eject from the target the hydrogen atoms, which are collected in a Si barrier detector. The quantification of H atoms gives information on the H sample content and their energy is related to the depth in the bulk.

2.1. The nuclear microprobe

The nuclear microprobe of the Pierre Süe Laboratory [2] is equipped with a single stage 3.75 MV Van de Graff accelerator, which provides a $^1H^+$, $^3He^+$, $^4He^+$ beam with an energy ranging from 500 keV to 3.75 MeV, or a $^2H^+$ beam of 500 keV–1.9 MeV. A selecting magnet assures the redirection of a monoparticle, monoenergetic beam in one of the two microfocusing lines. The first line, situated at 90° with regards to the accelerating section, is used to measure classical materials. The second one, at 45°, situated in a controlled and shielded area, is devoted to irradiating targets. The samples are placed into the relevant analysis chamber, and the detector information is stored in list-mode by a multiparametric acquisition program.

2.2. The ERDA setup of the 90° beam line

During the past few years, a particular effort has been realised to improve the μ-ERDA configuration, in order to increase the reliability of the results and to lower the detection limits. Measurements have been performed using a 8 × 2 μm^2 beam size on the target.

Three different detection modes are used simultaneously. A high purity Ge X-ray detector, set at 45° from the incident beam direction and shielded with a 150 μm mylar foil, is used to determine precisely the analysis region of interest, on the basis of Zr and main alloying elements (Fe, Cr, Sn) mapping in the alloy. The RBS detector signal, collected on an annular Si-barrier detector situated at 170° from the incident beam direction, is also used for localisation and serves as a charge monitor. The detector is partially collimated in order to avoid a shadowing effect of the sample holder in the tilted ERDA position. The RBS solid angle of detection, measured with reference samples in normal and tilted positions is 12 mSr. The H atoms are detected at 30° with regards to the incident beam direction, by a plain Si-barrier detector, shielded by a 14 μm Al filter, with a solid angle of 12 mSr, measured with a reference sample.

Computer driven beam scans on the target of 400 × 300 μm^2 size allow mapping cartographies of Zr, H and alloying elements (Fe, Cr, Sn).

2.3. The ERDA setup of the 45° beam line

The analysis chamber of the 45° beam line, dedicated to radioactive samples, is situated in a controlled shielded area. The samples, transported in a shipping cask, are unloaded and handled in hot cells with slave arms.

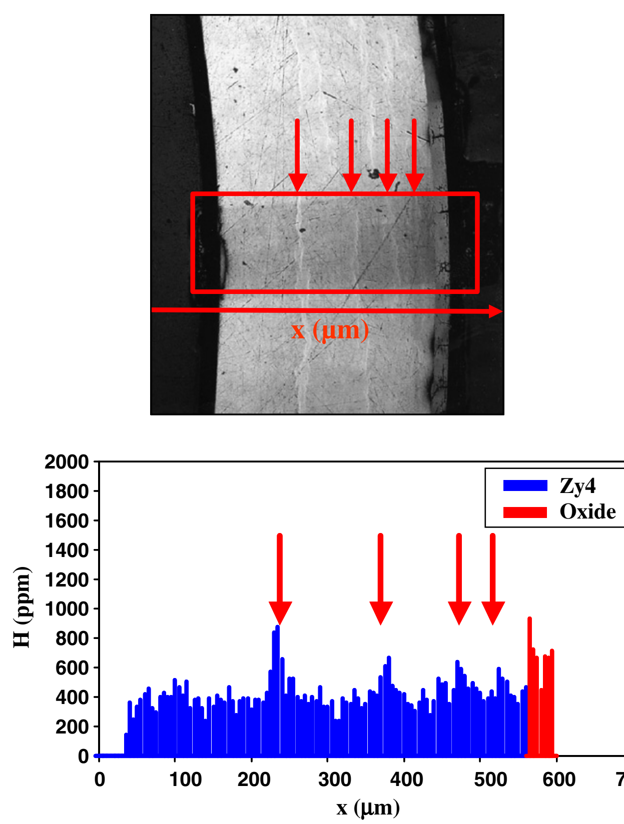

Fig. 1. Metallographic observation of the hydride distribution on Zircaloy-4 irradiated for 5 PWR cycles, and corresponding hydrogen profile, obtained by ERDA.