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Use of ion beam analysis techniques to characterise iron corrosion under water radiolysis

S. Lapuerta [a,b,*], N. Moncoffre [a], N. Millard-Pinard [a], E. Mendes [c], C. Corbel [c], D. Crusset [b]

[a] *Institut de Physique Nucléaire de Lyon, 4 rue Enrico Fermi, 69622 Villeurbanne Cedex, France*
[b] *ANDRA, Parc de la Croix Blanche 1-7 rue Jean Monnet, F-92298 Châtenay-Malabry Cedex, France*
[c] *Commissariat à l'Energie Atomique, CEA-Saclay, DSM/DRECAM/SCM/Laboratoire CEA de radiolyse, Bat. 546, F-91191 Gif-sur-Yvette, France*

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Abstract

The aim of this paper is to study the effect of water radiolysis under 12 MeV proton irradiation on the corrosion behaviour of pure iron. Oxygen and hydrogen playing a crucial role during the corrosion process have been specifically investigated. Heavy desaerated water (enriched at 99.9% in D) was also used to determine the origin of hydrogen at the iron surface. Proton irradiations were performed at the CERI cyclotron in Orléans. Both sides of the Fe foil (respectively in contact with air and with water) were analysed with ion beam techniques: alpha Rutherford backscattering spectrometry was used to profile oxygen, elastic recoil detection analysis has allowed to profile hydrogen. The use of D_2O gives evidence that the hydrogen concentration present on the water face could originate from wet air. In addition, in case of the aerated deionised H_2O media, it is shown that the irradiation process induces a strong corrosion. Scanning electron microscopy experiments confirm the formation of oxide precipitates.

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Keywords: Iron corrosion; Radiolysis; Ion beam analysis

* Corresponding author. Address: Institut de Physique Nucléaire de Lyon, 4 rue Enrico Fermi, 69622 Villeurbanne cedex, France. Tel.: +33 4 72 43 10 63; fax: +33 4 72 44 80 04.
E-mail address: lapuerta@ipnl.in2p3.fr (S. Lapuerta).

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1. Introduction

In the perspective of a geological disposal, high level nuclear wastes will be embedded in stainless steel. The second barrier will be very likely a low alloyed carbon steel overpack. It is assumed that, progressively, water will penetrate the geological site and after some hundred years, overpacks will be in contact with water and submitted to γ-irradiation. Because of the radioactive environment, water radiolysis will occur. This radiolysis produces both molecular (H_2O_2, H_2) and radical products ($OH^{\bullet}, O_2^{-\bullet}, HO_2^{\bullet}, e_{aq}^-, H^{\bullet}$) which could accelerate the corrosion process. Lots of works deal with the comparison of the radiolysis product primary yields induced by either γ, electron or proton irradiations [1]. Moreover these yields have been estimated as function of LET (linear energy transfer) by Monte Carlo simulations [2]. In this context, we propose a fundamental study to better understand the corrosion mechanisms of pure iron, considered as a model material, under proton irradiation. We have chosen proton irradiation for two main reasons. First, the protons produce a high ionisation density correlated with a high LET compared to γ-irradiation and it has been shown that water radiolysis strongly depends on the LET value [3,4]. Second, the beam energy control allows to study the corrosion process precisely at the iron/water interface. Several papers have put in evidence enhanced corrosion under charged particle irradiations in different materials: ceramics [5], zircaloy [6] and other metals [7]. Few works have been done on mild steels or pure iron. Burns [8] shows that, under γ-irradiation, an increase of the H_2 production occurs and he proposes specific reactions to explain the strong observed corrosion effect.

In this paper, we will study the corrosion of pure iron in contact with water under 12 MeV proton irradiation. In order to follow the iron surface evolution, an Eu marker introduced by ion implantation is used and analysed by Rutherford backscattering spectrometry (RBS). In addition, oxygen and hydrogen profiles are determined by ion beam analysis. Two types of experiments were realised with two different water media: aerated deionized water (H_2O) and heavy water (D_2O) packaged under argon. By the comparison of these two experiments, we have been able to determine the origin of hydrogen at the iron/water interface and the influence of dissolved species in water.

2. Experimental

The studied material is pure iron (99.995%). The samples are 250 μm thick discs with a 8 mm diameter. Since the iron disc thickness is very thin, only the iron surface in contact with water is mechanically polished with a 3 μm diamond paste. The measured rugosity is consequently of 50 nm on the non polished side and 5 nm on the polished one.

Samples are Eu implanted on the polished face at an energy of 300 keV and a fluence of 5×10^{15} at. cm^{-2}. These implantation conditions correspond to a 30 nm range given by the SRIM simulation [9] and an Eu concentration maximum value of 1.1 at.%.

The irradiation experiments are performed with the CERI (Centre d'Etudes et de Recherches par Irradiation) cyclotron at Orleans, which delivers a 12 MeV proton beam. The external proton beam enters the irradiation cell ($V = 20$ mL) through the 250 μm thick iron foil and emerges into water with a 4.5 MeV energy where it finally stops after a 290 μm range. Fig. 1 presents a schematic drawing of the irradiation set up. The LET at this energy is equal to 8.8 eV nm^{-1} which is much higher than that induced by γ-irradiation (0.2 eV nm^{-1}). Samples are irradiated with a 30 nA beam intensity. This intensity, which corresponds to an irradiation flux of 6.7×10^{11} p cm^{-2} s^{-1}, is set constant and measured carefully both by a Faraday

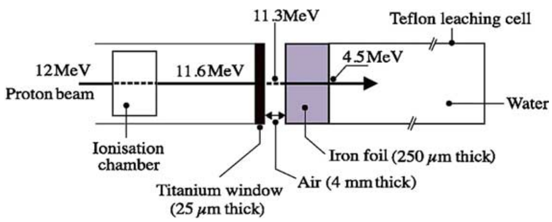

Fig. 1. Schematic representation of the irradiation set up for iron leaching under proton beam.