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# Studying processes of hydrogen interaction with metallic surfaces in situ and in real-time by ERDA

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**Abstract**

A special cell was constructed for studying the interaction of hydrogen with materials. It enabled exposure of samples to a neutral hydrogen atmosphere and simultaneous concentration depth profiling of H and D. The concentration of individual hydrogen isotopes was measured by elastic recoil detection analysis (ERDA) using a 4.2 MeV $^{7}Li^{2+}$ probe beam. The measurements reported here were performed on Ti and W. The time evolution of processes occurring on the surface and in the bulk was studied by successive recording of ERDA spectra. For specific conditions in the cell a model describing adsorption and desorption of deuterium molecules on a W surface was proposed. Sticking and recombination coefficients were obtained by fitting the time evolution of deuterium concentration.


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*Keywords:* Hydrogen; Deuterium; ERDA hydrogen depth profiling; Surface processes; Adsorption

## 1. Introduction

Interaction of hydrogen molecules (e.g. [1]) and atoms (e.g. [2]) with materials is an extensively studied subject due to its fundamental and practical importance. Special attention is recently devoted to the role of the hot-atoms from the surface in atom recombination and abstraction (e.g. [3–5]). Many of experimental and theoretical studies dealing with hydrogen reactions at surfaces are performed with well defined, ordered and clean surfaces. Such studies increase understanding of these fundamental processes and they are applicable to the clean surface technologies. However, similar processes at higher pressure occurring on the polycrystalline and possibly contaminated surfaces are also of great practical importance for understanding the gas-wall and plasma-wall interaction. It was shown [6] that vibrationally excited hydrogen molecules are produced by atom recombination under such conditions on the way that strongly depends upon surface composition. We have started studies of hydrogen (deuterium) adsorption, desorption, abstraction, recombination and diffusion on materials of practical importance in order to elucidate the role of hydrogen present in the near surface bulk. This work is motivated by the increased interest on retention of hydrogen isotopes in the wall of the fusion reactors [7,8] and also by the importance of vibrationally hot molecules for the fusion edge plasma [9,10].

Here we describe details and procedures of the experimental set-up that we developed for real time monitoring of hydrogen distribution in material while it is exposed to the thermal hydrogen molecules and atoms. A special chamber was constructed for this purpose that allows simultaneous material exposure to the test atmosphere and in situ hydrogen depth profiling by ion beam analytical method elastic recoil detection analysis (ERDA) [11]. In order to illustrate experimental performances of the new set-up we present some results obtained with Ti and W samples – H outgassing from Ti and hydrogen and deuterium adsorption, desorption and isotope exchange on W.

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A simple model for adsorption of deuterium molecules on W was developed and compared to the experimental results.

## 2. Experimental method

### 2.1. Chamber for sample exposure to hydrogen

A special cell was constructed for sample exposure to a controlled neutral hydrogen atmosphere and simultaneous in situ measurements of hydrogen concentration depth profiles by ERDA. This hydrogen exposure cell (HEC) is schematically shown in Fig. 1. The cell has the shape of a half cylinder, with an inner radius of 35 mm and a height of 50 mm. The sample is mounted on the flat side of the cell and heated from the rear side by a thermocoaxial heater mounted in the sample backing; its temperature is monitored by a thermocouple, mounted on the sample holder plate. The cylindrical envelope of the HEC is cooled by a water or air flow and its temperature is also monitored by a thermocouple. Two windows were cut in the envelope to allow in situ ion beam analytical method ERDA to be applied. One is the entrance window for the incoming high energy ion probe beam and the other window is for allowing the recoiled hydrogen atoms to reach the silicon detector. The windows are positioned at the half height of the cylindrical envelope and at $\pm 75°$ with respect to the sample surface normal. In order to reduce gas leaks from the cell, the windows are tightly covered by Al foils, 0.8 μm thick at the entrance window and 6 μm thick at the detector window. The 6 μm Al foil at the detector window acts together with another 6 μm Al foil, placed at the silicon detector, as a stopping barrier for the scattered incident ions. Gas inlet and pressure measurement ports are placed on the bottom of the cell. Gas pressure is measured by the Baratron, a capacitive pressure gauge. A tungsten filament, 0.2 mm in diameter, is mounted on the top of the cell. When heated, this filament is used to dissociate hydrogen molecules

$$H_2 + W(T > 1200\,K) \rightarrow H + H + W(T). \quad (1)$$

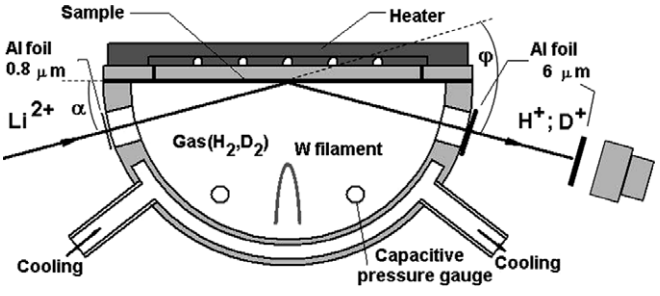


Fig. 1. Schematic drawing of the cell (HEC) for sample exposure to a controlled neutral hydrogen atmosphere and in situ hydrogen depth profiling. Elastic recoil detection analysis (ERDA) is used for in situ measurement of the hydrogen depth profile in a sample. The angle between the sample surface and the incoming ion beam is 15° and the angle between incoming ion beam direction and the silicon detector is 30°. Incident projectile ions are 4.2 MeV $^7Li^{2+}$.

The temperature of the filament was determined by an optical pyrometer as a function of the heating current. The temperature of the tungsten filament in the present experiments was 1600 °C. The characteristics of the cell atmosphere are varied by changing the hydrogen isotope composition and by turning on and off the filament.

### 2.2. ERDA method

The cell was mounted in the RBS/ERDA scattering chamber in the 10° beam line of the 2 MV Tandem accelerator at the Jožef Stefan Institute (JSI) in Ljubljana [12]. A 4.2 MeV $^7Li^{2+}$ ion beam was used for probing the studied sample mounted in the HEC. The beam was collimated with a vertical 1 mm × 4 mm rectangular shaping slit placed in front of the entrance of the experimental chamber at the distance of 35 cm from the target sample. A mesh charge collector mounted behind the shaping slit was used to determine the number of ions hitting the target [13]. A conventional silicon detector for detecting recoiled particles (H and D in the present case), later labelled as ERDA detector, was placed at an angle of 30° ($\varphi$ in Fig. 1) with respect to the direction of the incident beam. The angle between the beam direction and the sample surface was 15° ($\alpha$ in Fig. 1). Two Al foils with a total thickness of 12 μm were used to stop scattered Li ions. The experimental set-up of the ERDA is described in more detail elsewhere [14]. The background vacuum pressure is $5 \times 10^{-6}$ mbar.

Energy spectra of recoiling hydrogen and deuterium atoms were measured with the ERDA detector. Recoiled H and D from the same depth are well separated in energy due to the mass dependence of the recoil kinematic factor.

Accurate elastic recoil cross-section (CS) is needed for quantitative application of ERDA. CS for $^7Li$ on H is not Rutherfordian [14] but accurate experimental CS does exist [15] and it was used for the present spectra evaluation. For the case of $^7Li$ on D accurate CS is not available and we used Rutherford recoil CS for the present spectral analysis. Once the more accurate CS for $^7Li$ on D will be available the present values for D concentration can be easily scaled. For determination of the concentration depth profile of H and D [16] we employed the SIMNRA program [17].

## 3. Real-time measurement by ERDA

The experimental cell was used for exposure of the sample to hydrogen and simultaneous ERDA measurements of hydrogen depth profiles. In this experiment the following parameters could be varied:

- Composition of gas in the cell ($H_2$ or $D_2$).
- Gas pressure in the cell.
- Sample temperature.
- Degree of dissociation (variation of filament temperature).