

AMS MEASUREMENTS OF DEUTERIUM CAPTURED IN TUNGSTEN LAYERS DEPOSITED BY MAGNETRON SPUTTERING*

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Abstract. The paper presents the upgrading of the Accelerator Mass Spectrometry (AMS) facility of IFIN-HH, Bucharest, performed in order to measure depth profile of hydrogen isotopes concentration in various host materials. Results are also presented concerning the retention of deuterium in different tungsten layers deposited by magnetron sputtering procedure. The tungsten layers were deposited on a silicon substrate separately or simultaneously with the capture of deuterium. Our AMS measurements show that deuterium penetrates in different ways into the material according to the kind of tungsten deposition and it is easily captured below layers of tungsten.

Key words: accelerator mass spectrometry, depth profiling, deuterium in materials.

1. INTRODUCTION

The selection and study of the materials suitable for development of Plasma Facing Components (PFC) used currently in tokamaks are topics of wide interest. The Plasma Facing Materials (PFM) selected to the moment for design and realization of PFCs are carbon, tungsten and beryllium. Plasma wall interactions (occurring when the plasma escapes magnetic confinement) result, through chemical and physical sputtering, in the ejection of surface atoms and molecules,

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which either reenter the core plasma or are redeposited on other wall surfaces as mixtures of the wall components, forming the so-named codeposited layers. The interaction between the plasma and the PFCs (like for example protection tiles that are on the inside first wall and the divertor) leads to both modification of internal structure of the protection tiles and to safety issues due to tritium accumulation in wall in quantities exceeding the safety limits. For these reasons, the control and monitoring of deuterium and tritium retention in PFC is an important issue [1, 2].

With respect to this subject we have prepared in the laboratory samples of tungsten with incorporated deuterium (simulating tungsten codeposited layers containing tritium) and we studied the retention of deuterium in the prepared samples. The main task of this work is to determine, using the high sensitivity method of Accelerator Mass Spectrometry (AMS) the concentration of deuterium captured in deposited layers. For radioactivity safety reasons in all sample preparations only deuterium was used.

2. EXPERIMENTAL METHODS

A series of tungsten thin films were deposited on silicon substrates by magnetron sputtering (MS). Incorporation of deuterium in tungsten layers was performed in two approaches. In the first one, thin deuterated tungsten layers were deposited by magnetron sputtering of tungsten in argon and deuterium gaseous mixture [3]. In the second one, deposition of tungsten was performed by MS only in argon, followed by sample exposure to a deuterium radiofrequency (RF 13.56 MHz) plasma.

The experimental setup used for samples preparation is presented in Fig. 1. It consists in a vacuum chamber on which a magnetron sputtering gun (equipped with a 2 inches tungsten target, 99.8% purity) is mounted. The vacuum chamber is pumped using a turbo molecular vacuum pump while the processing gases (argon and deuterium) are introduced in the chamber using mass flow controllers. During the deposition of tungsten by MS (either in pure argon or argon and deuterium mixture) the substrate holder is grounded.

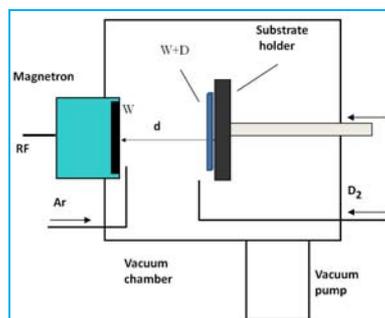


Fig. 1 – Magnetron sputtering chamber.

Deposition of pure tungsten layers by MS was performed using 47W power, and 20 sccm mass flow rate of argon, corresponding to a pressure of 8.9×10^{-3} mbar. Deposition of deuterated tungsten layers by MS was performed mixing 10sccm of deuterium with the already present 20 sccm of argon, resulting in a deposition pressure of 1.3×10^{-2} mbar. In both cases, the distance between tungsten target and the substrate was of 7cm. The deposition rate of pure tungsten layers was of 8.3 nm/minute while of the deuterated tungsten of 10.3 nm/minute. By using these deposition parameters thin layers of tungsten (deuterated or not) with up to 1000 nm thickness, being stable on the silicon substrate, were deposited.

Supplementary, the experimental setup presented in Fig. 1 was used for exposing pure tungsten thin films (1000 nm thickness) to deuterium plasma. Such as, after deposition of the tungsten thin films by MS (using only argon, as presented before), they were exposed to a deuterium plasma generated by applying an RF bias on the substrate holder. The parameters of the deuteration process are: 10 sccm deuterium mass flow rate, pressure of 7.5×10^{-2} mbar; 40W RF applied power; 3 hours duration of treatment. Except from plasma, no additional heating was used. After deuteration, the samples were cooled down for 30 minutes in a constant flow of deuterium (10 sccm) at a pressure of 4×10^{-1} mbar.

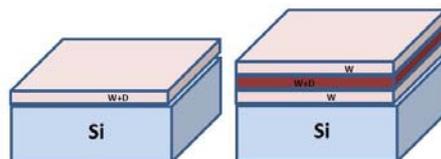


Fig. 2 – Schematics representation of the deuterated tungsten thin film on silicon substrate (left), three tungsten thin layers sandwich on silicon substrate (right).

The following samples were prepared: a) a silicon substrate exposed to radiofrequency deuterium plasma; b) a 1000 nm thick layer of pure tungsten deposited by MS and deuterated by exposure to deuterium plasma; c) a 500 nm thick layer of deuterated tungsten, deposited by MS in mixture of argon and deuterium; d) a sandwich of three tungsten layers deposited by MS on silicon substrate: the in between layer (660 nm thick) is deposited in mixture of argon and deuterium while the outer layers (500 nm thickness) in pure argon. The layers distribution in the samples deposited by MS is presented in Fig. 2. All samples are square, presenting around 7.4×7.4 mm²; this dimension was imposed by Accelerator Mass Spectrometry (AMS) sample holder.

The AMS facility is located at the 9 MV Tandem from the “Horia Hulubei” National Institute of Physics and Nuclear Engineering – IFIN HH – Bucharest. The experimental AMS system is presented in detail in [4, 5, 6]. The main components of the AMS facility are: the ion source (the Multi-Cathode Source of Negative Ions by Cesium Sputtering, 40 MC-SNICS), the injector deck, the linear tandem

accelerator, the analyzing magnets and the detector system. The AMS analysis method uses samples with very low concentrations of the measured isotopes. For these samples, the analyzed ion currents extracted from the ion source are very small, even to a few isotope ions per second. This extremely low currents cannot be detected by the diagnose systems between the ion source and the detector system situated at the end of the AMS facility. The AMS detector system is the only one capable of detecting and measuring the events from the rare isotope – detector system interactions. This isotope beam is invisible for the entire AMS facility, but not for the detector system. In order to find and optimize the transport of extremely low intensity beam, the AMS uses a so-called “pilot beam” [7, 8]. The guidance and beam filtering through the entire system is achieved by changing and tuning the values of the electrostatic and magnetic filters. Our measured isotope beam consists in extremely low intensity deuterium beams that are extracted from the tungsten or silicon matrix (*i.e.* the deposited samples presented before). The deuterium beam was accelerated to energies of approximately 6 MeV, with the state of charge 1+. For this experiment we have used as pilot a ^{12}C beam. Carbon is the visible beam that provides the optimum route for deuterium beam along the entire analysis in the AMS system. This beam of carbon ions was accelerated to energies of approximately 25 MeV, with the state of charge 5+.

3. RESULTS

3.1. UPGRADE OF THE AMS FACILITY FOR DEPTH PROFILING EXPERIMENTS

In order to achieve the in depth profilometry of the hydrogen isotope concentration, the standard AMS facility was upgraded. For this purpose we assembled a suitable detector system for measuring very light isotopes, such as deuterium and tritium. The semiconductor detector system that was made consists of two, three or four silicon Surface Barrier Detector (SBD), arranged as a sandwich, positioned one behind the other, perpendicular to the beam direction. In addition, to achieve the in depth profilometry of the hydrogen isotope concentration, it was necessary to design a suitable AMS sample holder.

The AMS modified sample holder was made in order to measure depth profile of hydrogen isotopes concentration in various host materials. This modified sample holder allows the use of squared plates having the side up to 7.4 mm or rounded plates up to 10.4 mm in diameter. It is shown in Fig. 3. During the measurements the samples from the holder are bombarded with Cs ions provided by a sputtering source, which are producing in-depth sample erosion. From the sputtered material from various depths the deuterium ions are selected by the AMS setup and directed into detector.



Fig. 3 – Modified AMS sample holder.

3.2. DEPTH PROFILING OF DEUTERIUM CONCENTRATION IN THE PREPARED SAMPLES

We have set out to make a qualitative estimate (not a quantitative one) of the in depth deuterium concentration, in order to characterize deuterium retention in tungsten and silicon layers.

First, raw data has been acquired as deuterium counts *versus* time (seconds). Then the selected samples have been analyzed by profilometry measurements using a Dektak device. Sample depths were measured and sputtering rates were calculated. The results were corrected to the background value which was obtained by measuring the deuterium concentration in samples of deuterated silicon substrate and pure tungsten thin layer deposition on silicon substrate.

The presented graphs show the depth profile of deuterium counts. The different layers were delineated by dotted lines marking. The Fig. 4 show marked layers of silicon, tungsten and deuterated tungsten.

In Fig. 4a is presented the depth profile of implanted deuterium concentration in a silicon sample. The implant occurs up to a depth of 350 nm with a maximum at 50 nm. In Fig. 4b, it is showed the depth profile of implanted deuterium concentration in a silicon sample with a deposited tungsten layer. The implant presents a peak at approximately 500 nm. A diffusion phenomenon in silicon substrate can be observed. In Fig. 4c is presented the depth profile of deuterium concentration in a tungsten layer deposited in mixed Ar+D₂. The deuterium distribution is constant in the tungsten layer. In Fig. 4d is presented the depth profile of deuterium concentration in a sandwich sample (Fig. 2 right, too). The different layers were delineated by dotted lines marking.

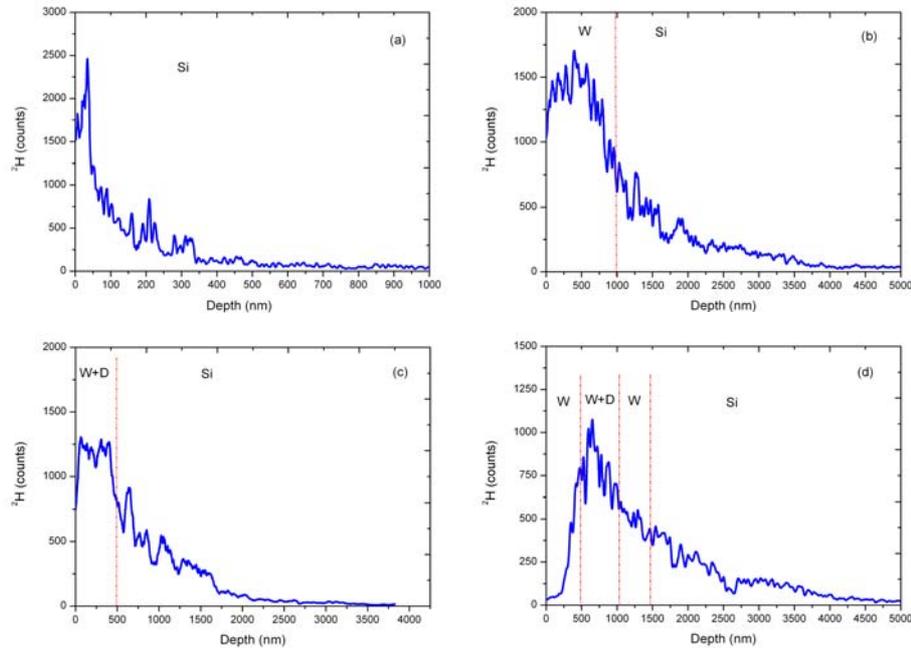


Fig. 4 – Dependence of Deuterium counts as function of depth in: a) deuterated silicon substrate by radiofrequency deuterium plasma; b) pure tungsten thin layer deposition on silicon substrate by magnetron sputtering and then deuterated by RF; c) deuterated tungsten thin layer deposition on silicon substrate by magnetron sputtering; d) three tungsten thin layers sandwich on silicon substrate by magnetron sputtering.

4. CONCLUSIONS

We have obtained in depth qualitative estimate of deuterium distribution in deuterated tungsten samples. In all the deposited samples, deuterium diffuses beyond the tungsten film up to depths of 3–4 μm , reaching the silicon substrate. In the silicon sample (type a) exposed to a deuterium RF plasma is observed an in-depth penetration of only of 350 nm, while in the deposited samples this is about a few μm . This shows that the deposited tungsten layer favors deuterium diffusion in the silicon substrate.

In the case of the sample deposited by MS and deuterated by exposure to deuterium plasma (type b) a non-uniform in-depth distribution of deuterium in the tungsten layer is observed. Contrary, in the case of the sample deposited by MS in a mixture of argon and deuterium (type c) an uniform distribution of deuterium in the tungsten layer is observed, according to an homogeneous incorporation of deuterium during the process. In the sandwich sample (type d) we observe that the

uniform distribution of deuterium disappears, which leads to the conclusion that the adjacent tungsten layers do not act as barriers for the deuterium diffusion

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