Diagnostics for plasma-material interaction studies

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Abstract

The manuscript is accompanying the corresponding lecture at the 12th Carolus Magnus Summer School on Plasma and Fusion Energy Physics. The contribution is focused on surface analysis and erosion-deposition diagnostics frequently applied in the fusion relevant research on plasma-material interaction. The definitions of real-time, in-situ, in-vacuo and ex-situ diagnostics are given. The working principles of the following surface analysis tools are described and examples of their applications are given: thermal desorption spectrometry (TDS); scanning electron microscopy (SEM) including the option of focused ion beam (FIB); ion beam analysis (IBA) methods of nuclear reaction analysis (NRA) and Rutherford back-scattering (RBS); and secondary ion mass spectrometry (SIMS). Two methods of erosion-deposition measurements are introduced: quartz microbalance (QMB) and marker techniques.

1. Introduction

The availability of the fusion reactor is crucial for its economic operation and is to a large extent determined by plasma material interaction (PMI). There are two main PMI issues limiting the availability of the reactor: the lifetime of the wall components and the operational safety. The first wall will suffer from heat loads and particle bombardment, causing erosion of materials and limiting the lifetime of the first wall components. The safety issues are connected to the restrictions on the amount of radioactive tritium and dust stored in the reactor vacuum vessel.

PMI studies in both large-scale, at close-to-reactor conditions, and lab-scale, at well-defined conditions, devices are necessary for a better understanding of the background processes, refined predictions for the reactor and finding solutions to critical issues [1]. Therefore, the methods to determine PMI related values, i.e. material erosion and deposition and fuel retention, are important experimental tools in the fusion research.

There are numerous surface analysis techniques analysing the physical and chemical properties of the material surface. E.g. an overview table in the book by G.A. Somorjai [2] lists 60 "most frequently used surface characterization techniques". In this paper, only a fraction of the techniques, which are most relevant to the PMI research, are covered, along with the methods for studying erosion and deposition of the wall materials.

2. Definitions of real-time, in-situ, in-vacuo and ex-situ

Material samples can be analysed in different ways concerning the temporal and spatial scale of analysis. The following nomenclature has been established for the description of different methods, here listed in order of the time point of their application or data availability: "real-time", "in-situ", "in-vacuo" and "ex-situ" ("post-mortem").

The real-time techniques deliver the data during the investigated process. They are often used for the real-time, or feedback, control of the process. Usually, less complex, robust methods are applied real-time for the control purposes.

In-situ (lat.: "in position") methods are, similarly to the real-time ones, applied during the investigated process, e.g. during the plasma exposure of a material sample. However, the data analysis often requires additional efforts after the actual experiment, e.g. for the conversion of the raw diagnostics data to the meaningful data describing the particular PMI process. Therefore, the data of the in-situ measurements typically becomes available after the experiment. Nevertheless the in-situ data often provides valuable insights in the temporal evolution of the investigated process. In-situ methods of PMI including optical emission spectroscopy are the topic of a different lecture.

In-vacuo (lat.: "in vacuum") methods are applied after the investigated process, e.g. after the plasma exposure of a material sample, but before the exposure of samples to air. Typically they are conducted in the same vacuum chamber. Plasma-exposed surfaces are often chemically activated, exposure to air changes the chemical state of the surface, e.g. by oxidation. Therefore, it is favourable to analyse the sample surface in-vacuo.

Ex-situ (lat.: "out of position") methods are applied after the investigated process and after the exposure to air. In some cases the influence of air is minimised by the use of sealable enclosures, called "desiccators", which are pumped down or filled with inert gas, for the sample storage and transport. However, it cannot be excluded, that the sample surface changes its characteristics between the exposure and the ex-situ analysis. "Post-mortem" (lat.: "after death") is a term equivalent to "ex-situ".

It is usually quite challenging to install and operate a diagnostic for the in-situ or in-vacuo surface analysis. However, the largely increased value of the data with respect to the ex-situ methods and the technical progress in the analysis tools make the use of these techniques more popular. Despite their advantages, in-situ and in-vacuo analysis solutions have a significant downside of high complexity and costs. Therefore, most PMI experiments still rely on the ex-situ surface analysis techniques. Another reason for the predominant use of ex-situ is that the material samples can be transported to various laboratories specializing in particular analysis techniques, thus increasing the versatility and quality of analyses.

One example of in-situ analysis in the DIONISOS plasma facility [3] at MIT, USA is given in figure 1(a). Here, an ion beam accelerator is used for the ion beam analysis simultaneously during the plasma exposure, providing insight on the dynamics of PMI processes. Figure 1(b) shows the in-vacuo surface analysis station of PISCES-B [4]. The samples are extracted from the target station, after termination of the plasma discharge, by a swing-linear manipulator and inserted in the surface analysis station, where Auger electron spectroscopy (AES), X-ray photoelectron spectroscopy (XPS) and secondary ion mass spectrometry (SIMS) techniques can be applied. For the PSI-2 device [5], the application of the laser based techniques including LID and LIBS is envisaged for in-vacuo surface characterization in the target exchange and analysis chamber (figure 1(c,d)). The sample carrier can be retracted after the exposure to plasma by a linear manipulator to the target exchange and analysis chamber, where it can be analysed by the laser based techniques. Mass spectrometry can be used for measurement of the desorbed deuterium, and a 2D optical spectroscopy system can be applied to detect the light intensity at a wavelength corresponding to a certain element on the surface. The plasma operation in PSI-2 can be continued with the retracted sample carrier using a removable plasma dump. The vacuum valves separate the exposure and the analysis regions of the vessel.

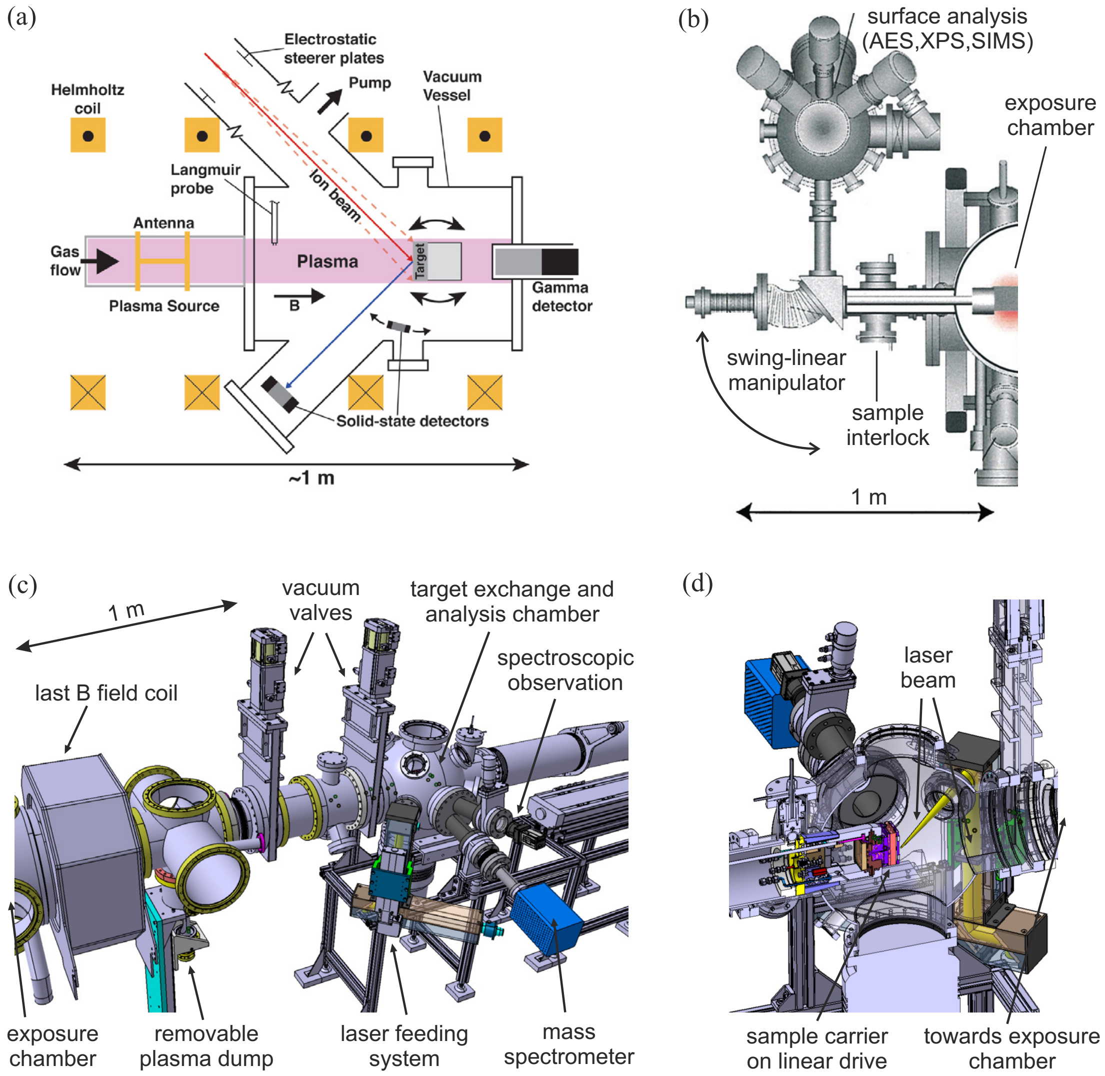

Figure 1. (a) In-situ ion beam analysis in DIONISOS [3]; (b) in-vacuo analysis station of PISCES-B [4]; (c) PSI-2 target exchange and analysis chamber with a laser feeding system for in-vacuo analysis; (d) cross-section of the opposite side of the PSI-2 target exchange and analysis chamber including an illustration of a sample exposed to a laser beam [5].

3. Surface analysis techniques

3.1 Thermal desorption spectrometry (TDS)

Thermal desorption spectrometry (TDS) is a frequently applied technique for the analysis of the gas, i.e. hydrogen isotopes, retention in samples. The gas is retained in various trapping sites in the material. By heating up of the sample the gas atoms escape the traps, diffuse to the sample surface and are released from the sample. In a TDS apparatus (figure 2(a)), the samples are heated in a compact vacuum chamber by a surrounding furnace. The ramp rate of temperature is defined by the power of the furnace feedback controlled by the thermocouple measurements at the sample inside the vacuum chamber. Therefore, the technique is also often referred to as temperature programmed desorption (TPD). Partial pressures of different gas components are measured by a residual gas analyser (RGA), which is calibrated for the relevant masses by calibrating leaks. Typical temperature ramp rates are 0.1 K/s – 10 K/s. A faster ramp results in a higher signal but a lower temperature resolution in comparison with a slower ramp. The typical heaters which are used for TDS are infrared lamp or resistive

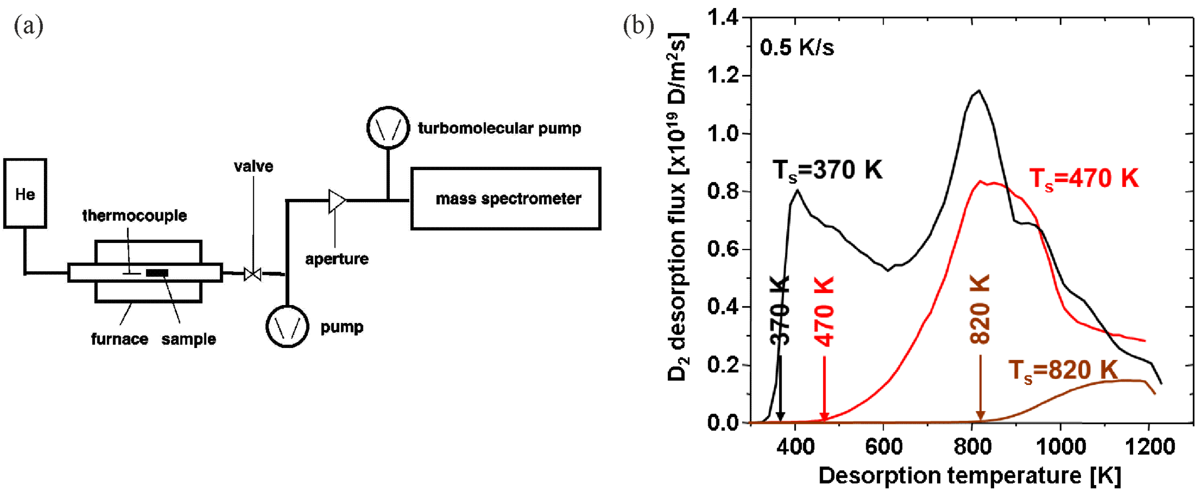

Figure 2. (a) Scheme of TDS system; (b) examples of spectra from thermal desorption. Temperatures at which the samples were exposed to deuterium plasma are indicated.

furnace.

Figure 2(b) shows an example of thermal desorption spectra of mass 4 (deuterium D_2) for carbon fibre composite (CFC) samples exposed to PISCES-A deuterium plasma at different sample temperatures [6]. The peaks correspond to different trapping sites of D in CFC. The distribution of deuterium in the sample bulk and a finite diffusion time out of the sample during the heating ramp lead to the peak broadening. The integral of the curve gives the total amount of deuterium in the sample released as D_2 molecules. Various methods are available to analyse the TD spectra, including sophisticated numerical codes describing the transport of solute gases in solids such as TMAP [7] and CRDS [8].

Laser induced desorption (LID) is a method in which a laser is used to heat up samples. The method can be seen as TDS with an extremely fast (~1-10 ms) heating ramp. In this mode, the temperature resolution of the desorbed signal is completely lost, only the total amount can be measured. However, the method is fast and can be applied with a lateral resolution along the sample surface, e.g. to obtain a 2D mapping of the gas retention.

3.2 Scanning electron microscopy (SEM) and focused ion beam (FIB)

Scanning electron microscopy (SEM) is an electron beam based method of the surface analysis. The incident e-beam of ~1-30 kV is scanned across the surface and causes emission of secondary electrons (SE mode of operation) and backscattered electrons (BSE mode of operation) (figure 3(a)).

The SE mode is the most frequently used operation mode of SEM. It provides the surface topology by measuring the intensity of secondary electrons, which is a function of the angle between the surface and the detector. The imaging is provided by the rastering (scanning) of the e-beam along the surface. Figure 3 (b) shows an example of an SEM image taken in the SE mode.

The BSE mode provides the information on the elemental composition of the surface. It uses the fact that the intensity of backscattered electrons is a function of atomic mass of the scattering element. Heavy elements on the surface correspond to a bright part of the image, while light elements result in a dark part of the image (figure 3(c)).

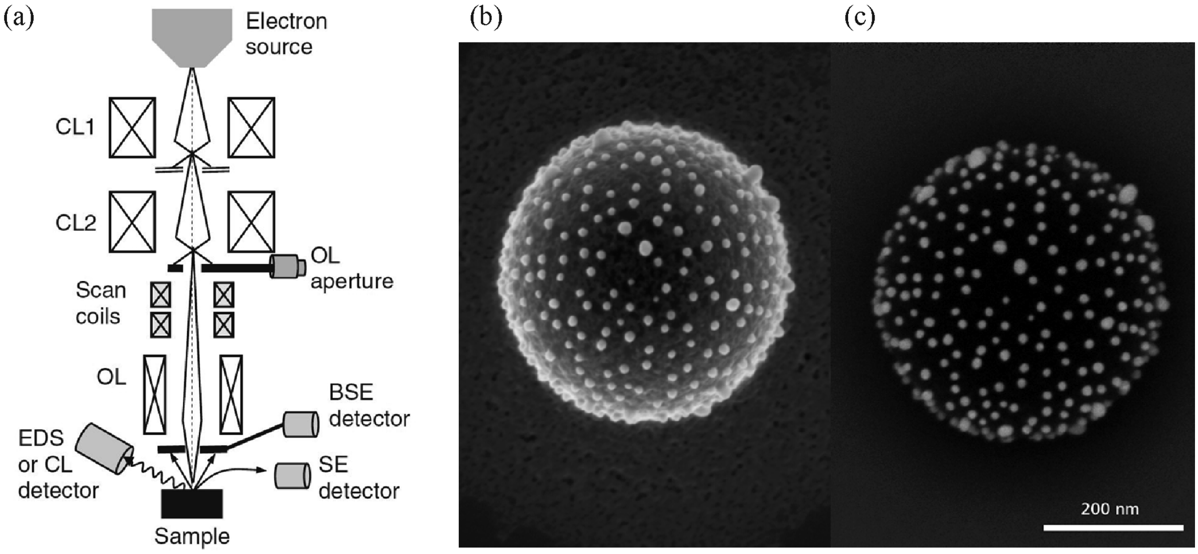

Figure 3. (a) Scheme of a SEM apparatus [9]; examples of (b) SE and (c) BSE operational modes of a SEM system: gold nanoparticles on polystyrol sphere. The SE image provides surface topography, while the BSE image gives material contrast [10].

The focused ion beam (FIB) technique is used for the nano-machining of the surface, e.g. surface cross-sectioning or preparation of small specimens. After the FIB treatment the surface is typically analysed by SEM. In a FIB instrument, an ion beam, typically Ga^+, is rastered over the sample causing well controlled sputtering. Figure 4 shows a scheme of the application of FIB for the surface cross-sectioning with the subsequent SEM analysis, as well as SEM images of the surface topology and cross-section of a nano-structured tungsten surface, also known as tungsten fuzz.

3.3 Ion beam analysis (IBA): nuclear reaction analysis (NRA) and Rutherford back-scattering (RBS)

Ion beam analysis (IBA) techniques rely on a high energy (~1 MeV) ion beam produced, e.g., by a tandem accelerator. Several beam lines for different experiments are usually attached to the accelerator. In the example of the in-situ surface analysis given in figure1(a), the linear plasma device DIONISOS is attached to one of the beam lines delivering the ion beam from the accelerator. There are numerous IBA techniques available, with nuclear reaction

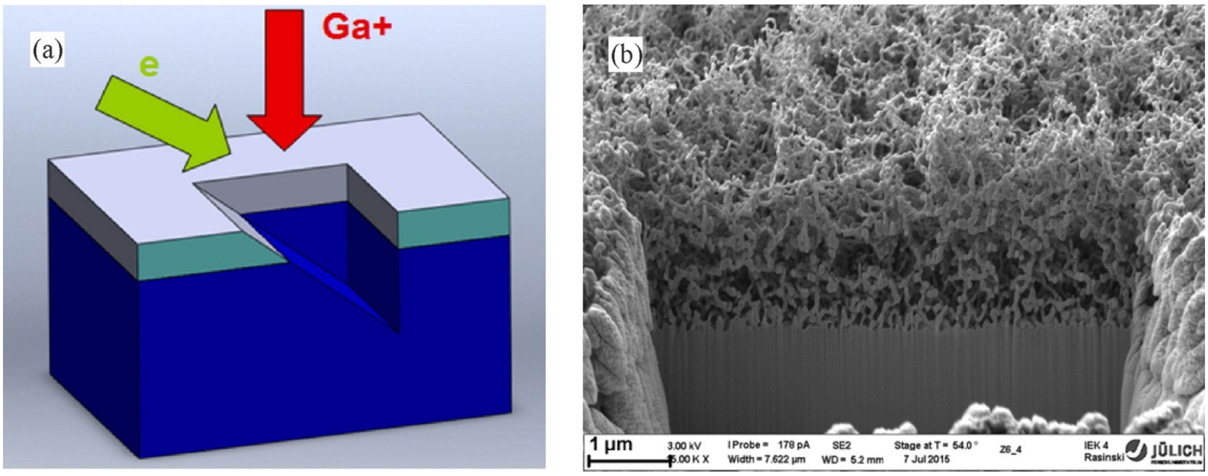

Figure 4. (a) Scheme of FIB application; (b) SEM image of a FIB cross section for nano-structured surface of a tungsten sample exposed to He plasma, also known as tungsten fuzz [11].

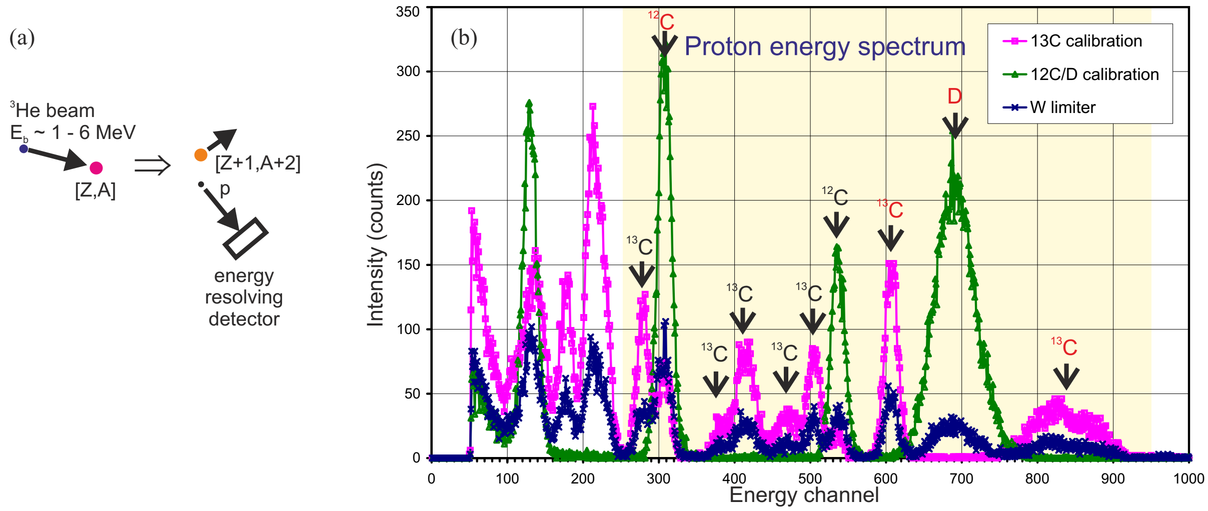

Figure 5. (a) Measurements scheme of NRA and (b) examples of NRA spectra for carbon-deuterium co-deposition layer on a tungsten test limiter after a $^{13}CH_4$ tracer injection experiment in TEXTOR as well as ^{12}C/D and ^{13}C reference samples [13]. Peaks from reactions which were used for the analysis are marked red.

analysis and Rutherford back-scattering being the most frequently used in the fusion related PMI research.

For nuclear reaction analysis (NRA), a ^{3}He beam in a range of energies 1-6 MeV is usually used, because ^{3}He reacts with many fusion relevant materials. The frequently used nuclear reactions with ^{3}He are: D (^{3}He,p) ^{4}He, ^{12}C (^{3}He,p) ^{14}N, ^{13}C (^{3}He,p) ^{15}N, ^{9}Be (^{3}He,p) ^{11}B and ^{11}B (^{3}He,p) ^{13}C, all of them having protons as one of the reaction product. The detection of further elements using other beams, i.e. H, D and Li, and reactions is possible. Figure 5(a) shows a measurement scheme of NRA. The method can determine the absolute amount of material in a maximal analysis depth of ~1-10 μm, depending on the material and the ion beam energy. It has a moderate sensitivity of ~10^{15} atoms/cm^2. The amount of the material can be determined by a cross-calibration with a sample with a known amount of material, or by using available reaction cross-section. Most of the PMI relevant reactions are incorporated in the IBA data analysis software SIMNRA [12]. A typical lateral resolution of ~1 mm, corresponding to the beam diameter, can be reduced down to ~10 μm using the μ-NRA technique. NRA has a poor depth resolution, which can be improved by the use of beams of different energies and therefore different probing depths for the same sample. Figure 5(b) shows examples of NRA spectra for ^{12}C/D and ^{13}C reference samples and from a mixed ^{12}C/^{13}C/D layer deposited on a tungsten test limiter in the tokamak TEXTOR [13].

For Rutherford back-scattering (RBS), a ^{4}He beam in a range of energies 1-6 MeV is usually used. A ^{3}He beam can also be used, thus RBS and NRA measurements can be combined. However, the market price of ^{3}He gas is significantly higher than of ^{4}He. Figure 6 shows a scheme of the RBS technique and an example of RBS spectrum. The energy of backscattered beam particles is $E_R = K(m_b, m_s, \theta, d) \times E_b$, where the kinematic factor K is a function of the beam particle mass m_b, scattering particle mass m_s, scattering angle θ and depth d and E_b is the beam energy. The method provides quantitative depth profiles of elements on the sample surface. The maximal analysis depth is typically ~5 μm. The technique has a high sensitivity ~10^{12} atoms/cm^2. The latera resolution is of ~1 mm (beam diameter), similarly to NRA. RBS suffers from a pure discrimination of heavy elements with similar masses and has a limited applicability for mixed material layers and high-Z substrates.

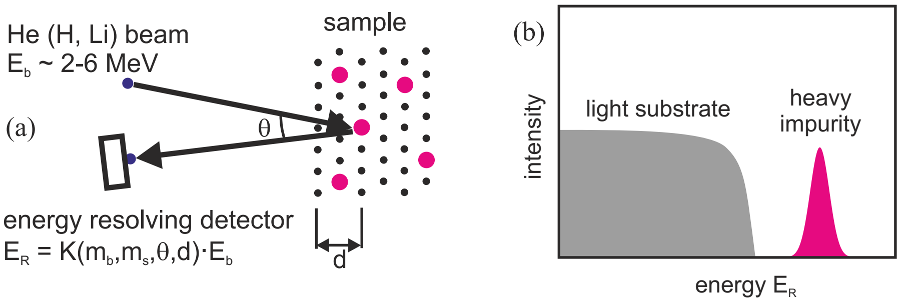

Figure 6. (a) Measurements scheme of RBS and (b) typical RBS energy spectrum for a heavy impurity on a light substrate.

3.4 Secondary ion mass spectrometry (SIMS)

Secondary ion mass spectrometry (SIMS) utilises ion probing beams in a range of energies of ~1-30 keV. The primary beam causes the emission of secondary ions from the sample, which are then analysed by a mass spectrometer delivering information on the elemental surface composition. Figure 7(a) shows the scheme of a SIMS apparatus. It usually incorporates at least two primary ion beam sources, e.g. an oxygen primary ion beam for probing of electropositive surface constituents and a caesium primary ion beam for electronegative elements. In some SIMS devices, an additional ion beam is used for the surface sputtering to obtain a high quality depth profiling. The method has a high lateral resolution of ~100 nm - 1 μm. The modern SIMS devices, especially those equipped with a time-of-flight (TOF) mass spectrometer, have a high mass resolution and are able to resolve different molecular fragments. One drawback of the method is that only sputtered ion are analysed. The neutrals, which are the majority of sputtered particles, get lost. The process of the production of secondary ions is complex. Therefore, SIMS typically delivers qualitative but not quantitative information on the material composition. If the depth profiling is applied, the sputtering rate of the SIMS apparatus can be calibrated by measuring the depth of the SIMS crater by a surface profilometer.

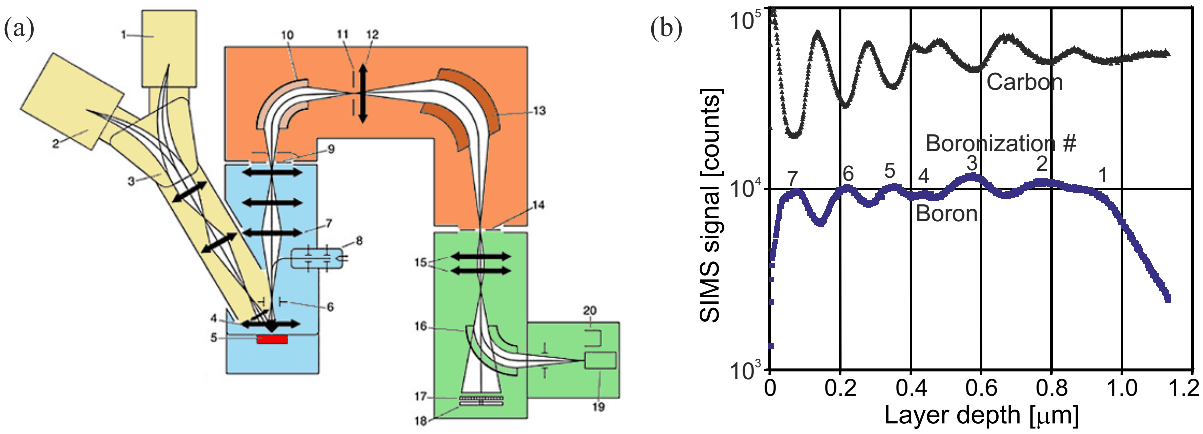

Figure 7. (a) Scheme of a SIMS apparatus: (yellow) primary ion column consisting of (1) and (2) ion sources, (3) primary beam mass filter, (4) immersion lens, (blue) secondary ion extraction system consisting of (5) specimen, (6) dynamic transfer system, (7) transfer optical system, (8) electron flood gun, (orange) mass spectrometer consisting of (9) entrance slit, field aperture, (10) electrostatic analyser, (11) energy slit, (12) spectrometer lens, (13) electromagnet, (14) exit slit, (green) detection system consisting of (15) projection lenses, (16) electrostatic sensor, (17) and (18) ion detectors and (19) Faraday cup [14]; (b) SIMS depth profile of a layer deposited on a collector probe installed for one experimental campaign in TEXTOR showing anti-correlated signals of carbon from the tokamak operation and boron from the boronizations [15].

Figure 7(b) shows an example of the SIMS depth profiling of a mixed layer deposited on a collector probe during a full experimental campaign in the tokamak TEXTOR [15]. The SIMS measurement was stopped when the signals dropped by a factor of two, corresponding to the bottom of the layer. Then the crater depth of 1.1 μm was measured by a stylus profilometer, thus, the sputtering rate of SIMS was obtained. The layer consists mainly of carbon from the normal plasma operation, with carbon being the main plasma impurity in TEXTOR, and boron from the boronisation wall conditioning. Seven boronisations were performed during the experimental campaign, which is reflected in the SIMS depth profiling.

4. Measurements of erosion and deposition

4.1 Quartz microbalance (QMB)

The quartz microbalance (QMB) technique uses the fact that the quartz (SiO_2) crystals have a resonant frequency changing with their mass. Quartz crystal microbalance (QCM) is a different notation for the technique. For commercially available quartz crystals for QMB, the resonant frequency is usually a few MHz, decreasing when gaining mass with a rate of $\sim 10^{-08}$ g/Hz, thus providing a high mass sensitivity corresponding to about one monolayer of deposition on the quartz surface. Electronics measuring the crystal frequency have to be placed close to the detector, which increases the complexity of the QMB application in fusion devices. Another drawback is the sensitivity of the resonant frequency on the surrounding temperature. In fusion devices the particle flux is typically accompanied by a significant heat flux. Therefore, the influence of temperature on the QMB signal has to be monitored. It is often done by a second, reference, quartz, which is placed close to the first, measuring quartz. The reference quartz has similar temperature as the measuring one, but is mechanically covered to prevent any deposition on it and to keep its initial mass. Thus, the reference quartz has only the temperature response and can therefore be used for the temperature compensation of the measuring quartz.

The piezoelectric constant of quartz drops sharply for temperatures >300°C and reaches zero at the Curie point of 573°C. Therefore, QMB systems are typically installed in remote areas of a fusion device, where the heat flux is low. Material eroded from plasma facing components under the plasma impact is transported to remote areas where it is collected by QMB systems. Therefore, although actually measuring deposition, QMB systems are used as in-situ

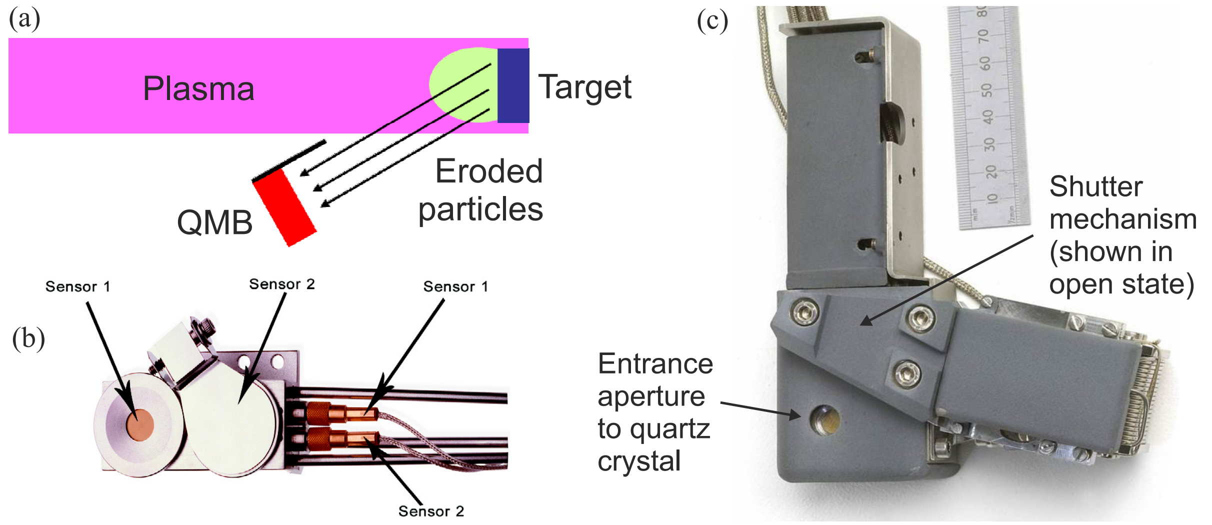

Figure 8. (a) Measurement scheme of QMB; (b) commercial dual QMB sensor [16]; (c) QMB diagnostic used at JET is protected by a graphite cover against heat loads [17,18].

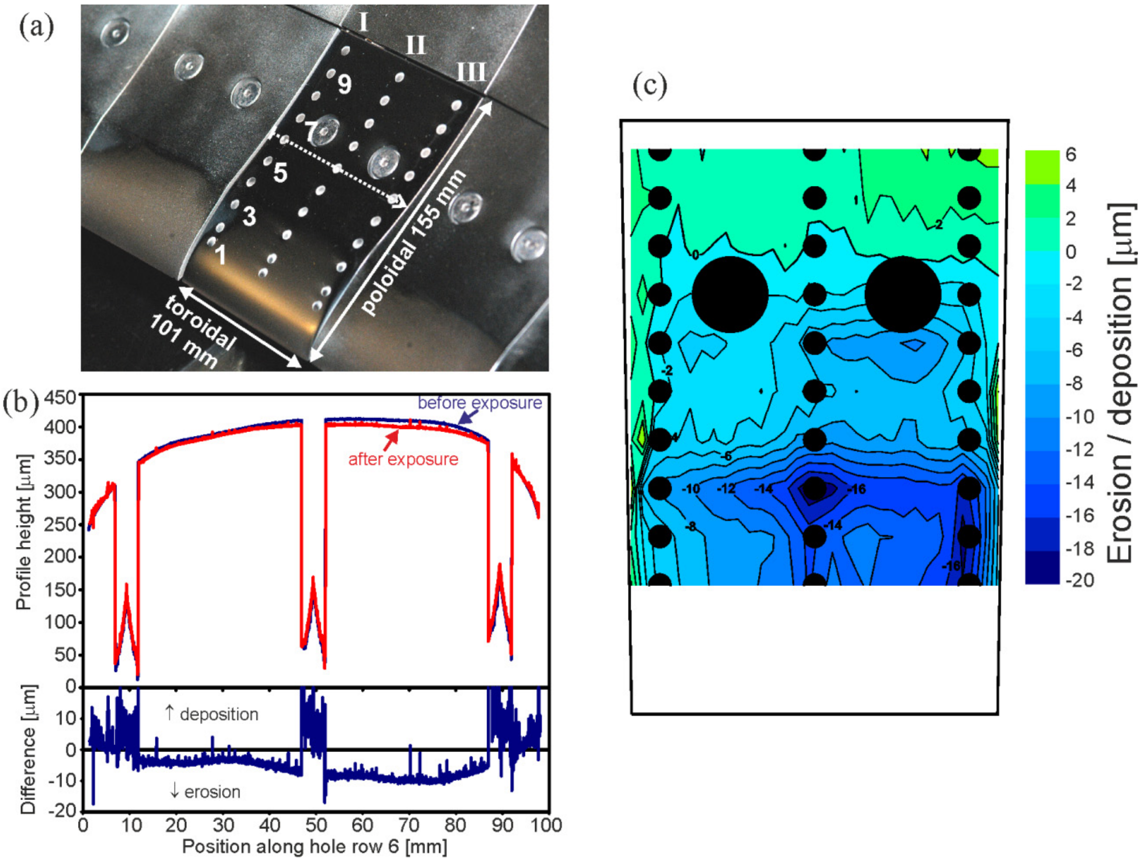

Figure 9. (a) View of the special limiter tile in TEXTOR after the exposure for one experimental campaign. The dimensions of the tile and the positions of the marker holes are indicated. The dotted line represents the position and the direction of the X axis in (b); (b) surface height profile measured by optical profilometry before and after the exposure; (c) 2D reconstruction of the erosion and deposition pattern of the limiter tile. Negative values represent erosion, positive values deposition.

erosion monitors of the plasma facing components.

Figure 8 shows the scheme of QMB application in a plasma device as well as photographs of commercially available QMB systems [16] and of a QMB system for the in-situ application in the tokamak JET [17,18].

4.1 Application of marker tiles

There are numerous methods which can quantify the amount of deposition on a certain sample or a tokamak wall tile. It is significantly more complicated to measure the erosion, something which is absent. A well-established method is marker techniques. Markers have to be prepared on samples or wall tiles in advance, before their exposure. The wall components equipped with markers are also called "smart tiles". Typically the installation of "smart tiles" at the wall needs the air vent of the vessel, which is done between experimental campaigns. Therefore, unlike the in-situ QMB method, the marker technique is campaign-averaged. Markers can be e.g. well-defined layers of material deposited on a substrate in a combination suitable for the applied surface analysis techniques. The marker layers are characterised before and after the exposure. The difference is then the amount of eroded material. Techniques like RBS or SIMS are often used for the marker characterisation.

Another approach of markers is the application of reference points on the sample surface which are not subjected to erosion. The reference points can be e.g. recessed grooves or pits, which bottom is shaded from the eroding plasma flux. Figure 9(a) shows a marker tile of the main toroidal limiter in TEXTOR made of isotropic graphite, which was used to determine the erosion-deposition pattern over the full tokamak campaign [19]. 3 × 10 drill holes were used as the reference points. It was found out by SIMS that up to several μm of deposition was accumulated in the recessed holes during the experimental campaign. It was taken into account when comparing the surface profiles measured by the optical surface profilometry before and after the exposure (figure 9(b)). A full erosion-deposition pattern of the tile surface was reconstructed by combining the surface profiles across the recessions done in both toroidal and poloidal directions (figure 9(c)).

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