A quantitative analysis of hydrogen isotope retention in tokamak first wall materials by optical spectroscopy and mass-spectrometry

© E.V. Smirnova[1–3], O.S. Medvedev[1,4], A.G. Razdobarin[1,2], D.I. Elets[1,2,4], L.A. Snigirev[1,2], I.V. Miroshnikov[1], I.M. Bukreev[1], Yu.M. Gasparyan[2,4]

[1] Ioffe Institute, St. Petersburg, Russia
[2] St. Petersburg State University, St. Petersburg, Russia
[3] ITMO University, St. Petersburg, Russia
[4] National Research Nuclear University „MEPhI", Moscow, Russia
E-mail: evsmirnova@mail.ioffe.ru

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In this work, the opportunity of laser-induced ablation for diagnostics of hydrogen isotope retention is demonstrated. The analysis of residual gases based on mass spectrometry and Penning discharge optical spectroscopy were used as methods for measuring the deuterium flux. It is shown that the optical spectroscopy of the Penning discharge measures a larger number of deuterium atoms than mass spectrometry analysis. The difference of the quantitative evaluation relates to the registration of D_2, H_2, HD molecules by mass spectrometer, while the Penning discharge optical spectroscopy shows the presence of protium and deuterium regardless of the chemical composition of the material.

Keywords: Hydrogen isotopes, Penning discharge spectroscopy, mass-spectrometry, laser-induced ablation, tokamak first wall, tritium retention diagnostic.

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Thermonuclear reactor such as ITER offer operation modes with a deuterium–tritium (D–T) mixture used as fuel. Hydrogen isotopes tend to accumulate in structural materials of thermonuclear facilities that come into contact with plasma (the first wall and the diverter). The retention of thermonuclear fuel has the following negative effects: (1) limited reactor service life due to accumulation of radioactive tritium; (2) possible release of isotopes from the walls during a discharge, which alters the ratio of fuel components; (3) low economic efficiency [1,2].

Ex-situ methods, such as thermal desorption spectroscopy and nuclear reaction analysis, are used widely to study the accumulation of hydrogen isotopes in laboratory conditions. However, in the case of thermonuclear reactors, these methods require either vent of the vacuum vessel, which entails a significant expenditure of time and money, or the presence of witness samples, which limits the regions of study [3]. Due to the radioactivity of tritium and the limited capabilities of *ex-situ* methods, *in-situ* methods for diagnostics of the retention of hydrogen isotopes in the first wall of a tokamak based on the interaction of laser radiation with matter are currently being developed. Laser diagnostic methods are free from the above-mentioned disadvantages inherent in *ex-situ* methods; in addition, they exclude the influence of isotopic exchange upon contact of samples with the atmosphere and the interaction of personnel with radioactive structural elements. Various methods of residual gas analysis are potentially suitable for quantitative measurements: mass spectrometry, optical Penning discharge spectroscopy, and laser-induced breakdown spectroscopy [3,4].

Mass spectrometry is a widely used method for measuring the concentration of resudial gas components. However, quantitative analysis requires calibration for each mass being monitored. The detection system sensitivity depends on the characteristics of the vacuum system and varies from one chemical compound to another.

Penning discharge emission spectroscopy involves the analysis of intensity of lines of the chemical elements being studied upon excitation of a neutral gas. The change in intensity depends linearly on the concentration of elements in the gas mixture [2,3]. The spectral positions of the Balmer α-lines of hydrogen isotopes (H_α — 656.27 nm, D_α — 656.11 nm, and T_α — 656.03 nm) require a high resolution spectrometers (no worse than 0.1 Å). The use of a Penning

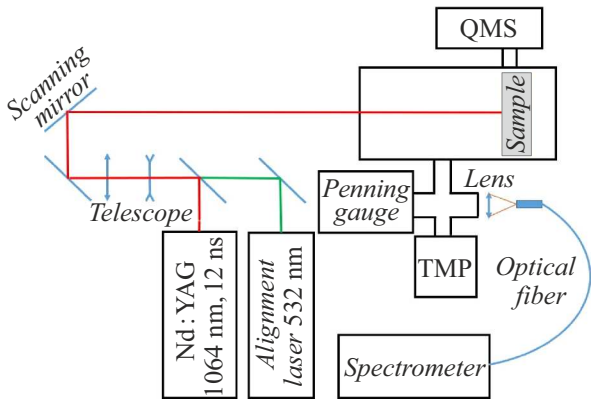

Figure 1. Experimental setup of laser diagnostic of hydrogen isotopes retention in first wall elements.

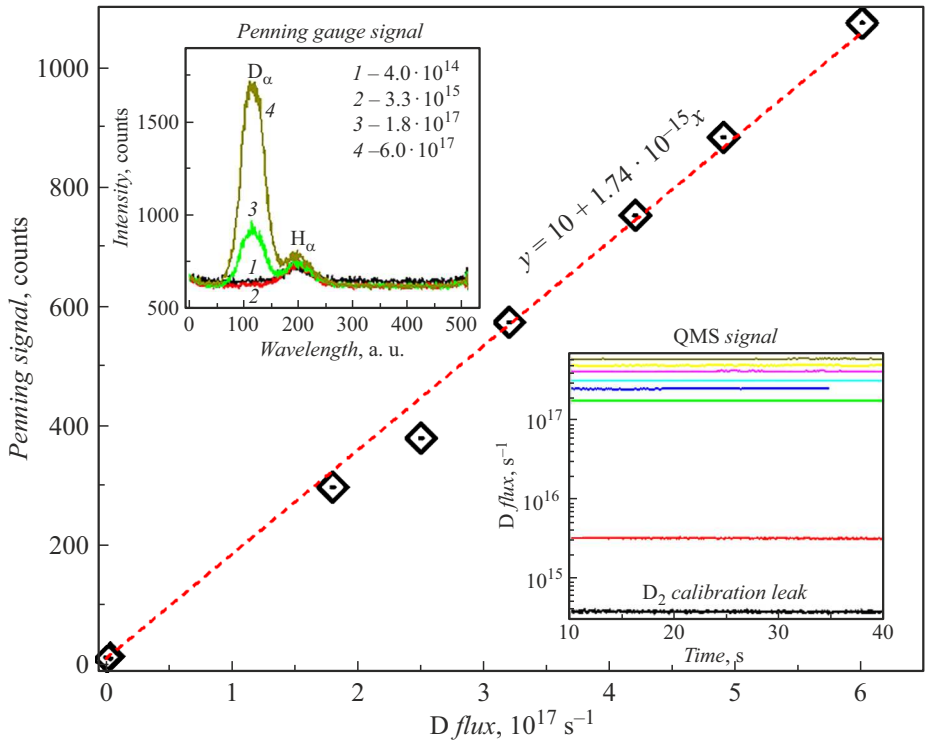

Figure 2. Dependence of the D_α lines intensity on the flux of pure deuterium. The upper inset shows the initial signal recorded in a Penning cell with an exposure time of 10 s. The atomic flux recalculated from the QMS signal is presented in the lower inset.

Flux of deuterium atoms (in s^{-1}) recorded by quadrupole mass spectrometry and Penning discharge optical spectroscopy

Detection method	Sample	
	FWE_3	FWE_5
QMS	$4.3 \cdot 10^{15}$	$1.3 \cdot 10^{16}$
Penning cell	$2.2 \cdot 10^{16}$	$3.4 \cdot 10^{16}$

cell at a residual gas pressure of 10^{-5}–10^{-4} Pa translates into a low intensity of recorded lines of hydrogen isotopes; therefore, a high spectrometer luminosity and highly sensitive detectors based on EMCCD, ICCD, or ICMOS are needed to record the signal. Penning discharge optical spectroscopy has the following advantages: the capacity to detect He as a product of the D–T reaction, operate within the 10^{-5}–10^{-1} Pa pressure range [5], and detect hydrogen isotopes regardless of the chemical compound (HD, HDO, C_xD_y, etc.) [6].

In the present study, the possibility of application of laser-induced ablation combined with quadrupole mass spectrometry (QMS) and optical spectroscopy of a Penning discharge in quantitative analysis of the hydrogen isotope content in redepositions on first-wall elements (FWEs) is demonstrated for the first time.

The experimental setup (Fig. 1) features a laser radiation input system and a Penning discharge light collection system. A modified Alcatel CF2P cell with the magnetic field strength and the potential difference raised to 0.18 T and 5 kV, respectively, was used to increase the plasma glow intensity in a Penning discharge [3]. The detection system included a lens with a fiber-optic connector and a spectrometric analysis system based on an SPT-DDHR-4 high-resolution diffraction spectrometer. The distance from the Penning cell to the lens was 0.4 m.

A vacuum stand with a volume of 70 l was used for the study. The base pressure was $8 \cdot 10^{-5}$ Pa. A pulsed Nd:YAG laser with a radiation wavelength of 1064 nm, a pulse duration of 12 ns, a pulse energy of 450 mJ, and a pulse repetition rate of 10 Hz was used as a radiation source. In the ablation mode, the energy density was 10 J/cm^2 with a spot diameter of 0.8 mm. The samples were scanned with a step of 0.5 mm; the measurement time per point was 0.1 s.

To determine the optical sensitivity of the system to a deuterium flux, the signal was calibrated by introducing pure deuterium into the vacuum chamber via the gas injection system. In the process of calibration, absolute values of gas fluxes were measured using calibrated data from the quadrupole mass spectrometer. The dependence of intensity of D_α lines on the deuterium flux was approximated by a linear dependence with a slope coefficient of $1.74 \cdot 10^{-15}$ (Fig. 2). The full range of fluxes used in calibration was determined by the QMS characteristics and the gas injection system and varied from $\sim 2 \cdot 10^{17}$ s^{-1} at a chamber pressure of $1.8 \cdot 10^{-3}$ Pa to $6 \cdot 10^{17}$ s^{-1} at a pressure of $5.6 \cdot 10^{-3}$ Pa.

In the current configuration of the experimental setup, a reliably detected deuterium flux level is $\sim (2-5) \cdot 10^{16}\,\mathrm{s}^{-1}$.

Samples of hydrocarbon deposits on the tungsten lining of the diverter exposed at the Globus-M2 tokamak [7] were used in laser-induced ablation experiments. Layers with a thickness of 300–600 nm (FWE_3 in [7]) and 5–6 μm (FWE_5 in [7]) were studied in this series of experiments. The D_α line intensity in the Penning cell and the signal of the 3rd and 4th masses in QMS were recorded in the process of ablation of hydrocarbon deposits. According to the QMS data (with only the 3rd and 4th masses taken into account), the surface density of deuterium atoms is $\sim 4 \cdot 10^{17}$ and $\sim 2.5 \cdot 10^{18}\,\mathrm{cm}^{-2}$, respectively.

The fluxes of deuterium atoms determined by QMS and Penning discharge optical spectroscopy are listed in the table. A coefficient of $1.74 \cdot 10^{-15}$ obtained as a result of calibration was used to calculate the flux of deuterium atoms from the intensity of the optical signal of the D_α line. It can be seen from the table that the deuterium flux calculated by the optical method is 3–4 times higher than the flux determined by QMS with only the 3rd and 4th masses recorded. One possible reason for the observed difference is the potential release of deuterium as part of volatile hydrocarbons in the case of redepositions at the Globus-M2 tokamak with a graphite first wall. Therefore, when QMS is used, one needs to monitor a significantly larger number of masses to perform a more accurate quantitative analysis, which, in turn, will require calibration for each recorded mass. Another reason is the unaccounted contribution of atomic and molecular fractions to the H_α and D_α line signal intensity in a Penning discharge, which is related to their different emission cross sections [8].

Thus, the feasibility of application of a Penning cell in analysis of the accumulation of hydrogen isotopes in hydrocarbon deposits at the Globus-M2 tokamak has been demonstrated for the first time. Similar experiments with films made of other deuterium-saturated materials (e.g., tungsten, which excludes the possibility of formation of volatile hydrocarbons) are necessary for a more in-depth examination of the observed discrepancy between the results of optical spectroscopy and mass spectroscopy. To use Penning discharge optical spectroscopy as a method for quantitative analysis of hydrogen isotope content, one needs to improve the experimental data processing model and raise the sensitivity of the optical signal recording system for the purpose of expanding the dynamic range.

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Conflict of interest

The authors declare that they have no conflict of interest.

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