INTERMEDIATE REPORT 3

Investigation of Water Fragments

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Feasibility of utilizing of Vis/IR system and proposals on its enhancement allowing realization of spectroscopic method over the band range 615-700nm

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1. Short summary of measurements and calculations

The main results described in Report 1 and 2 can be summarized as follows:

1. The emission spectra of hollow cathode discharge were studied in details in the interval 300-900 nm. The gases are the water vapor or its mixture with rare gases He, Xe. This type of discharge can be considered as a model for the examination of water leaks at the places of blanket joints in ITER water cooled wall.

2. Spectra have been recorded with high spectral resolution ~0.04 nm, intensity dynamic range ~10^4 and signal to noise ratio $S/N>10$.

3. Optical transitions have been identified and the emissivity for the most of them was measured in absolute scale.

4. Even for such relatively simple gas mixtures composition plasma emission demonstrates the developed atomic, molecular and ion spectra. The most developed spectra are concentrated in the short wavelength interval 300-600 nm thus providing the flexible approaches for water leaks diagnostics.

5. The most characteristic electronic molecular bands of O$_2$ (Shuman-Runge) and H$_2$O (Kitagawa) have not been observed.

6. The main purpose of these studies was in searching the possibility for water and water fragments spectroscopy in the region of 615-700 nm with the center at the H$_\alpha$ hydrogen line 656nm. In this region quite few lines are presented. These are the rather strong lines of H, He and Xe and very slow H$_2$ bands.

2. Concepts for spectroscopic method for water leaks diagnostics

1. Presently the most promising water leaks diagnostics remains the approach based on the registration of OH lines intensities near 300 nm. Being only preliminary and qualitative in the early studies in 2002-2012 it is developed now as a strict absolute well localized technique. The possibility to measure the water vapor fluxes at the level 10^{-7} Pa\cdot m^3\cdot s^{-1} is demonstrated [1]. The problem is that the optics in UV might be not stable enough in the conditions of ITER operation mode. If this problem can be solved then this technique can be considered as the most ready to apply.

2. Only lines of atomic hydrogen as a potential water fragment are presented in 615-700 nm range at reasonable intensity (10^4 dynamic range). The absolute amount of H in plasma can be determined by the methods under developing now in Lebedev Institute (LPI) but the origin of atoms might be questionable in ITER conditions. On the other hand the measurements of H
atoms concentration might be useful in the "cold" mode of ITER operation during the cleaning of
the walls by discharge in rare gases for searching the leaks.

3. Despite the mentioned above disappointing fact of absence of the other then hydrogen
characteristic water fragments strong enough spectral lines in the 615-700 nm it is possible to
provide the new possibilities at a slightly longer wavelengths. In the range 750-850 nm the lines
of Xe and atomic oxygen O lines are observed. Recently in LPI the new technique was
demonstrated for the measurements of absolute O atoms density using the combination of these
lines intensities [2]. If the Vis/IR optics is stable in this spectral region then this technique can be
specially elaborated in more details and applied in rather simple technical form. The photo-
detectors in this region are sill high sensitive. It can be useful for different aspects of ITER
operation including water leaks detection also. It have to be noticed that the Xe in this technique
is not suggested to be used as a marker solved in water (discussed previously but without clear
enough perspectives) but has to be injected directly in plasma camera in controlled trace
amounts.

4. Performing the measurements of the densities of H$_2$O molecules (in UV) and O atoms
(in red and near IR) systematically it is possible to study the correlations between the densities of
these particles. The example of such preliminary measurements is shown in fig. 1 [3]. In this case
the O line 777.2 nm and Xe line 823 nm are used.

5. After some additional elaboration the technique of absolute particle densities
measurements can be extended for the atomic hydrogen and deuterium atoms. One of the
convenient spectral structures is the H$_\alpha$- D$_\alpha$ (656nm) doublet. The lines can be distinguished even
at moderate spectral resolution like that was realized with the small size spectrometer in LPI
experiments. The example is shown in fig. 2.
Fig. 2. Hα - Dα (656nm) doublet. Recorded with the help of spectrometer MDR-204 (used in optical scheme of LPI)

The spectra of similar resolution have been recorded also with the help of the spectrometer included in the MEPhI installation.

Such type of measurements can be used to establish the dependencies like that in fig.1 but for hydrogen and deuterium atoms. The penetration of H₂O vapors in plasma chamber can lead to the deviation of H/D ratio from those in the plasma background thus can be used for the water leakage flux. This proposal needs in quantitative studies.

Some additional comments on the intensity of Hα radiation in cleaning discharge can be added. Usually the concentration of H* atoms is proportional to H⁺. Fig.8 (INTERMEDIATE REPORT 2) demonstrates that in He plasma the addition of the water vapor leads to predominant generation of H⁺ ions relative to H₂⁺ ions. With injection of molecular hydrogen, in contrary, the H₂⁺ peak prevails. This effect can be related to primary formation of H atoms during water molecules fragmentation in plasma. In addition to the water molecule hydroxyl OH dissociation and hydrogen atom H excitation by an electron impact, some principal plasma-chemical reactions can be included into consideration. Namely.

In the presence of excited metastable He⁺ atoms (19.8 eV) water dissociation can be accompanied with H excitation or even ionization:

\[ \text{He}^+ + \text{H}_2\text{O} \rightarrow \text{H}^+ + \text{OH} + \text{He}. \]

Dissociation of water molecules in plasma can be also stimulated by a vibrational excitation:
\[ \text{H}_2\text{O}^* + \text{H}_2\text{O} \rightarrow \text{H} + \text{OH} + \text{H}_2\text{O}. \]

With electron excitation of hydrogen atom to 3p sublevel (12.09 eV) the H\text{\textalpha} quantum can be radiated.

Dissociative attachment can also influence the fragmentation of water molecules:

\[ e + \text{H}_2\text{O}^* \rightarrow \text{H}^- + \text{OH}. \]

This reaction is followed by electron detachment:

\[ \text{H}^- + e \rightarrow \text{H} + e + e. \]

Electrons can be involved in dissociative process many times (playing the role of catalyzer) and this mechanism of hydrogen atoms generation from water molecules can be very effective. So, for obtaining of maximum H\text{\textalpha} radiation in cleaning He discharge, its parameters are to be optimized.

The possibility of H\text{\textalpha}/D\text{\textalpha} ratio measurement for water leakage detection was detailed demonstrated at our experiments in PR-2 due to the effect of isotopic shift of the emission lines of deuterium and hydrogen atoms. It is possible to apply this method for finding water leakage in ITER, using Vis/IR optical system for SOL emission monitoring. In a strong magnetic field relevant to ITER the high-resolution spectrometer is needed for this purpose. So for this purpose the spectrometers with ~ 0.01 nm resolution should be included into the Vis/IR procurement.

3. Possible increasing of H\text{\textalpha}/D\text{\textalpha} method sensitivity with auxiliary laser

The lower level of hydrogen Balmer series has the metastable 2s sublevel (about 10.2 eV) with living time of about 2ms. The concentration of H atoms (as fragments of H\textsubscript{2}O dissociation) in this metastable state in cleaning discharge can be rather high. The same is true for D atoms. But as its energy of excitation is slightly higher, population of 2s H\textsuperscript{+} atoms exited due to transfer from 2sD\textsuperscript{+} states will increase. Charge exchange collisions of D\textsuperscript{+} atoms with H\textsuperscript{+} ions can also enhance the population of 2sH\textsuperscript{+} states. The following excitation of H\textsuperscript{+} by electrons with thermal energies (of about 2eV) to 3p level responsible for H\text{\textalpha} can enhance the ratio of H\text{\textalpha}/D\text{\textalpha} emission and as result to increase sensitivity of the H\text{\textalpha}/D\text{\textalpha} method.

The metastable 2sH\textsuperscript{+} state can be also used for resonance laser light absorption and excitation to the 3p level. The wavelength for hydrogen excitation to 3p state with 2s-3p transition 656.274 nm hydrogen and 656.094 for deuterium. Provided the auxiliary light beam generated by a dye laser with corresponding to H 2s-3p line wavelength illuminates the edge of discharge plasma with water plume, the induced local emission of this line would take place only for 2sH\textsuperscript{+} atoms. The sensitivity and contrast of H\text{\textalpha}/D\text{\textalpha} method for water vapor ingress localization in this regime of the laser induced fluorescence can be risen manifold.
One of the drawbacks of laser fluorescence technique is the laser light reflection and dispersion by walls. So, the light beam must be directed parallel or with minimal grazing angle relative to wall surface for space separation of spots were the light beam intersects the wall and were the induced H⁺ luminescence is analyzed. In principle, this drawback can be overcome with the use of vacuum ultraviolet monochromator for Lyβ registration of H⁺ luminescence (12.09 eV, 102.57 nm) but it is far from Vis/IR spectral range.

Rather long living time of 2sH⁺ states excited in cleaning discharge can be used in another variant of water leak localization method. Namely, a laser beam with resonant wavelength 656.3 nm after reflection from the wall can be included into laser sensitive to resonance absorption of this wavelength. So, the reflected (dispersed) laser beam part passes through the “water” plume with metastable atoms two times before returning into the active media. In this case registration system must react on appearance of Hα by decreasing signal in the absorption spectrum. So, on this basis some kind of water leakage laser LIDAR can be proposed.

4. Conclusion

The results of this contract fulfillment can be summarized as follows:

1. In 615-700 nm wavelength range only lines of atomic hydrogen as a potential water fragment are presented in 615-700 nm range at reasonable intensity (10⁴ dynamic range).
2. It is possible to provide the new possibilities at slightly longer wavelengths (within high transmitting efficiency of Vis/IR) in the range 750-850 nm with lines of Xe and atomic oxygen O.
3. For measurements of absolute density of O atoms, technique with combination of O and Xe lines intensities elaborated in Lebedev institution can be used (with very limited controlled Xe injection directly into plasma camera).
4. Measurements of Hα-Dα (656nm) doublet ration can be used for water leak detection. Absolute particle densities measurements can be extended for the atomic hydrogen and deuterium atoms after some additional elaboration.
5. It is shown that in cleaning discharge in He enhanced generation of atomic hydrogen from water molecules takes place with subsequent excitation to long living metastable 2sH⁺ level and Hα radiation.
6. The possibility of Hα/Dα ratio measurement with water ingress into plasma was demonstrated in experiments in PR-2 facility. It is possible to apply this method for finding water leakage in ITER, using Vis/IR optical system for SOL emission monitoring. In a strong magnetic field relevant to ITER a high-resolution spectrometer (~ 0.01 nm resolution)
is needed for this purpose. So, for water leak detection based on H\(_{\alpha}\)/D\(_{\alpha}\) ratio measurement such spectrometer should be included into the Vis/IR procurement.

7. It is suggested that sensitivity of the H\(_{\alpha}\)/D\(_{\alpha}\) method for leak localization can be increased with laser induced fluorescence. Provided the auxiliary light beam generated by a dye laser with corresponding to H 2s-3p line wavelength illuminates the edge of discharge plasma with water plume, the induced local emission of this line would take place only for 2sH\(^*\) atoms. The sensitivity and contrast of H\(_{\alpha}\)/D\(_{\alpha}\) method for water vapor ingress localization in this regime of the laser induced fluorescence can be risen manifold.

8. Some kind of water leakage laser LIDAR with dry laser and registration of absorption spectrum near H\(_{\alpha}\) wavelength can be also considered.
References

