Simultaneous Measurements of Work Function and H⁻ Density Including Caesiation of a Converter Surface

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Abstract. Negative hydrogen ion sources rely on the surface conversion of neutral atomic hydrogen and positive hydrogen ions to H^-. The efficiency of this process depends on the actual work function of the converter surface. By introducing caesium into the source the work function decreases, enhancing the negative ion yield. In order to study the impact of the work function on the H^- surface production at similar conditions to the ones in ion sources for fusion devices like ITER and DEMO, fundamental investigations are performed in a flexible laboratory experiment. The work function of the converter surface can be absolutely measured by photoelectric effect, while a newly installed cavity ring-down spectroscopy system (CRDS) measures the H^- density. The CRDS is firstly tested and characterized by investigations on H^- volume production. Caesiation of a stainless steel sample is then performed in vacuum and the plasma effect on the Cs layer is investigated also for long plasma-on times. A minimum work function of (1.9±0.1) eV is reached after some minutes of plasma treatment, resulting in a reduction by a value of 0.8 eV compared to vacuum measurements. The H^- density above the surface is $(2.1\pm0.5)\times10^{15}$ m^{-3}. With further plasma exposure of the caesiated surface, the work function increases up to 3.75 eV, due to the impinging plasma particles which gradually remove the Cs layer. As a result, the H^- density decreases by a factor of at least 2.

INTRODUCTION

The work function is an important parameter for negative hydrogen ion sources based on surface conversion of neutral atomic hydrogen and positive hydrogen ions. In particular a low work function is required in order to enhance the negative ion formation. For this purpose, caesium is introduced in negative hydrogen ion sources, since it reduces the converter work function [1, 2]. The effect of partial caesium coverage on the work function χ of a metallic substrate is well known under ultra-high vacuum conditions (background pressure below 10^{-9} mbar) [3–5]. The surface work function decreases with Cs coverage reaching a minimum of 1.5–1.7 eV at 0.5–0.7 monolayers. Between 0.5–0.7 and 1 monolayer the work function increases again, until the bulk Cs work function is obtained (2.14 eV) [6]. For coverages above 1 monolayer the bulk Cs work function is maintained. However in negative hydrogen ion sources the experimental conditions are different: the background pressure is higher (10^{-7}–10^{-6} mbar) and formation of Cs compounds with residual gases can affect the work function in vacuum, as shown in [7]. Furthermore, the low-temperature hydrogen plasma interacts with the converter surface, resulting in a redistribution of the Cs atoms. Additionally, this interaction can lead to a cleaning effect of the Cs layer performed by plasma particles, like neutral hydrogen atoms and hydrogen positive ions, probably together with the UV light emitted by de-excitation of neutral hydrogen, producing a decrease of the work function [8, 9].

The impact of the surface work function on the negative hydrogen ion yield has been studied in several works, both theoretically [10] and experimentally by scattering of protons on caesiated surfaces in UHV conditions [11–13]. In plasma environment, some investigations on the correlation between H^- yield and work function are presented, e.g. [14–18]. In these studies, the work function of the converter surface is obtained by the photoelectric method using an Ar^+ laser and by conversion of the photoelectric currents to the quantum efficiencies, which depend on the work function and on the photon energy. However, the relationship between the work function and the quantum efficiencies at the laser wavelengths is achieved in a different experimental setup, a high-vacuum test chamber for work function measurements. The converter work function is then obtained by comparison between the quantum efficiencies in the two different apparatus. Thus an absolute quantification of the work function was not possible.

Absolute measurements of work function at the same plasma conditions of negative hydrogen ion sources are highly desirable for the investigations on the correlation between work function and H^- density above the surface, in order to reduce the Cs consumption in view of ion sources for fusion devices like ITER and, beyond ITER, DEMO. Since the work function is not easily accessible in ion sources, dedicated investigations are performed in a laboratory experiment, where the work function can be absolutely determined by the photoelectric effect and to which a cavity ring-down spectroscopy system has been recently installed for absolute H^- density measurements. The plasma parameters of the planar ICP discharge are close to the ones of ion sources. Cs is evaporated into the chamber and the experiment is equipped with several diagnostic systems, which can be operated simultaneously. In the present paper, the work function of a caesiated sample is monitored after plasma exposure and its influence on the H^- density is investigated.

EXPERIMENTAL SETUP

The laboratory experiment ACCesS (Augsburg Comprehensive Cesium Setup) [7, 19, 20] consists of a cylindrical stainless steel vessel, with a diameter of 15 cm and a height of 10 cm, as shown in Fig. 1. The background pressure within the vessel is of the order of 10^{-6} mbar. A planar coil is located on the top of the vessel and generates the low temperature plasma via inductive RF coupling (frequency 27.12 MHz, maximal RF power 600 W). The vessel walls are controlled to a temperature below 40 °C via an active water cooling system, in order to assure reproducible conditions. A caesium dispenser oven is placed at the bottom plate in order to evaporate caesium inside the chamber [21]. The sample holder is located near the center of the experimental chamber and it is electrically and thermally insulated from the vessel. For the plasma phases the chamber is filled with H_2 gas at a pressure of 10 Pa.

The diagnostic systems adopted for the present investigations are shown in Fig. 1: the Cs evaporation rate is monitored via a surface ionization detector (SID) located directly upon the Cs oven nozzle; the impurity content is recorded by a residual gas analyzer (RGA); the negative hydrogen ion density is measured via a cavity ring-down spectroscopy (CRDS) system and the work function of the sample is evaluated considering the photoelectrical effect induced by a high pressure mercury lamp (100 W).

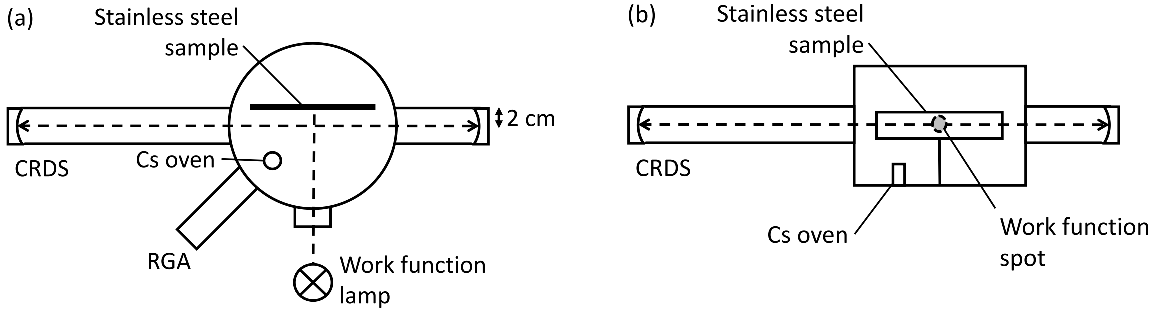

FIGURE 1. (a) Top view and (b) front view of the experimental setup, including the orientation of the diagnostics and the sample applied for the present campaigns.

The work function is evaluated by means of the Fowler method [22], following the enhanced procedure described in [23]. The photocurrent $I_{ph}(h\nu)$ depends on the energy $h\nu$ of the incident photons and on the work function χ of the converter surface. Hence, by using different interference filters to select the photon energy and measuring the corresponding photocurrents, it is possible to absolutely determine the work function. The sensitivity of this diagnostic system has been recently improved for the region below 3.0 eV by the installation of new interference filters with lower photon energy. The size of the spot on the sample has a diameter of 1.5 cm. The work function measurements are not possible in plasma phases, due to the massive presence of electrons which disturbs the photocurrent measurement. In [8], the evaluation of the work function is performed by pulsing the plasma with several hertz and measuring the photocurrents during plasma-off phases. This method cannot be applied at the current experimental setup due to a lower duty cycle, hence the work function is measured within the first minutes after plasma pulses of typically several minutes. The uncertainty associated to the work function measurements is of the order of 0.1 eV.

For the evaluation of the negative hydrogen ion density near the sample surface, a cavity ring-down spectroscopy system is installed [24–26]. The setup was previously installed in a different experiment, and its description can be found in [27]. The optical cavity is 1.5 m long and the laser beam is reflected back and forth by two planar-confocal highly reflective mirrors (curvature of 1 m and nominal reflectivity of 99.999 %). The line-of-sight of the laser beam is close to the converter surface, at 2 cm from the sample as shown in Fig. 1, in order to detect especially the negative hydrogen ions created by surface production. The obtained densities are absolute values but averaged over the plasma length (15 cm). In order to increase the ratio of surface to volume produced H^-, a stainless steel sample of 2.4 cm height and 11 cm length is installed. The error associated to the H^- density is 0.5×10^{15} m^{-3}. The mirrors conditions determine the detection limit for the H^- density evaluation: a modest loss of the reflectivity (−0.003 %) of the CRDS mirrors was observed after several months of operations, leading to a change of the detection limit from 3.5×10^{14} m^{-3} to 1.0×10^{15} m^{-3}. This event does not affect the measurements performed, but only the detection limit.

RESULTS

Investigations on H^- Volume Production

In order to study the influence of the plasma parameters on the negative hydrogen ion density, measurements of volume produced H^- density are performed. For this purpose, the sample holder was removed together with the Cs oven, and the newly installed CRDS was characterized in ACCesS for the first time. Figure 2 shows the behavior of the H^- density for different pressures and different RF powers. The electron density obtained by means of a Langmuir probe is also shown. The H^- density shows a maximum at 10 Pa of $(2.0\pm0.5)\times10^{15}$ m^{-3}, while at 20 Pa the density is below the detection limit (3.5×10^{14} m^{-3}). Furthermore, the H^- density increases with increasing power. The H^- to electron density ratio is always below 11 % and it decreases with increasing pressure. In particular, at 10 Pa and 250 W, the ratio is around 9 %.

In order to characterize the volume processes, the measured H^- density is investigated via a 0-dimensional model for the volume produced negative hydrogen ion density [27], where the relevant production and destruction reactions are balanced and the plasma parameters are taken as input. As a result, the H^- density depends mainly on the vibrational temperatures of the vibrationally excited hydrogen molecules in the electron ground state $H_2(X,v)$, where the vibrational population of $H_2(X,v)$ is given by a superposition of two Maxwell-Boltzmann distributions with two different vibrational temperatures $T_{vib,x}$ (with $x = 1, 2$) [27, 28]. While $T_{vib,1}$ (for $v < 4$) is evaluated via optical emission spectroscopy, $T_{vib,2}$ (for $v \geq 4$) is a free parameter obtained by adjusting the calculations to the experimental results. The production of negative ions via dissociative electron attachment to $H_2(X,v)$ increases strongly with increasing $T_{vib,2}$. On the other side, the main destruction mechanism observed in volume regime at ACCesS is the associative and non-associative detachment of negative hydrogen ions with atomic hydrogen. For pressures below 10 Pa, the high vibrational temperatures ($T_{vib,1} = 3500$ K and $T_{vib,2} > 6500$ K, with an error on the estimated temperatures of ±500 K) counterbalance the increase of the atomic hydrogen density with the pressure, while the decrease of the H^- density for pressures over 10 Pa is due to the lower vibrational temperatures of the $H_2(X,v)$ molecules ($T_{vib,1} = 3000$ K and $T_{vib,2} < 6500$ K).

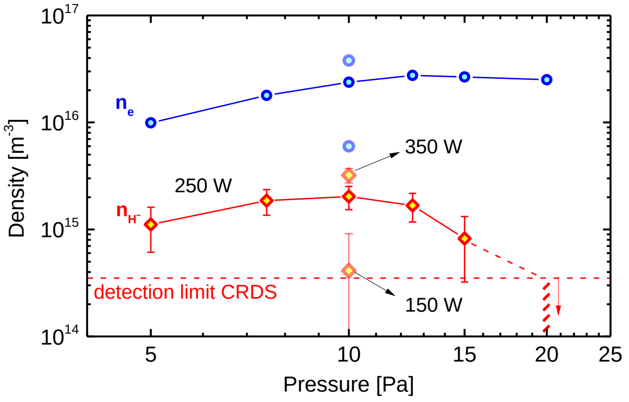

FIGURE 2. Volume produced H^- density and electron density for different pressures and RF power. The H^- density at 20 Pa is below the detection limit (3.5×10^{14} m^{-3}).

Influence of H_2 Plasma on a Caesiated Surface

In order to investigate the effect of plasma exposure on a caesiated surface, caesiation is performed in vacuum with a background pressure of the order of 10^{-6} mbar. The Cs evaporation rate is monitored by the oven SID. Figure 3 shows the gradual decrease of the work function of the stainless steel sample, with the increasing SID signal, hence with increasing Cs flux. Before caesiation the work function is above 4.3 eV, which is the upper limit for the work function evaluation with the current setup. This is consistent with the iron work function (4.5 eV) [6]. At the end of the caesiation process in vacuum the work function reaches the stable value of $\chi_{vacuum} = (2.7 \pm 0.1)$ eV. This result is in accordance with previous measurements on a different sample [7]. From [7] it is known that χ_{vacuum} represents the minimum of the work function in ion source relevant vacuum conditions: a further increase of the Cs evaporation rate does not affect the surface work function, due to the presence of Cs compounds with residual gases on the surface. The non-metallic nature of these compounds leads to a higher work function compared with bulk Cs.

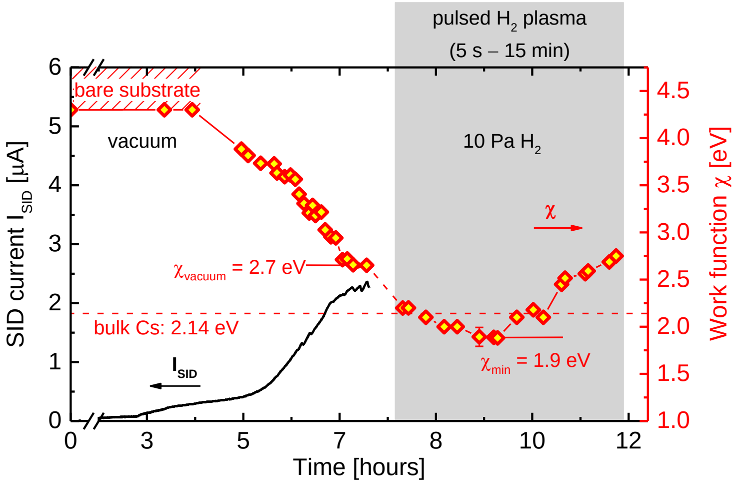

FIGURE 3. Work function of a stainless steel sample during caesiation in vacuum (background pressure 10^{-6} mbar) and after pulsed hydrogen plasma treatment (plasma-on time repeatedly between 5 s and 15 min). The current signal collected on the SID is an index of the Cs evaporation rate.

After caesiation, the evaporation of Cs from the oven is stopped, as well as the SID current measurement, and the chamber is filled with H_2 gas at a pressure of 10 Pa. The impurity content is constantly monitored by the residual gas analyzer and no particular events are observed during the operations. Plasma pulses with an RF power of 250 W are then performed. The plasma-on time was stepwise increased from 5 s to 15 min during this phase, only interrupted to measure the work function. The interval between one pulse and the other is usually of 5−10 minutes. The sample potential is floating during the plasma phases. Already after the first 5 s plasma pulse, the work function reaches the value for the bulk Cs, indicating a cleaning effect of the Cs layer. Due to additional plasma exposure and the interaction of the plasma particles, like neutral hydrogen atoms and positive hydrogen ions, probably together with the UV light emitted by de-excitation of neutral hydrogen, the Cs is then released from the surface. As a result, the work function reaches a minimum $\chi_{min} = 1.9$ eV after an overall plasma-on time of 15 min. The minimum work function after plasma treatment thus is 0.8 eV lower than χ_{vacuum}. It is important to remind that the Cs evaporation was stopped at the end of the vacuum phase, hence the Cs layer cannot be restored but it is gradually removed by the impinging plasma particles. The work function of 1.9 eV is below the work function of bulk caesium, indicating a Cs coverage below 1 monolayer on the sample surface. The work function of 1.9 eV has been observed in several, but not all the campaigns at the same experimental conditions: in few campaigns the work function decreased just down to 2.1 eV. Hence this aspect is still under investigation.

After reaching χ_{min}, the work function starts to increase again due to the interaction between the plasma particles and the surface. After an overall plasma-on time of 1.5 hours, the surface work function is 2.8 eV, already higher than the vacuum minimum value.

Influence of the Work Function on the H^- Density

In order to measure the H^- density for different work functions, a pulsed plasma campaign is performed with a caesiated surface: a longer plasma exposure compared to the previous campaign is applied, in order to increase further the work function by interaction of plasma particles on the Cs layer. It should be noted that the presence of this long sample inside the vessel can affect the plasma parameters compared to the H^- volume production investigations [29], hence the H^- density measurements previously performed in volume regime are no more valid for the current setup. The purpose of the current campaign then is to investigate the influence of the work function on the H^- density, both absolutely measured.

Figure 4 shows the work function of a caesiated sample. The bare substrate is a new stainless steel sample, treated with the same procedure shown in the previous section. After the caesiation process in vacuum, the work function is 2.7 eV before applying any plasma. The evaporation of caesium is stopped before injection of H_2 gas. By the interaction with the H_2 plasma at 10 Pa, the work function decreases down to (2.1±0.1) eV after some minutes of plasma treatment.

The H^- density measured via CRDS for the first plasma pulses is stable at $(2.1\pm0.5)\times10^{15}$ m^{-3}. As already mentioned, this value cannot be compared to the one measured during volume operations at 10 Pa.

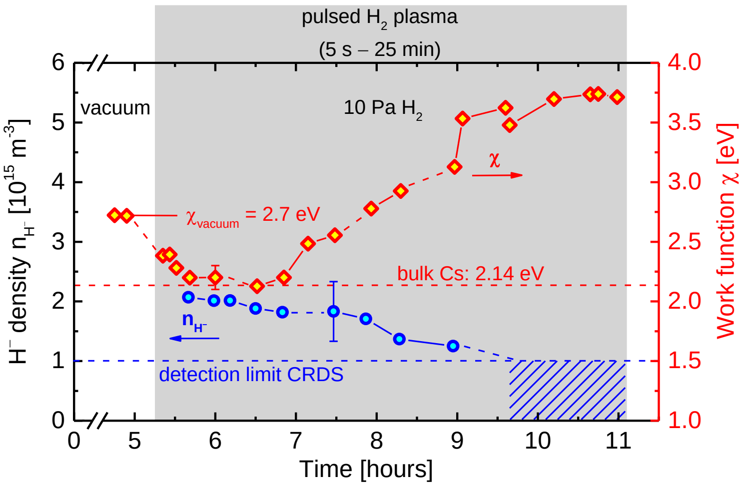

FIGURE 4. Work function of a caesiated stainless steel sample under plasma treatment (plasma-on time between 5 s and 25 min). The H^- density measured during the plasma phase is also plotted. The H^- density is below the detection limit (1.0×10^{15} m^{-3}) in the last part of the campaign, where a higher work function is achieved.

By subsequent plasma pulses with different plasma-on time, the caesium deposited on the sample surface is progressively removed by the plasma particles. As a result, the work function increases gradually up to a value of 3.75 eV after an overall plasma-on time of 3 hours, and the H^- density decreases down to values below the detection limit. The detection limit is 1.0×10^{15} m^{-3}, higher than during the volume investigations due to the degradation of the CRDS mirrors after several months of operations.

The volume produced H^- density represents an offset for the detection of surface produced H^-. For these measurements, this offset is given by the H^- density measured for high work function values, hence it is below the detection limit.

The H^- density against the work function is plotted in Fig. 5. The work function increases from 2.1 eV to 3.75 eV while the H^- density decreases by at least a factor of 2, showing that the surface production of negative hydrogen ions directly depends on the actual work function of the surface. For low work functions a higher H^- density is observed.

In order to discuss the obtained results, it is worth comparing the measured H^- densities to the one achieved at the BATMAN testbed, dedicated to development of H^- production and equipped with the RF-driven prototype source for the fusion experiment ITER [30]. Starting from a non-caesiated source, the caesiation process produces an increase of the H^- density by more than a factor of 10 during the first day of caesiation, as shown in [31] (RF power 60 kW, pressure 0.3 Pa, extraction voltage 5 kV). Since in ACCesS (RF power 250 W, pressure 10 Pa) at least a factor of 2 is observed, first considerations can be made on the possible influences of the plasma on the H^- density.

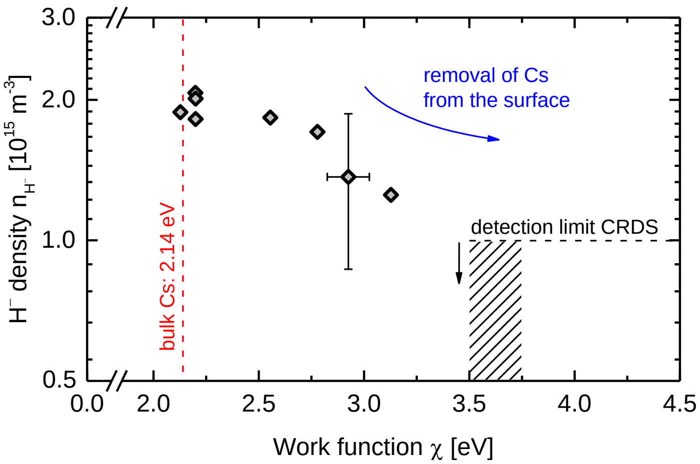

FIGURE 5. H^- density with increasing surface work function during the plasma treatment. For work functions above 3.5 eV the H^- density is below the detection limit (1.0×10^{15} m^{-3}).

As already mentioned, the surface production of negative hydrogen ions relies on the conversion of neutral hydrogen atoms and positive hydrogen ions approaching the low work function surface. The flux of produced negative ions for each of the two channels depends on the flux of the impinging particles (H or H_x^+) onto the surface multiplied by the corresponding conversion yield Y. For prototype sources, the conversion of neutral hydrogen atoms is the dominant surface production mechanism, due to the higher atomic flux compared with the positive ion flux and to an adequate energy, as shown in [32]. Also in ACCesS, the atomic hydrogen flux is at least two orders of magnitude higher than the positive ion flux, due to the high atomic hydrogen density (of the order of 10^{19} m^{-3}). The temperature T(H) of the atomic hydrogen particles is around 0.05 eV and it is obtained by considering the Doppler effect on the broadening of the hydrogen Balmer lines in the visible range via optical emission spectroscopy during several experimental campaigns in ACCesS. The resulting atomic hydrogen flux has been calculated in ACCesS considering an isotropic thermal distribution of the neutral atoms with temperature T(H) and it is in the range of $(2–4)\times10^{22}$ m^{-2} s^{-1}, which is comparable to the values calculated for BATMAN source in [33], that is $(1–3)\times10^{22}$ m^{-2} s^{-1}.

On the other hand, the conversion yield factors depend on the work function, as experiments performed in vacuum have shown [34, 35], and on the energy of impinging particles [34–37]. In particular, considering a constant work function, experiments performed in vacuum with caesiated surfaces show that the conversion yield for atomic hydrogen atoms increases with increasing T(H) [37]. As already mentioned, the atomic hydrogen temperature in ACCesS is around 0.05 eV, while in BATMAN ion source it is 0.8 eV [32], indicating a higher conversion yield in BATMAN source compared with ACCesS if the same low work function is assumed. However, it should be considered that the plasma environment might have effects on the resulting conversion yields as well.

In summary, absolute measurements of work function and H^- density present a clear trend: first results at ACCesS experiment have shown that the H^- density increases at least by a factor of 2 with decreasing work function from 3.7 eV to 2.1 eV. Systematic investigations of the correlation between negative hydrogen ion density and surface work function will be performed, and improvements to the experimental setup will be applied. The attachment of a tunable diode laser absorption spectroscopy is planned in the early future, in order to measure the Cs density in vacuum and plasma conditions. The sensitivity range for the CRDS will be enhanced with the application of new reflective mirrors, reducing the detection limit for the H^- density. The improved experimental setup will allow to investigate the plasma influence on the work function and on the H^- density in different experimental conditions, e.g. with different substrates materials, applying additionally a bias. It will be investigated the case of caesiation during plasma operations, with different pressures and RF powers, and possibly with different amounts of impurities in the H_2 gas.

CONCLUSIONS

The formation of negative hydrogen ions in negative hydrogen ions sources based on surface conversion is strictly depending on the work function of the converter surface and on the energy of impinging particles on the surface. In view of ion sources for fusion devices like ITER and DEMO, investigations on the correlation between work function and H^- density are highly desirable. Low work functions can be achieved by caesiation of the surface. Since absolute values of work function are not easily accessible at the ion sources, simultaneous and absolute measurements of H^- density and work function are performed at a flexible laboratory experiment, where a cavity ring-down spectroscopy system for H^- detection has been recently installed and the work function is measured by the photoelectrical effect.

The performed investigations show that caesiation of a stainless steel sample leads to a minimum work function of 2.7 eV in vacuum (background pressure of 10^{-6} mbar). Without additional Cs evaporation in the experiment during plasma phase (pressure 10 Pa), the effect of plasma interactions with the caesiated surface allows to reach a work function of 1.9–2.1 eV, with an H^- density of 2.1×10^{15} m^{-3}. The impinging plasma particles, like neutral hydrogen atoms and positive hydrogen ions, probably together with the UV light emitted by de-excitation of neutral hydrogen, gradually remove caesium from the surface. Consequently, the work function increases, giving the opportunity to measure the H^- density for different work functions. First results show that the work function increases from the minimum value of 2.1 eV to 3.75 eV and the H^- density decreases consequently by at least a factor of 2, down to values below the detection limit of the current apparatus. It is then proved that a change in the actual work function has direct effects on the surface production of negative hydrogen ions and an experimental correlation between H^- density and work function can be achieved at the ion source relevant plasma conditions.

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