

Efficiency of Cs-free Materials for Negative Ion Production in H₂ and D₂ Plasmas

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Abstract. High power negative ion sources use caesium to reduce the work function of the converter surface which significantly increases the negative ion yield. Caesium, however, is a very reactive alkali-metal and shows complex redistribution dynamics in consequence of plasma-surface-interaction. Thus, maintaining a stable and homogenous low work function surface is a demanding task, which is not easily compatible with the RAMI issues (reliability, availability, maintainability, inspectability) for a future DEMO fusion reactor. Hence, Cs-free alternative materials for efficient negative ion formation are desirable. At the laboratory experiment HOMER materials which are referred to as promising are investigated under identical and ion source relevant parameters: the refractory metals Ta and W, non-doped and boron-doped diamond as well as materials with inherent low work function (lanthanum-doped molybdenum, MoLa and lanthanum hexaboride, LaB₆). The results are compared to the effect of in-situ caesiation, which at HOMER leads to a maximal increase of the negative ion density by a factor of 2.5. Among the examined samples low work function materials are most efficient. In particular, MoLa leads to an increase of almost 50 % compared to pure volume formation. The difference to a caesiated surface can be attributed to the still higher work function of MoLa, which is expected to be slightly below 3 eV. Using deuterium instead of hydrogen leads to increased atomic and positive ion densities, while comparable negative ion densities are achieved. In contrast to the low work function materials, bulk samples of the refractory metals as well as carbon based materials have no enhancing effect on H⁻, where the latter materials furthermore show severe erosion due to the hydrogen plasma.

INTRODUCTION

Neutral beam heating (NBI) systems for future fusion devices like ITER and beyond require high-performance ion sources for negative hydrogen ions. In order to achieve the required heating powers of several tens of MW the H⁻/D⁻ source has to deliver stable and spatially homogeneous beams (cross section ~m²) with current densities of 200–300 A/m² for pulse lengths of one hour (or longer beyond ITER). Furthermore, the low temperature hydrogen discharge of the ion source needs to be operated at pressures of 0.3 Pa or below in order to reduce stripping losses within the accelerator stages. Such high current densities at low pressure can only be achieved by ion sources based on the surface conversion mechanism for negative ion formation: the so-called converter surface is covered with the alkali metal caesium to reduce its work function which significantly enhances the conversion yield of atomic and ionic hydrogen particles to negative ions [1–3]. Caesium, however, is a very reactive alkali metal and has a very high vapor pressure. This leads to complex physical and chemical ad-, desorption and degradation processes during vacuum phases of ion source operation as well as to cleaning, sputtering and redistribution processes during plasma phases. Since the work function of Cs layers significantly depends on the thickness and the purity of the Cs coating, this dynamical behavior has a direct impact on the extracted currents [4]. Especially in view of the required stability and homogeneity of the negative ion beam and its reliability and availability, this behavior poses several difficulties. Hence, alternative materials to Cs covered surfaces for efficient negative ion production are a key interest in current ion source research and the investigations are included in the EUROfusion work package for heating and current drive (WPHCD) of future fusion devices like DEMO.

Several groups of materials are suggested throughout the literature to produce negative hydrogen ions via different mechanisms [5]:

- Like Cs (bulk work function 2.14 eV) also other materials with a low work function are expected to directly produce negative hydrogen ions at their surface by transferring one or more electrons from the solid state to impinging atoms or ions. First investigations in this direction were already reported by [6] showing the principle applicability of a lanthanum hexaboride cathode for H^- extraction.
- For the refractory metals tantalum and tungsten several experiments showed an enhancing effect on the H^- yield (see e. g. [7, 8]). The effect is often explained by an increasing population of highly vibrationally excited H_2 molecules which thus indirectly enhances the H^- volume production via the dissociative attachment process: $H_2(v \geq 5) + e \rightarrow H + H^-$. On the other hand, also diminishing effects on negative ion yields using these metals as bulk material are reported in [9] for instance. Hence, results on refractory metals seem to be dependent on the respective plasma conditions and furthermore on the surface structure of the material, since most of the experiments showing enhanced H^- yields rely on evaporated materials.
- Carbon materials, in particular non-doped and boron-doped diamond (BDD) have also shown to produce negative hydrogen ions (see e. g. [10, 11]). However, in contrast to low work function materials, the formation mechanism at the insulating diamond surface (work function above 4 eV) and thus the actually required conditions for efficient H^- production are not fully understood at present. The capability of diamond layers to show negative electron affinity under certain circumstances is often discussed in this context, since this characteristic helps to emit electrons from a solid state [12].

Until now investigations on these materials were performed in dedicated experiments aiming at specific characteristics of the corresponding mechanism. Hence, the results might not directly compare to each other and furthermore might not be transferable to ion sources, since absolute yields under ion source relevant plasma conditions are missing.

Thus, dedicated comparative studies on these material groups regarding the efficiency for negative ion formation are performed under controlled and identical plasma conditions at the laboratory experiment HOMER [13–15] including comparison to the current state-of-the-art, i. e. a caesiated surface, as well as to pure volume formation. The setup follows the tandem concept by which plasma parameters comparable to the ones close to the plasma grid in negative ion sources are obtained for pressures as low as 0.3 Pa. The negative ion density is measured via laser photodetachment, while a Langmuir probe and optical emission spectroscopy (OES) are applied to characterize and monitor the plasma. Besides measurements also modeling of the H^- volume production is performed [16] in order to deduce the influence of different material surfaces on the plasma itself. Like this, possible H^- surface effects can be distinguished from the influence on the background plasma.

EXPERIMENTAL SETUP

HOMER [13] is an ECR (Electron Cyclotron Resonance) discharge driven by a microwave generator at 2.45 GHz with powers up to 1 kW. A schematic of the experimental setup is shown in figure 1. The magnetic field of 87.5 mT is generated by Helmholtz coils. The cylindrical stainless steel vacuum vessel has a diameter of 15 cm and a height of 31 cm in total. In the region of the upper magnetic coil a meshed grid is installed which separates the volume into a heated driver region above the grid (height 12 cm) and a diffusive downstream region. The latter part houses the sample holder and gives diagnostic access to the plasma above the sample surface. This tandem concept provides plasma parameters comparable to the conditions close to the converter surface in negative ion sources for fusion: discharge pressures down to 0.3 Pa, electron temperature 1–2 eV, electron and positive ion density 10^{16} – 10^{17} m^{-3} and atomic density 10^{18} – 10^{19} m^{-3} .

The sample holder is insulated from the vessel walls and can hold material samples up to sizes of $8.5 \times 6 \text{ cm}^2$. It can be moved in vertical direction by $\pm 2 \text{ cm}$, biased against the vessel walls up to $\pm 40 \text{ V}$ and heated up to temperatures of 550°C . Negative ion densities n_{H^-} are measured locally above the sample surface via laser photodetachment: a short laser pulse (1064 nm, 8 ns, 23 mJ) destroys the negative hydrogen ions within the laser channel and the detached electrons are collected by the Langmuir probe which is biased to voltages within the electron saturation region. A fast trigger circuit is used to synchronize the laser pulse and the detection of the electron current. The measured relative negative ion density n_{H^-}/n_e is translated to absolute values via the ion density from Langmuir probe measurements. The absolute density n_{H^-} has been benchmarked against cavity ring-down spectroscopy [16]. Furthermore, the Langmuir probe is used to determine the ion density, the electron energy distribution function and the plasma and floating potential. Due to the magnetic field required for ECR coupling a direct determination of the electron density via the

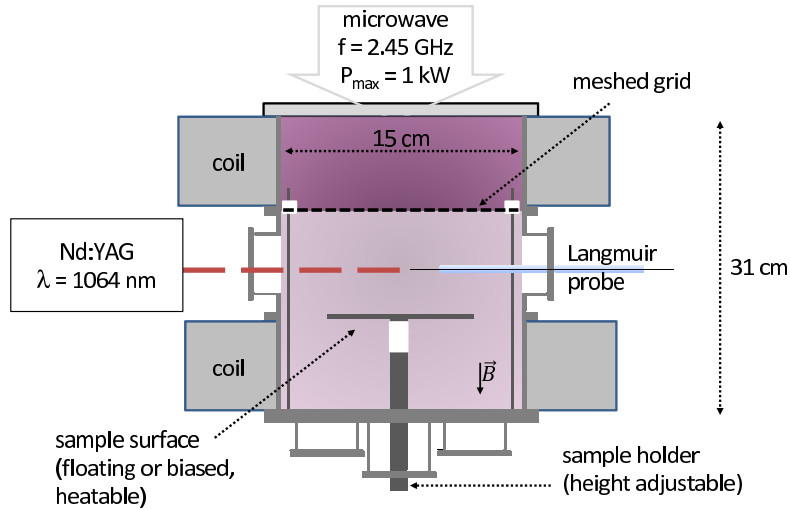


FIGURE 1. Schematic of the ECR tandem experiment HOMER illustrating the separation into driver and downstream region by the meshed grid and the measurement position relative to the material samples.

Langmuir probe is not possible. Thus, quasi-neutrality is used to determine n_e via the ion density. Optical emission spectroscopy (OES) is applied for the determination of the atom-to-molecule density ratio n_H/n_{H_2} , the gas temperature and the vibrational temperature of the molecular ground state. Further information about the diagnostic systems can be found in [13].

RESULTS

In order to assess different Cs-free materials regarding their capability for negative ion production, their influence on the negative ion density needs to be compared to pure volume formation as a lower limit and to the presently known most efficient formation scheme via a caesiated surface.

For the former case a dummy surface needs to be installed in order to maintain the plasma geometry compared to the following investigations with different material samples. Here, a stainless steel (V2A) sample can be applied, since for stainless steel no material induced influence on the negative ion density is known. Thus, results obtained with V2A can act as a reference describing the offset of pure H^- volume formation at the particular plasma conditions during the performed measurement series. Any candidate for a Cs-free converter material must then at least overcome the limit given by the V2A reference in order to result in a detectable material induced enhancement of the negative ion density.

Furthermore, the negative ion yield needs to be compared to the effect of Cs. Here it is known e. g. from the ITER prototype source that caesiation can lead to an increase of the negative ion yield by a factor of ten [17]. This increase, however, depends on the present plasma parameters, i. e. the flux of impinging particles and their energy. Hence, the actual influence of a caesiated surface is examined by caesiating a V2A sample in-situ.

Analysis of H^- volume processes at HOMER – Experiment and Modeling

The dependency of the negative hydrogen ion density above the stainless steel dummy surface (45 mm distance) on the pressure is shown in figure 2 (a), while the corresponding electron density as well as a vibrational temperature determined by modeling (see below) is shown in part (b). For a pressure of around 0.5 Pa a maximum in n_{H^-} develops reaching about $2.8 \times 10^{15} \text{ m}^{-3}$ which corresponds to an H^-/e ratio of about 13 %. For lower and higher pressures the density decreases rapidly to below 10^{15} m^{-3} in minimum at pressures above 2 Pa. The electron density, however, is peaked at 1 Pa with a maximum of about $3.2 \times 10^{16} \text{ m}^{-3}$.

In order to explain the observed pressure dependence and furthermore to determine the dominant destruction processes for negative ions within the plasma volume 0-dimensional modeling of the H^- density is performed using

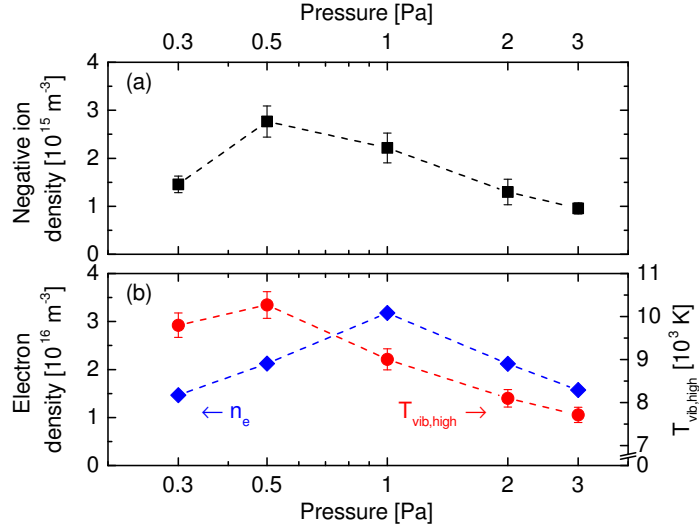


FIGURE 2. (a) H^- density and (b) electron density and vibrational temperature for the highly excited vibrational states of H_2 above the stainless steel reference sample for 300 W microwave power in a distance of 45 mm to the surface.

the solver YACORA [16, 18]. H^- production via dissociative attachment (DA) is balanced by the well-known destruction mechanisms: electron detachment (ED), mutual neutralization (MN) with positive hydrogen ions (H , H_2^+ , H_3^+), associative and non-associative detachment (AD, nAD) by collisions with hydrogen atoms and collisional detachment (CD) by hydrogen molecules. Cross section data are taken from [19]. Measured plasma parameters from OES and Langmuir probe are used as input parameters. The tandem concept is taken into account by assuming an ensemble of hydrogen molecules which are vibrationally populated in the heated driver region and diffuse to the downstream region above the sample surface. This ensemble can be characterized by a vibrational temperature $T_{\text{vib,high}}$. Like this, a two-temperature vibrational population distribution for the H_2 ground state is produced above the sample surface, characterized by two temperatures $T_{\text{vib,low}}$ and $T_{\text{vib,high}}$ for the low-energy and high-energy range, respectively. Here, $T_{\text{vib,low}}$ is diagnostically accessible via OES [20], while $T_{\text{vib,high}}$ is not accessible and is used as a free parameter in the modeling procedure. Further input parameters like electron density and temperature are taken from measurements directly in the downstream region. Modeled H^- densities are then fitted to measured ones by variation of $T_{\text{vib,high}}$ [16].

In figure 2 (b) the pressure dependent vibrational temperatures $T_{\text{vib,high}}$ required to fit the measurements are shown additionally to n_e . Agreement of model and measurement is thus obtained for values of $T_{\text{vib,high}}$ in the range of 8,000–10,000 K decreasing with pressure, while the measured $T_{\text{vib,low}}$ is almost constant around 3,500 K.

For these modeled densities the relative share of the destruction processes to the total H^- destruction rate is shown in figure 3. Over the whole pressure range associative and non-associative detachment of negative ions by hydrogen atoms is the dominant destruction channel while its share increases with pressure. For pressures below 0.5 Pa mutual neutralization as well as electron detachment contribute with around 10 % in maximum. Collisional detachment by H_2 is negligible at these plasma conditions.

Thus, the negative hydrogen ion density produced in the volume is sensitive to the atomic hydrogen density dominantly determining the destruction rate on the one hand, and to the electron density and the vibrational population determining the production rate on the other hand. The presented H^- pressure dependence (figure 2), however, is mainly linked to the H^- production side only: the behavior of n_e (maximum at 1 Pa) together with the decreasing evolution of $T_{\text{vib,high}}$ determines the observed maximum in n_{H^-} around 0.5 Pa. For more details on this analysis see [16].

Effect of caesiation at HOMER

In order to determine the H^- level with which Cs-free materials have to compete a stainless steel sample is caesiated in-situ. A caesium dispenser (Bi_2Cs alloy from Alvatec [21]) is installed above the sample surface such that evaporation of Cs directly covers the stainless steel surface while contamination of the surrounding discharge vessel is mostly prevented. Nevertheless, after this campaign the experimental vessel was completely cleaned from any possible Cs

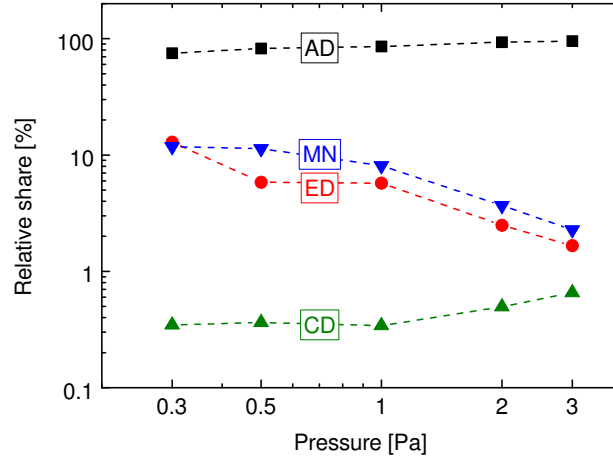


FIGURE 3. Relative share of the H^- destruction channels to the total destruction rate determined via YACORA H^- for the measurements with V2A. 'AD' stands for the combined rate of associative and non-associative detachment.

residues thus assuring a Cs-free environment for the following investigations. Figure 4 shows the evolution of the Cs emission line at 852 nm, the negative ion density as well as the positive ion density during onset of Cs evaporation.

Cs evaporation is monitored at the emission from the caesium atom. It can be seen that the negative ion density directly reacts on the coating of the surface while the positive ion density as well as other plasma parameters like electron temperature stay constant, which is in line with the investigations by [22]. Thus, the volume production of H^- should not be influenced by the presence of Cs in the plasma and the additionally detected negative ions can be attributed to surface conversion at the caesiated V2A surface. Measurements of the work function under comparable plasma parameters at the laboratory experiment ACCesS have shown values down to 2.1 eV for caesiated surfaces [23–25]. Hence, it can be concluded that in this system formation of H^- at a surface with 2.1 eV work function leads to an increase of the negative ion density by about a factor of 2.5 in maximum. This is the goal for any candidate as Cs-free converter material.

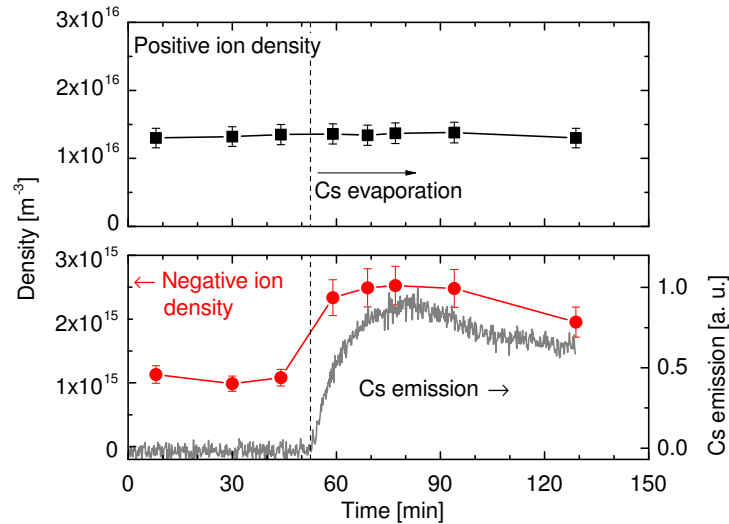


FIGURE 4. Monitoring the caesiation of a stainless steel sample for 300 W microwave power and 0.3 Pa H_2 pressure in a distance of 25 mm to the surface.

Cs-free materials for H^- production

The following materials were investigated concerning their efficiency for negative hydrogen ion formation: bulk samples of the refractory metals Ta and W, non-doped and boron-doped diamond (BDD) and materials with inherent low work function, namely lanthanum-doped molybdenum (99.3 % Mo : 0.7 % La, MoLa) and lanthanum hexaboride (LaB_6). Comprehensive measurement series were performed regarding the variation of the hydrogen pressure, the distance to the sample surface, the bias potential applied to the surface as well as the sample temperature [14, 15]. Figure 5 summarizes the results at one specific but representative point within the investigated parameter space: the maximally achieved H^-/e ratios at 0.3 Pa in a distance of 25 mm are shown for the different materials compared to the reference case of a stainless steel surface (V2A) and to the case of the in-situ caesiated surface (V2A + Cs).

The employed bulk samples of the refractory metals Ta & W do not show an enhanced negative ion yield compared to V2A. Furthermore, comparable vibrational temperatures have been measured via OES for the different materials. Hence, these bulk materials do not show a significant impact on the H_2 vibrational population (or any other plasma parameter) and consequently also no effect on the H^- volume production is expected. It has to be noted, that the enhancing effect of these refractory metals cited in the literature was mostly achieved by evaporation of the metals. Since adsorbed layers of evaporated materials have a much higher surface roughness than bulk materials their impact on the H_2 vibrational population might be much higher. However, it is disputable if replacing Cs in future NBI sources by another material which needs to be evaporated is advisable, since issues correlated with plasma induced redistribution or penetration into the accelerator stages would still be present.

Among the carbon based materials, two different non-doped and three different boron-doped diamond samples were investigated differing in the substrate material (silicon, molybdenum, niobium), the thickness of the diamond layer (3–750 μm), the average grain size (1.8–150 μm) as well as the crystallinity (epitaxial, polycrystalline). However, none of them resulted in H^- densities exceeding the volume production. Moreover, all of the carbon samples have shown severe erosion due to the high flux hydrogen plasma (plasma-on-time ~ 10 h). Weight loss rates in the range of mg/h were detected and confirmed by surface analyses with SEM and Raman spectroscopy. Thus, these materials can not be seen as suitable alternatives to Cs for future negative ion sources. The results on diamond materials are presented in more detail including parameter studies in [15].

In contrast, lanthanum-doped molybdenum as well as lanthanum hexaboride, both of which are expected to have a work function of slightly below 3 eV, show an increase of the negative ion yield of almost 50 % compared to stainless steel. Furthermore, no signs of plasma induced erosion could be detected for these bulk materials. Thus, basic requirements for being an alternative to caesiated surfaces in negative ion sources are fulfilled. However, the impact on the negative ion density is still much smaller than via the application of Cs which can be attributed to the higher work function. Measurements of the actual work function under ion source conditions aiming at the identification of possibilities for tuning are scheduled at ACCesS [25–27].

Concluding, direct surface conversion at low work function surfaces seems to be the most promising way for efficient H^- production under ion source relevant conditions. Among those, caesiated surfaces still stand alone due to their so far unmatched low work function.

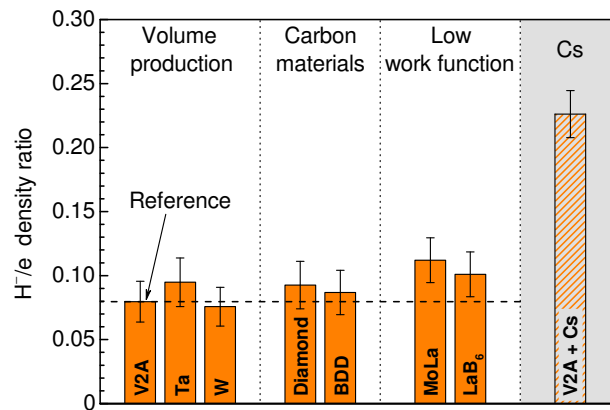


FIGURE 5. Negative-ion-to-electron ratio for several materials at 300 W microwave power and 0.3 Pa H_2 pressure in a distance of 25 mm to the surface.

Isotopic differences

Ion sources for fusion applications have to be driven in both H_2 and D_2 and the respective performance varies depending on the isotope [28]. Thus, for the most effective Cs-free converter material –MoLa– investigations comparing hydrogen and deuterium are performed. Exemplarily the measurement campaign varying the bias voltage applied to the sample surface with respect to the grounded vessel walls is considered. Varying the bias leads to a variation of the energy of the impinging particles as well as of the emitting negative ions which can have an influence on the surface conversion probability. The performed bias scan ranges from -30 to $+20$ V, where the plasma potential is always higher leading to a variation of the ion energy from 20 eV at -30 V to slightly below 1 eV at $+20$ V (assuming a collisionless sheath). Figure 6 shows the corresponding influence on the negative ion density above the MoLa sample in H_2 and D_2 compared to the case with stainless steel in H_2 .

Besides the ion energy, biasing the sample surface has a significant influence on the plasma parameters of the bulk plasma, since the spatial potential distribution is varied. This affects the H^- related volume processes which can be seen at the V2A measurements. It is shown by modeling [14] that the variation of the volume H^- density with bias is mainly determined by a varying electron energy distribution function (EEDF) showing a decreasing mean electron energy with increasing bias. This leads to steadily increasing rate coefficients for H^- production via dissociative attachment, since for this process lower electron energies are beneficial. The enhancing effect is, however, slightly compensated by a correspondingly decreasing vibrational population of H_2 . The combined effects lead to a slightly increasing H^- density with increasing bias.

Changing the sample material to MoLa has virtually no influence on the evolution of the EEDF or the vibrational population of H_2 . Furthermore, almost identical atomic-to-molecular density ratios are measured. Thus, the volume formation of H^- should not be influenced by the low work function material. However, it can clearly be seen that for voltages above -20 V the negative ion density is higher for MoLa than for V2A reaching the maximum enhancement with almost $+60\%$ at around -5 V. This enhancement indicates an additional source for negative ions besides volume formation and can clearly be attributed to surface production. Figure 7 shows on the right side to the bottom the atomic-to-molecular density ratio for MoLa in H_2 where an increasing atomic density can be observed. This is accompanied with an increasing flux of atomic hydrogen onto the low work function surface showing the steepest increase between -20 and -10 V. This again is directly linked to an increasing amount of surface produced negative ions with increasing bias. Additionally, the energy of negative ions leaving the surface is decreasing with increasing bias which leads to decreasing destruction rates. Hence it can be concluded that for voltages above -20 V the influence of surface produced negative ions is sufficiently high to overcome the respective volume density.

Changing from H_2 to D_2 has a significant influence on the plasma parameters, as can be seen in figure 7: Both the positive ion density and the atomic density are increased in D_2 . While the increased positive ion density, which is directly linked to a higher electron density, is regularly observed at HOMER and can be explained by a better

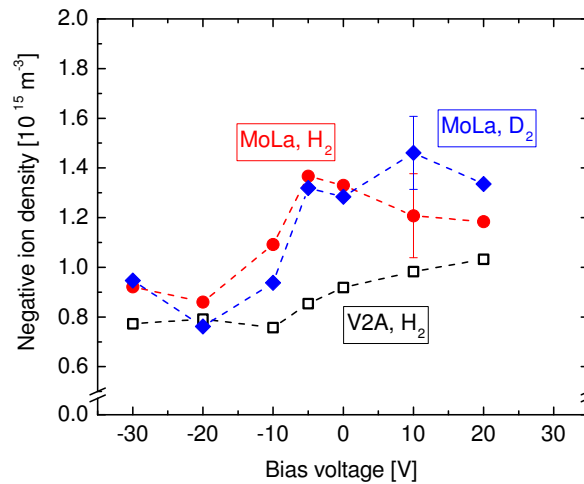


FIGURE 6. H^- density above lanthanum-doped molybdenum for 0.3 Pa in H_2 and in D_2 at 300 W microwave power in a distance of 25 mm to the sample surface. For comparison measurements with stainless steel in H_2 are depicted as well.

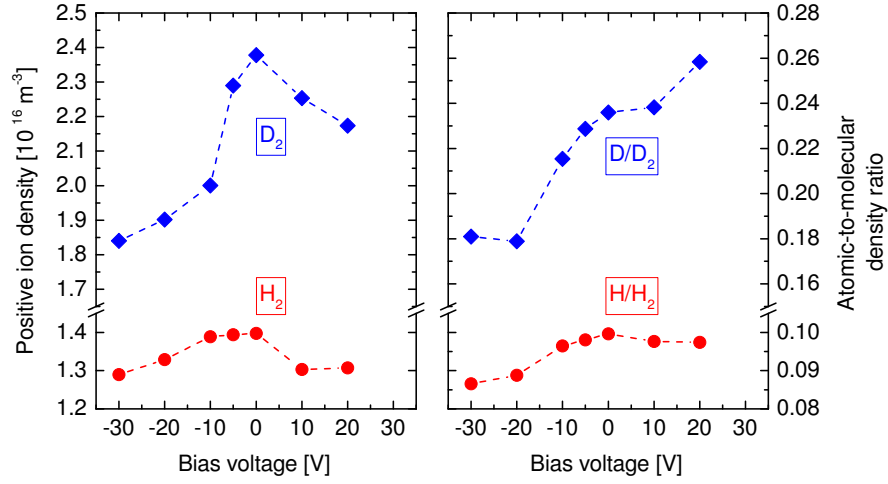


FIGURE 7. Positive ion density and atomic-to-molecular density ratio above MoLa in 0.3 Pa H_2 and D_2 at 300 W microwave power in a distance of 25 mm to the sample surface.

coupling of the microwave to the deuterium plasma [29], the increased atomic density is attributed to the higher dissociation cross section for D_2 [30] and is thus frequently observed in low temperature hydrogen discharges (see e. g. [22]). Other plasma parameters like gas temperature, the EEDF or the vibrational distribution of the molecular ground state show only very slight differences between the isotopes. Regarding negative ion formation, the higher electron density in D_2 indicates an enhanced negative ion volume production compared to H_2 . On the other hand, the higher atomic density in deuterium results in a comparably enhanced negative ion destruction within the volume due to the associative detachment process but also to an increased atomic flux onto the surface. Due to the only limited contribution of surface production in this case, the different effects seem to compensate each other leading to virtually identical negative ion densities in D_2 and H_2 as shown in figure 6.

Nevertheless, the difference between n_{D} and n_{H} increases with increasing bias leading to an increasing influence of surface produced negative ions with increasing bias. Hence, at least in the positive bias range an enhanced negative ion density for D_2 could be expected. However, within the measuring accuracy, this enhancement could not be detected. In caesiated high power negative ion sources, however, surface conversion is the dominant H^- formation mechanism. And indeed, in this case, an isotopic influence can be observed, as for example shown at the BATMAN test facility [28]: the typically increased dissociation degree in deuterium plasmas directly converts to an increased negative ion density.

CONCLUSION

Alternative materials to Cs for H^- production have been investigated under identical conditions with plasma parameters close to the ones at ion sources for fusion. The results are assessed compared to pure H^- volume formation and to the current state-of-the-art, i. e. an in-situ caesiated surface.

It is shown, that neither bulk samples of the refractory metals Ta or W nor any of the examined diamond samples have the potential to exceed negative ion volume formation for these plasma parameters. Furthermore, for the carbon based materials severe plasma induced erosion was observed already for plasma-on-times below 10 h. Thus, these material groups reveal to be unsuitable as possible alternative materials to caesium for future negative ion based NBI systems, since they neither fulfill the requirements on efficiency nor on stability of H^- production.

On the contrary, the tested materials with low surface work function, i. e. lanthanum-doped molybdenum and lanthanum hexaboride, have proven long term stability to the high flux hydrogen plasma and, more importantly, they clearly enhance the negative ion density via surface conversion: MoLa shows an increase of almost +50 % compared to a bare stainless steel surface. However, this effect is still far below that of caesiation (n_{H^-} increases by about a factor of 2.5). This difference can be attributed to the higher surface work function of slightly below 3 eV for MoLa in contrast to 2.1 eV for caesiated surfaces. Varying the isotope from hydrogen to deuterium shows an influence on the plasma parameters, while the resulting effects on the negative ion formation compensate each other leading to

comparable negative ion densities independently from the isotope.

Regarding application to ion sources, the bulk materials of MoLa and LaB₆ should moreover imply a substantially reduced complexity of handling compared to a volatile coating with a highly reactive alkali metal. Furthermore, a substantially reduced migration of material into the extraction system can be expected compared to a material which is necessarily evaporated into the source. However, possibilities have to be identified in order to further decrease the work function since a lower negative ion density at the converter surface also yields a reduced effect on the electron density in front of the converter surface and thus, the co-extracted electron current might be much less reduced.

Hence, further studies on efficient Cs-free materials for negative ion formation should be focused on materials with inherent low work function aiming at even lower but still stable surface work functions. Here, measurements of the actual work function under ion source conditions and determination of the parameters influencing the work function are mandatory.

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