Cs-Doped Mo as Surface Converter for H⁻/D⁻ Generation in Negative Ion Sources: First Steps and Proof of Principle

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Abstract. In a proof-of-principle study, molybdenum samples were implanted with a very small dose of Cs in order to test the properties of the compound as a surface converter for negative hydrogen ion production. First results on the properties of Cs doped Mo compounds show a reduction of the work function and a stable H⁻ yield up to four hours in low density hydrogen plasma. The implanted Cs atoms were stable in the Mo lattice over one year for samples stored in vacuum and not exposed to the plasma. The surface H⁻ generation mechanisms were identified and a comparison of the negative ion yield with pure Mo showed that the Cs doped Mo sample's yield was much larger.

INTRODUCTION

Negative hydrogen ion sources are an active research topic relevant to many fields of application. For example, in the frame of neutron therapy [1], for spallation neutron sources [2] as well as for neutral beam injection (NBI) systems on LHD [3]. For ITER, H⁻/D⁻ NBI systems based on the small [4,5] and "half-size" [6,7] IPP prototype sources will be used. Consorzio RFX is currently building the full size ITER prototype sources [8,9]. Negative ions are also foreseen to be used by the proton synchrotron booster at CERN [10].

Negative hydrogen ion sources for ITER neutral beam injectors are based on the conversion of positive ions and neutrals into negative ions on a converter surface. The conversion yield strongly depends on the work function of the converter material: a low surface work function was shown to greatly increase the rate of formation of negative ions on the surface. The reduction of the work function on the plasma grid (PG), the most relevant converter surface, is usually achieved by evaporation of Cs. This greatly enhances the negative ion conversion yield [11,12,13] and is mandatory to extract large negative ion currents from current negative ion NBI sources. Because of its high intrinsic reactivity, Cs is passivated by impurities such as oxygen and water, leading to an increase of the work function and thus a degradation of the source performance. The evaporation of fresh Cs allows recovering high source performance, i.e. a high negative ion current and low co-extracted electron current.

For the small IPP prototype sources the Cs consumption was determined to range between 5 and 10 mg h⁻¹ [13]. In comparison, the half-ITER-size source on the IPP test bed ELISE, which has a larger source volume, grid surface and volume-to-surface ratio, an upper value of 4 mg h⁻¹ for

each of the two Cs ovens was determined, which is lower than expected [14]. This result shows that the scaling of the Cs consumption to larger sources is not straightforward. A tentative scaling was proposed based on the Cs consumption in the former IPP long pulse test stand MANITU. For an ITER-like DEMO source (0.2 m² extraction area, 60 A extracted current) it lead to a prediction of 350–700 g annual consumption if the injector is operated 80% of the year (see [13] for details).

In the IPP prototype sources only small amounts of Cs (around 1 g) are stored in the ovens. For DEMO according to the prediction a much larger storage is needed as well as the development of a refilling procedure suitable for large amounts of Cs and a radioactive environment. Another critical issue is the removal of the Cs accumulated in the source, while source maintenance intervals should be as long as possible.

All of the points discussed above make clear that the development of techniques that reduce Cs consumption or at best allow for Cs-free operation would lead to significant advantages.

It is expected that working with a low-work-function base material will require less Cs evaporation in order to achieve high performance, which in turn will ease the operations by reducing the time needed for conditioning and re-conditioning. Cs-doped Mo was already shown to possess a lower work function than pure Mo [15,16,17]. However, for an application to negative ion sources additional properties are important. In particular, the long term stability of the implanted Cs atoms must be assessed. Moreover, the stability of the negative ion yield during long plasma pulses, the reproducibility of the results as well as the recovery of the negative ion yield after venting should be demonstrated.

The ultimate goal of this research is to obtain a material whose work function is low and stable – both in vacuum and plasma – in order to reach high performance without Cs seeding or at least a significant reduction of the Cs consumption. In this article the very first steps towards validation of Cs-doped Mo as surface converter for negative ion sources are presented as a proof-of-principle study.

EXPERIMENTAL RESULTS AND DISCUSSION

Small size samples of 1 cm² made of 99.95% Mo were implanted with a very small dose of Cs. The aim was to build up a near-surface reservoir of Cs and a surface Cs concentration of only a few percent, which was deemed sufficient for this proof of principle and helped to reduce implantation costs. In order to determine the most suitable energy and dose of Cs to reach the aforementioned two properties, the depth profiles of the implanted atoms were simulated with SDTrimSP [18]. A two steps process was found to be most suitable. First a dose of $5 \cdot 10^{15}$ cm⁻² Cs atoms was implanted at 15 keV followed by the same dose at 10 keV, resulting in the very small total dose of $\approx 10^{16}$ cm⁻² of implanted Cs atoms. The first step permits the implantation of

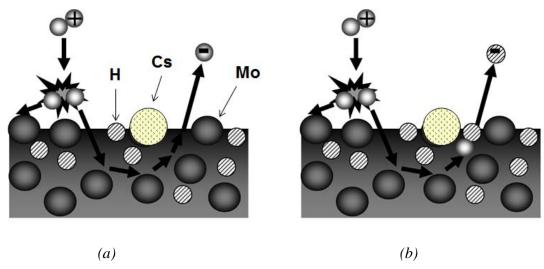


FIGURE 1. (a): Backscattering followed by electron capture of a H^0 originating from a dissociated H_2^+ ion, (b): sputtering as a negative ion of an adsorbed hydrogen atom by an H^0 originating from a dissociated H_2^+ ion.

the Cs deep into the Mo, up to 20 nm. However, the drawback is a large sputtering yield, leading to a poor surface and bulk atomic fraction. The second step at reduced energy reduces the implantation depth as well as the sputtering yield. Moreover the stopping power of the material is larger than pure Mo because of the

Cs atoms already implanted during the first step. After the second implantation step, one obtains samples with a surface Cs atomic fraction of 7% (0.07 monolayers) and a near surface reservoir of Cs (see [19] for more detail). The Cs reservoir is expected to counteract Cs diffusion into the bulk that would lead to a slow increase of the work function over time [19]. For samples stored in vacuum and not exposed to the plasma XPS depth profiling showed no diffusion of the Cs one year after implantation [19].

The negative ion surface generation mechanisms were investigated in hydrogen plasma. The hydrogen plasma density was around 10^{14} m⁻³, the electron temperature 0.5 eV and the plasma potential was 5 V. The positive ion composition determined by a mass spectrometer showed that it was largely dominated by H_3^+ ions with a proportion of 63%, followed by H^+ with 28% and H_2^+ with 9%.

The positive ions from the plasma are accelerated by the potential difference between the plasma and the sample potential and strike the sample. The negative ions generated on the surface by the positive ions are then repelled by the same potential difference and collected by an energy-resolved mass spectrometer (see [19,20,21,22]). Two H⁻ surface generation mechanisms are anticipated and presented in Figure 1: the backscattering followed by electron capture and the sputtering of hydrogen adsorbed on the surface as H⁻. As already shown for HOPG [19,20,21,22], the negative ions surface mechanisms can be distinguished by negatively biasing the surface (down to –130 V) and by increasing the sample temperature. Figure 2 shows a typical distribution function of the energy at which the H⁻ is created on the Cs-doped Mo and a pure Mo reference surface. For the backscattering mechanism, one can determine the theoretical maximum surface creation energy of a negative ion originating from any kind of positive ion: it is the sheath voltage drop between the sample and the plasma divided by the number of hydrogen atoms that the atomic or molecular ion is composed of, because the molecular ions dissociate upon impact on the surface. Here, for simplicity elastic backscattering with an infinitely heavy

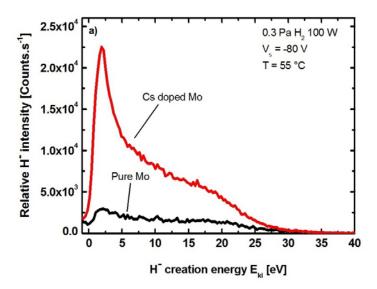


FIGURE 2. Comparison of the relative negative ion yield obtained with the Cs doped Mo samples and a pure Mo sample, both from the same raw material. The samples potential V_s was -80 V and the samples temperature 55 °C.

target atom is assumed, where no energy is lost during the interaction of the positive ion with the surface.

For the case presented in Figure 2, the sheath voltage drop between the plasma potential (5 V) and the sample (-80 V) is then 85 V. One can clearly see that the signal virtually vanishes above a surface creation energy of 28 eV (= 85 eV/3) because H_3^+ is the dominant positive ion from the plasma. The few negative ions having a larger energy than 28 eV were created from H_2^+ or a proton. Thus, the negative ions contributing to the high energy part of the distribution functions are created by the backscattering mechanism [19]. Similar conclusions could be drawn concerning HOPG [20,21,22].

At high temperatures [19] the main peak almost disappears which can be explained by the disappearance of the hydrogen adsorbed on the surface and trapped in the bulk, the remaining signal being attributed to the backscattering mechanism. The assignment of the sputtering signal to this main peak is confirmed by SDTrimSP calculations, showing that the expected energy of the hydrogen atoms sputtered from the surface is less than 7–8 eV at –130 V sample bias and 5–6 eV at –80 V bias.

From Figure 2, one can also clearly see the magnitude of the difference between the relative negative ion yields of the Cs-doped Mo and the pure Mo samples, both manufactured from the same raw material. It is possible to give a rough estimate of the work function reduction induced by the Cs implantation. For metals, it is known that the conversion probability into negative ions of incoming species is $P^- \propto e^{-\frac{\phi-A}{\varepsilon}}$ [23,24] with \emptyset being the work function, A the electron affinity and ε a parameter in the range of 0.2-0.4 eV [25,26].

Given the work function of pure Mo (4.6 eV), the ratio of the integrated curves from Figure 2 and the above relation, one finds a work function reduction for the Cs doped Mo samples between $-0.8 < \Delta \emptyset < -0.4$ eV, in good agreement with existing measurements [27]. The work function of the implanted samples will be measured and compared to this rough estimate in future experiments.

CONCLUSION

The very first results concerning the Cs-doped Mo as possible surface converter for negative ion sources are presented. In particular, the surface generation mechanisms were identified: the backscattering mechanism is responsible for the high energy part of the distribution function, while the main low energy peak is mostly due to the sputtering of adsorbed hydrogen as a negative ion with only a smaller contribution of the backscattering. A comparison of the relative negative ion yield of the Cs-doped Mo to pure Mo allowed to determine that the work function reduction was comprised between -0.8 and -0.4 eV, which appears substantial for only 7% surface Cs atomic fraction.

These results are encouraging for further investigations of the properties of Cs-doped Mo compounds. In particular, the stability of the implanted layers should be tested with samples implanted with a larger surface and bulk atomic fraction. Moreover, the stability of the negative ion yield and of the implanted atoms should be investigated when exposed to hydrogen plasma with a much larger flux and fluence.

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