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Interactions of Solid and Liquid Lithium
with Steady State Hydrogen and Helium Plasmas

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Abstract. A variety of innovative Plasma-Facing Component (PFC) concepts, employing moving solid or liquid surfaces, have recently been proposed in order to resolve technical issues, associated with the applications of currently used PFCs in future steady state fusion devices. As the first step to evaluate the concept using flowing-liquids for PFCs, steady state hydrogen and helium plasma interactions with solid and standing liquid lithium have been investigated in the present work, using the H_α and He-I spectroscopy at the ion bombarding energies up to 150eV and at the lithium temperatures between room temperature and 480°C. Data indicate that hydrogen recycling over liquid lithium is clearly reduced, relative to that over solid lithium, whereas helium recycling does not show the same trend. From the kinetic analysis of these recycling time constant data, the activation energies for the overall recycling processes have been evaluated to be 0.02 ± 0.01eV, both for hydrogen and helium plasmas. Also, it has been found that the activation energy is nearly independent of ion bombarding energy.

1. Introduction

It is widely recognized in the magnetic fusion research community that since the discovery of TFTR's Supershot in late 80's, high performance core plasmas tend to favor low edge recycling conditions. Therefore, wall conditioning such as boronization has routinely been conducted in many confinement experiments. Unfortunately, however, due to the surface saturation with implanted particles, the efficacy of boronization to maintain low edge recycling conditions has finite lifetime, necessitating re-conditioning. This clearly points to a need for enabling wall concept development to provide reduced recycling even at steady state for future long-pulse and/or steady state fusion devices beyond the International Thermonuclear Experimental Reactor (ITER).

In an attempt to provide a possible resolution to this steady state recycling issue, the concept of Moving-Surface Plasma-Facing Component (MS-PFC) was proposed nearly a decade ago [1, 2]. Recently, laboratory-scale Proof-of-Principle (PoP) experiments have been conducted, employing a continuously Ti- or Li-gettered rotating drum exposed to hydrogen plasmas, and the results indicate that recycling can be reduced down to 95% for Ti and 75% for Li even at steady state, demonstrating "unsaturable walls" [3, 4, 5].

The success on these PoP experiments on moving-solid surfaces has now redirected our interest to moving-liquid surfaces, the concept often referred to as "liquid (metal) waterfall". In the this work, because of its high hydrogen absorptivity, low melting point, and low atomic number, lithium has been selected as a candidate material for this application. However, lithium is not yet set for flowing in the present experimental setup because we would rather obtain fundamental knowledge on standing liquid lithium interactions with steady state hydrogen and helium plasmas.

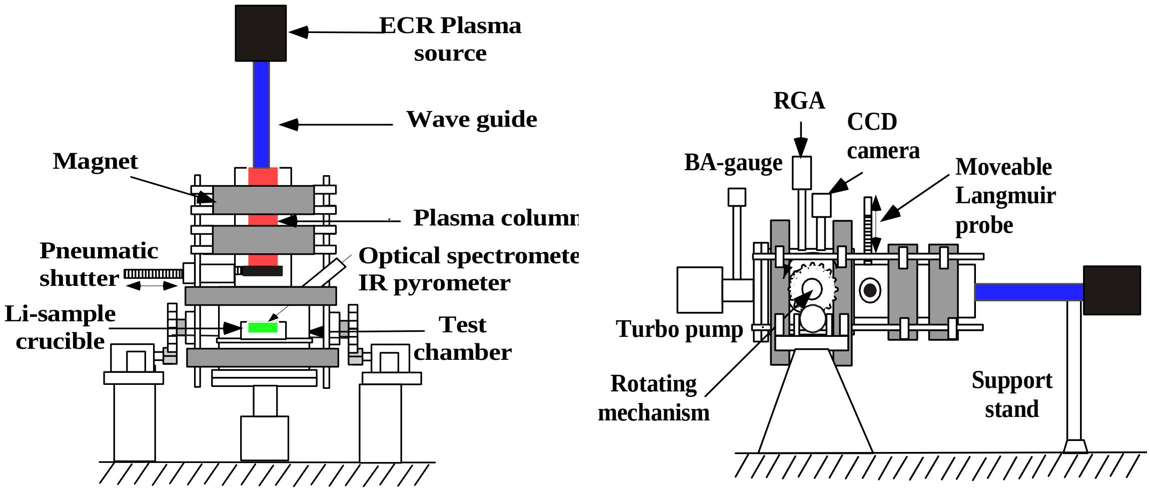
From this point of view, hydrogen and helium plasma recycling behavior over solid and liquid lithium has been observed in a newly build facility: Vehicle-1 (for the **Ve**rtical and **H**orizontal positions **I**nterchangeable test stand for **C**omponents and **L**iquids for fusion **E**xperiments) [6]. Also, the relevant data taken from moving-solid lithium PFC experiments [3, 4, 5] will be referred to, as-needed, to make a comparison with those obtained from the moving-liquid cases.

2. Experimental

A schematic diagram of the Vehicle-1 facility is shown in Fig. 1-(a) and (b). Unlike other plasma-surface interactions research facilities such as PISCES-B [7], Vehicle-1 can take two positions, as can be seen, and is used in its vertical position in the present work. All the details of this facility have already been described elsewhere. However, for the sake of completeness, some of the most important features of Vehicle-1 will be briefly described below.

Vehile-1 employs a 1kW ECR plasma source and can generate steady state hydrogen, helium, argon, nitrogen and oxygen plasmas with densities of the orders of $10^{10\text{-}11}$ 1/cm^3 and electron temperatures typically 4~5 eV. The ion temperature is believed to be near thermal unless associated with the Franc-Condon process. However, the ion bombarding energy can be controlled by applying a DC bias between the sample assembly and the plasma chamber at the floating and plasma potentials, respectively. The plasma column diameter is limited to about 3.5cm by a donut limiter made of tantalum, as shown later, providing a relatively flat density profile for the projected area over lithium in the form of circular disk with the diameter of 2.9cm and thickness of 2mm.

The diagnostics available for plasma-surface interactions include: a scanning Langmuir probe, partial and total pressure gauges, digital CCD camera, and an Optical Multi-channel Analyzer (OMA) connected with an optical fiber cable aiming at the pre-sheath region near the sample surface. Importantly, the temperature of lithium is measured with two thermocouples: one attached to the bottom of the crucible made of molybdenum; and the other attached to lithium directly on the plasma-facing surface. The crucible is 3cm in diameter and is mounted on a resistive heating assembly that can heat lithium up to around 650°C, sufficiently high to induce the decomposition of lithium hydride (LiH) in vacuum. Therefore, the sample temperature control was done using the combination of the plasma bombarding power flux and resistive heating.



(a) (b)

FIG. 1 A schematic diagram of the experimental facility: Vehicle-1[6] in its (a) vertical position; and (b) horizontal position.

3. Results and discussion

3-1. Plasma characteristics in Vehicle-1

As presented in our recent paper [6], the plasma density increases from 1 x 10^{10} to 1 x 10^{11} 1/cm^3 almost linearly with increasing ECR power from 100W to 1kW, whereas the electron temperature tends to remain 4~5 eV. In the present work, the ECR power was set at relatively low levels so that lithium temperature would not be overheated by the power flux associated with incident plasma ions.

Shown in FIG.2-(a) are the radial profiles of plasma density and electron temperature taken at the ECR power of 100W across the plasma column, the diameter of which is defined by a tantalum donut limiter. As seen, these profiles are rather flat.

Using the CCD video camera, the H_α intensity profile was taken at the ECR region as that is viewed for hydrogen recycling measurements to be described next. Results are shown in FIG. 2-(b) with and without a DC-bias. Notice that the H_α profile is raised with the DC-bias on, due to the enhanced reflection of electrons, while both exhibit the characteristic pre-sheath profiles.

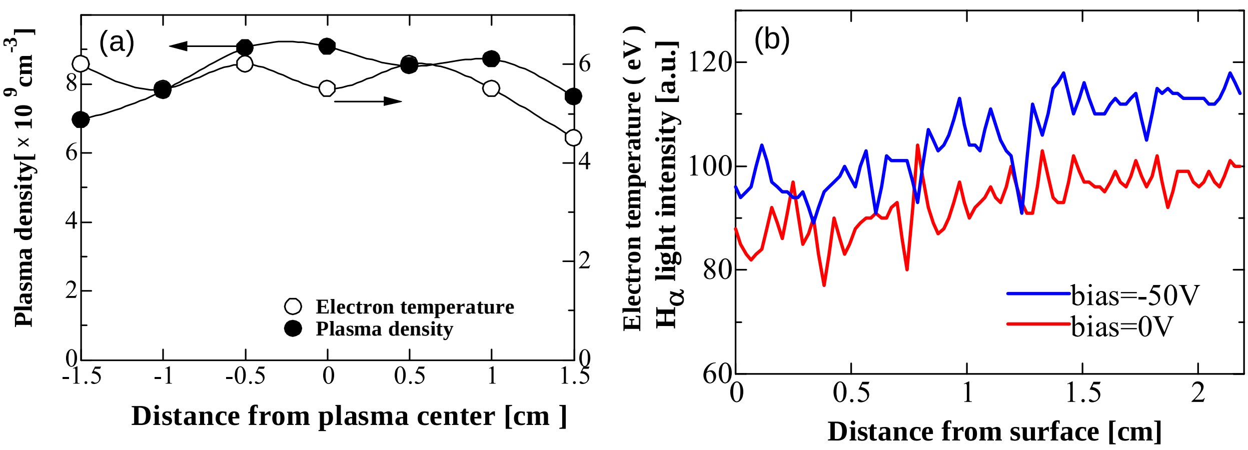


FIG.2 Plasma characteristics in Vehicle-1:(a) Radial profiles of plasma density and electron temperature; and (b) Axial profiles of H_α intensity in the pre-sheath region.

3-2. Hydrogen and helium plasma recycling measurements

Hydrogen and helium recycling measurements were conducted, setting the solid and liquid lithium temperature at 50°C and 300°C, respectively. Shown in FIG. 3-(a), (b) and (c) are the time evolution curves of H_α and He-I light intensities that are taken as the measures of respective plasma recycling for the following reasons. In our recent experiments [3, 4, 5], the H_α intensity has been found to be more closely related to the molecular hydrogen density in the pre-sheath region than to the atomic hydrogen density, the latter of which one would expect be more relevant though. Similarly, we assume that the He-I intensity is governed by the neutral helium density recycled from the surface rather than by that in the host plasma.

These time evolution curves are fitted with the following empirical formula [8] to evaluate the recycling time constant, τ_r:

$$I(t) = I^{\infty}\{1 - \exp(-\frac{t}{\tau_r})\} \qquad (1),$$

where I and I^{∞} are the intensities of observed H_α or He-I at t = t and at steady state (i.e.

$t=\infty$), respectively.

Notice that the steady state H_α measured for solid lithium (see FIG. 3-(a)) intensity generally increases with increasing ion bombarding energy, consistent with the data shown in FIG. 2-(b). It is also true that the recycling time constant tends to increase with increasing ion bombarding energy. This is because as the ion bombarding energy becomes larger, the implantation depth becomes larger, which then increases the particle retention capacity. Trends similar to these were observed in our

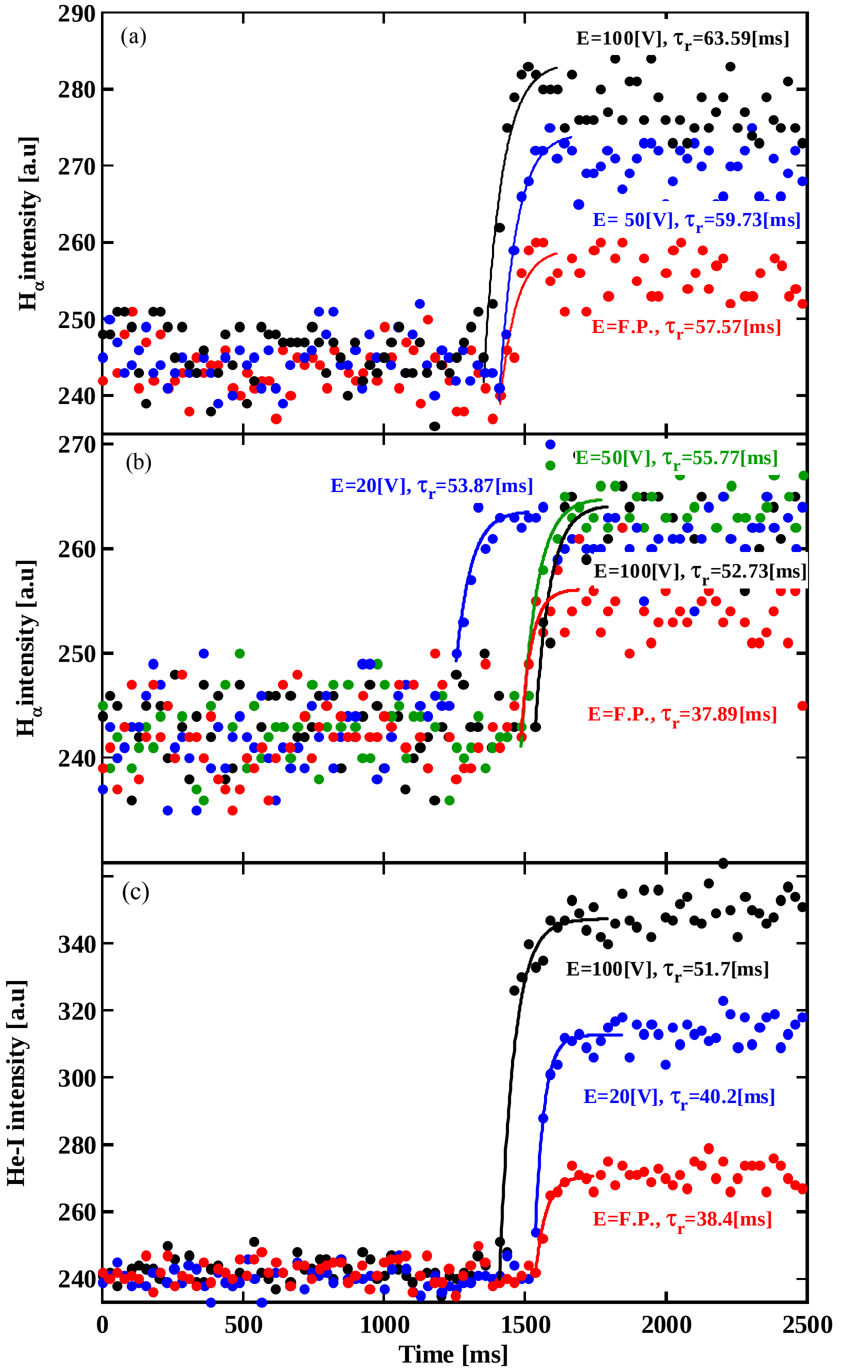


FIG. 2 Time evolution of (a) hydrogen recycling over solid lithium at 50°C, (b) hydrogen recycling over liquid lithium at 300°C, and (c) helium recycling over liquid lithium at 300°C, where E is the ion bombarding energy and F.P. is the floating potential (<10eV).

recent MS-PFC experiments using a lithium-gettered rotating drum, the temperature of which is maintained essentially at room temperature [5].

It is extremely important to note here that hydrogen recycling over liquid lithium does not show these trends (see FIG. 3-(b)). In contrast, the steady state H_α intensity tends to saturate only above 20V and the recycling time constant does not appear to increase even as the bias voltage increases. One might explain these findings as follows: Hydrogen migration in liquid lithium, obeying the Einstein-Stokes' law, is orders of magnitude faster than that in solid lithium, obeying the Fick's law. This in turn enhances the transport of hydrogen deeper into liquid lithium, which can then retard the saturation of the implantation range.

It is highly likely that under the present experimental conditions, implanted hydrogen exceeds the solid solution concentration within the range, forming lithium hydride (LiH). Interestingly, LiH is a solid up to its decomposition temperature of ~650°C, leading to the two-phase condition of the Li-H system [9]. Therefore, perhaps forming clusters, segregated LiH tends to "sink" in the host liquid due to the microgravity effect. This again enhances the transport of hydrogen into the bulk of liquid lithium.

Therefore, it is considered that when hydrogen plasmas interact with liquid lithium, reemission of molecular hydrogen back to the surface will most likely be suppressed until the liquid is fully hydrogenated. This allows us to expect to see reduced hydrogen recycling over flowing liquid lithium even at steady state, as has been predicted elsewhere [10].

As shown in FIG. 3-(c), helium recycling over liquid lithium generally exhibits the similar trends to those observed over solid lithium. This is presumably because lithium has very little solubility of helium in it and they do not form any chemical compounds.

3-3. Kinetic analysis of hydrogen and helium plasma recycling behavior

In addition to the definition by eq. (1), the recycling time constant can also be given by the following relation [11]:

$$\tau_r = \frac{D}{k_r \nu \Phi} \qquad (2),$$

where D is the diffusion coefficient of hydrogen in lithium, k_r is the surface recombination coefficient, ν is the sticking (i.e. trapping) coefficient, and Φ is the incoming particle flux. Generally, the reciprocal time constant is equivalent to the reaction rate constant, which may be expressed as follows:

$$\frac{1}{\tau_r} = \frac{k_o \nu_o \Phi}{D_o} \exp(-\frac{E_r - E_s - E_d}{kT}) \qquad (3),$$

where k_o, ν_o, and D_o, and E_r, E_s, are E_d are the frequency factors and the activation energies of the surface recombination, sticking and diffusion processes, respectively, and also k is the Boltzman constant, and T is the absolute temperature. Here, one must be aware that if the DC bias voltage is sufficiently large to induce ion implantation, ν_o, and E_s, would be eliminated from eq. (3). Likewise, in the case of helium plasma bombardment, k_o and E_r should be ignored in eq. (3). Interestingly, although one might consider otherwise, in eq. (3) there is no factor related to the depth of implantation, i.e., the ion bombarding energy.

Shown in Fig. 3-(a) and (b) are the Arrhenius plots of these ($1/\tau_r$) data, taken from hydrogen and helium recycling measurements, respectively. The activation energies evaluated from the straight lines have been found to be 0.02±0.01eV, surprisingly for both hydrogen and helium plasmas, regardless whether lithium is a solid or liquid.

Immediately from eq. (3), one predicts not only that the activation energy for the overall recycling process could be a minute yet positive value, but also that it might possibly be a negative value, depending on the process dominating the overall recycling, as has actually been found in our recent work [6].

Shown in FIG. 5 are these activation energies plotted as a function of ion bombarding energy. One finds that there is almost no dependence on ion bombarding energy, as expected from eq. (3). However, because not all these individual activation energies are available from the literature, the further evaluation of the present data can not be done at this point, which no doubt warrants future work in this area.

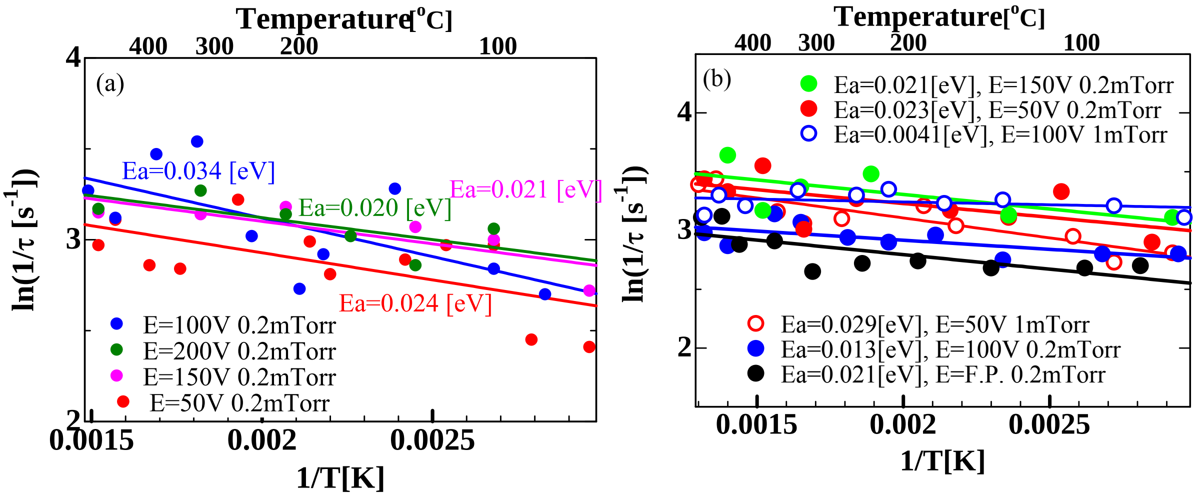


FIG. 4 Arrhenius plots of the reciprocal recycling time constants taken from:
(a) hydrogen; and (b) helium recycling experiments, where E_a is the activation energy.

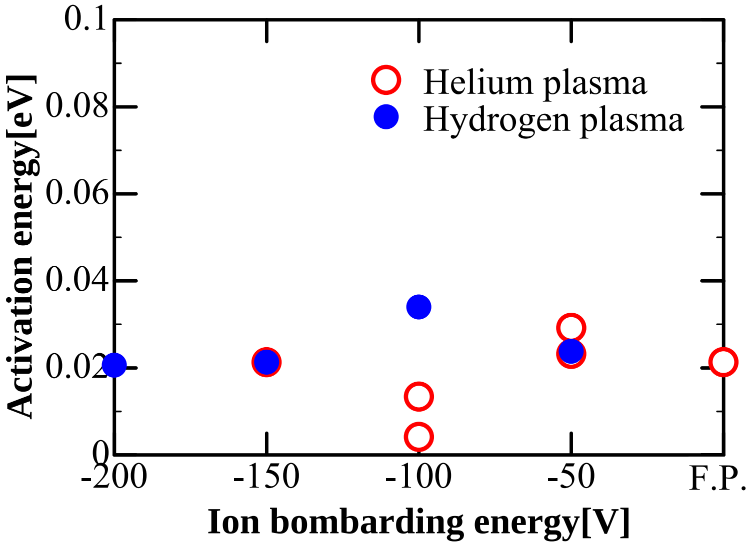


FIG. 5 Activation energies for hydrogen and helium plasma recycling over lithium as a function of ion bombarding energy.

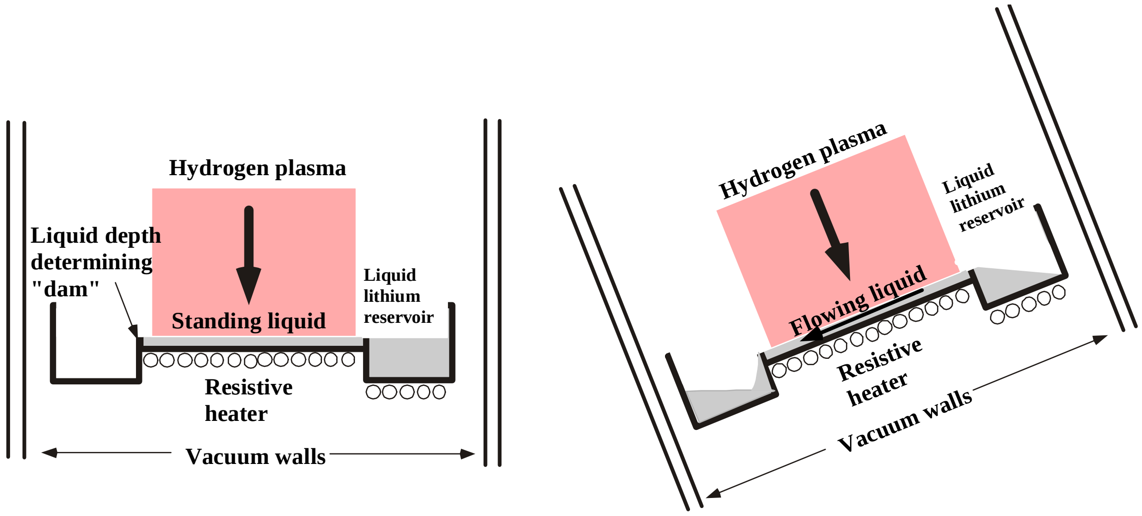
4. Summary and future plans

Steady state hydrogen and helium plasma recycling behavior over solid and liquid lithium has been measured with optical spectroscopy, using the newly built Vehicle-1 facility. Observations have indicated that hydrogen plasma recycling over liquid lithium tends to prevent the surface saturation, perhaps because of the rapid transport of implanted hydrogen into the bulk of liquid lithium. In contrast, this is not the case with helium because the solubility in lithium of which is known to be very little, hence leading to instant reemission.

From the kinetic analysis of the reciprocal recycling time constant data, the activation energies for the overall hydrogen and helium recycling have been evaluated and those obtained for hydrogen and helium are surprisingly close to each other and also they are nearly independent of ion bombarding energy.

In our separate experiments on the MS-PFC concept [5], employing a continuously lithium-gettered rotating drum, it has been demonstrated that steady state hydrogen recycling can be reduced down as low as 75%. From these data and together with the findings in the present work, one immediately expects that hydrogen recycling over flowing liquid lithium will also be reduced even at steady state. Nonetheless, more direct PoP experiments are awaited to be conducted on the liquid metal waterfall concept.

For this purpose, a two-reservoir "seesaw type" flowing liquid lithium setup is currently designed for the next step experiments. A schematic diagram of this setup is shown in FIG. 5. In this setup the liquid lithium flow is driven only by the gravity for the sake of simplicity. The depth of the liquid is determined by a small "dam" and the liquid flows from one reservoir into the other over a resistively heated bridge. Also, it is important to mention here that this seesaw setup takes advantage of the rotating mechanism of Vehicle-1(see FIG.1), so that the plasma column and all the diagnostics will be inclined together when the flowing liquid interacts with the plasma, eliminating the possible errors in data due to the possible changes in relative distances between the spot of measurements and sensor heads, etc.



(a) (b)

FIG. 4 A flowing liquid lithium experimental setup to be mounted in Vehicle-1 set in its: (a) vertical position; and (b) inclined position [towards the horizontal position].

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