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A new versatile facility: Vehicle-1 for innovative PFC concepts evaluation and its first experiments on hydrogen recycling from solid and liquid lithium

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Abstract

A new plasma facility: Vehicle-1 has been built for the evaluation of innovative plasma-facing component concepts. This facility can conduct experiments in such a way that standing liquids in a tray are exposed to vertically flowing plasmas, or that flowing liquids on a slope are bombarded with horizontally directed plasmas. Vehicle-1 can generate steady state hydrogen plasmas with densities of the order of 10^{10} cm^{-3} and electron temperatures around 4 eV. Hydrogen recycling behavior has been observed in Vehicle-1, and the Arrhenius plot of rate constants exhibits a break at around 300 °C. The activation energies are −0.0096 eV and 0.17 eV, respectively, below and above the break. To understand the kinetics of hydrogen recycling, particles sticking coefficients have been measured. Results indicate that at temperatures below the break the sticking process appears to be rate-limiting, whereas above the break surface recombination is important. The sticking coefficients for plasma species have been found to be orders of magnitude larger than those for hydrogen molecules.

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1. Introduction

Since the discovery of TFTR's Supershot [1], high performance confinement plasmas have often been found to favor low edge recycling conditions. Therefore, wall conditioning such as boronization is conducted in many confinement experiments. Unfortunately, however, due to the surface saturation with implanted fuel and impurity particles, the efficacy of boronization has finite lifetime to maintain low recycling conditions. Therefore, the application of these wall conditioning techniques will be limited in steady state fusion devices.

Currently used plasma-facing components would also suffer from other problems including erosion, tritium co-deposition, radiation damage, and pulsed heat cycles from ELMs. These arguments point to a need for enabling wall concepts development over the next several decades, perhaps while ITER is running.

To resolve these technical issues, the concept of moving-surface plasma-facing component (MS-PFC) was proposed about a decade ago [2,3]. Recently,

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laboratory-scale proof-of-principle experiments have been conducted, employing a Ti-or Li-gettered rotating drum exposed to hydrogen plasmas, and the results indicate that particle recycling can be reduced down to 90 ~ 95% even at steady state [4,5]. The success on these experiments has encouraged us to challenge the use of flowing liquids as plasma-facing surfaces in magnetic fusion devices beyond ITER.

Lithium is selected as a candidate material for moving-liquid PFC applications because of its low atomic number and melting point, and high hydrogen absorptivity forming hydrides (LiH). At early stages of the challenge, however, we must obtain fundamental understandings of hydrogen plasma interactions with solid and liquid lithium. In the present work, therefore, liquid lithium is not yet set for flowing but standing under plasma bombardment. Ideally, liquids should be held in a horizontally placed tray, and hydrogen plasmas impinge on them in the vertical direction. A new facility has been built to provide these conditions. This is the critical difference from existing experiments [6] where horizontally directed plasmas impinge on liquids, holding themselves against the gravity with surface tensions.

2. Experimental

A new facility: Vehicle-1 (the Vertical and Horizontal positions Interchangeable test stand for Components and Liquids for fusion Experiments) was proposed in our previous work [5] and has been built for the present work. As shown in Figs. 1(a) and (b), Vehicle-1 can take two positions: (1) horizontal position similar to other plasma facilities such as PISCES-B [7]; and (2) vertical position like no other facility. In the present work Vehicle-1 is used in the vertical position.

The Vehicle-1 facility employs a 1 kW 2.45 GHz ECR plasma source which can generate steady state hydrogen and helium plasmas. These plasmas are magnetized at around 300 G, flowing linearly down to interact with lithium. For well-diagnosed experiments, Vehicle-1 is installed with a movable Langmuir probe, a digital CCD camera, an optical spectrometer, a total and partial pressure gauges for plasma characterization, and equipped with an infrared pyrometer, two thermocouples and a resistive heater for temperature measurements and control, respectively.

A molybdenum crucible with the diameter of 3 cm, holding lithium in it, is mounted on the resistive heater. The two thermocouples are used such that: one pressed on the lithium surface (for the temperature to be denoted as T_t); and the other attached to the bottom of the crucible (for T_b). The ion temperature is at most a few electron volt, associated with Franck–Condon dissociation. However, the ion bombarding energy can be increased by applying a negative DC bias between the crucible and test chamber, respectively, at the floating and plasma potentials.

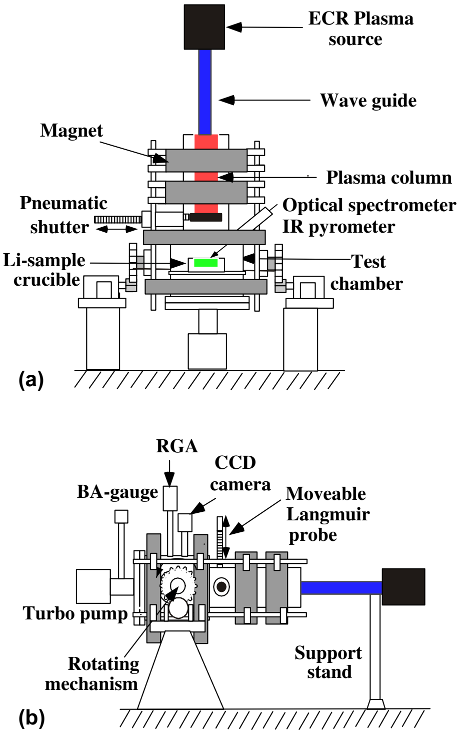

Fig. 1. A schematic diagram of the Vehicle-1 facility: (a) in the vertical position; and (b) in the horizontal position.

Because lithium tends to form surface oxide and hydroxide, all samples were heated to ~250 °C for about 10 min to reduce surface contamination prior to plasma exposure. Plasma exposure was controlled by a pneumatically-operated shutter made of molybdenum, the opening time of which is ~20 ms.

3. Results and discussion

3.1. Plasma characterization in Vehicle-1

The plasma densities and electron temperatures measured with the Langmuir probe are shown in Fig. 2 as a function of ECR power. The hydrogen pressure was maintained at 1.5×10^{-3} Torr in these measurements. The plasma density increases with increasing ECR power, which, however, is not the case with electron temperature.

The plasma column diameter is defined to be about 3.5 cm by a tantalum donut limiter positioned upstream. Although the radial profiles of plasma density profile and electron temperature are not shown here, these are generally found to be uniform within ±10% over the projected area of the sample crucible mentioned earlier.

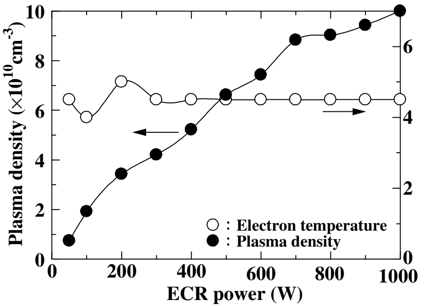

Fig. 2. Plasma density and electron temperature as a function of ECR power in Vehicle-1.

3.2. Hydrogen recycling behavior and its kinetic analysis

Solid and liquid lithium were bombarded with 400 W hydrogen plasmas to observe recycling behavior. As an indicator of recycling, the volume-integrated intensity of H_α was measured in the pre-sheath region. In our previous work [5], the H_α intensity in the pre-sheath region was found to be correlated with recycling hydrogen molecules rather than hydrogen atoms, the latter of which one would expect to be more important, though. The same is believed to be true in the present work.

The time evolution of hydrogen recycling is shown in Fig. 3(a). To refresh surfaces, 50 W helium plasma bombardment was done between hydrogen plasma exposures. These curves are fitted with the following empirical relation to determine the recycling time constant,τ_r [9]:

$$I_{H_\alpha}(t) = I^{\infty}_{H_\alpha}\left\{1 - \exp\left(-\frac{t}{\tau_r}\right)\right\}, \tag{1}$$

where t is the time, I_{H_α} and $I^{\infty}_{H_\alpha}$ are the intensities of H_α observed at $t = t$ and at steady state (i.e. $t = \infty$), respectively. The recycling time constant can also be expressed as follows [10]:

$$\tau_r = \frac{D}{k_r \Phi}, \tag{2}$$

where D is the diffusivity, k_r is the surface recombination coefficient, and Φ is the net implanted flux such that $\Phi = \nu\Gamma$, where ν is the sticking coefficient, and Γ is the incoming flux.

Because the reciprocal time constant is equivalent to the rate constant, $1/\tau_r$ data are plotted in Fig. 3(b) in the Arrhenius manner. Notice that there is a break at around 300 °C, and the activation energies have been calculated to be −0.096 eV and 0.17 eV, respectively, in the temperature ranges below and above the break.

The break in the Arrhenius relation indicates a change of the rate-limiting step. From the constituents in Eq. (2) one conjectures several rate-limiting processes: diffusion, recombination and sticking. Unlike diffusion and recombination, the sticking coefficient usually decreases with increasing temperature. This is generally true with molecular hydrogen. However, particles "compound" sticking coefficients during plasma exposure are yet-to-be explored and these will be described next.

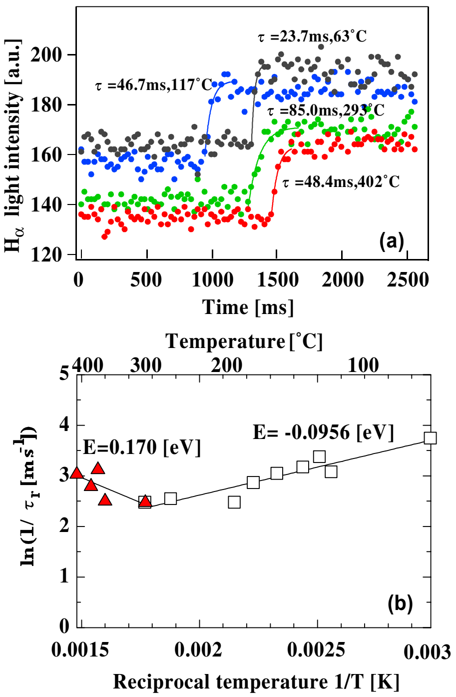

Fig. 3. Hydrogen recycling behavior from solid and liquid lithium: (a) time evolution of H_α intensities and its time constant evaluation; and (b) Arrhenius plot of reciprocal time constants.

3.3. Sticking coefficient measurements

Sticking coefficient measurements have been conducted at $T_t = 100$ °C and $T_t = 290$ °C, the latter of which is relevant to the break seen in Fig. 3(b). These temperatures are intended for solid and liquid lithium. First, as-received lithium was exposed to hydrogen gas at the pressure of 1.5×10^{-3} Torr, the same pressure as for plasma operation, for 30 min. Next, to see the effect of surface contamination, as-received lithium was bombarded with a 50 W helium plasma prior to hydrogen gas exposure, each for 30 min. Finally, lithium was bombarded helium and hydrogen plasmas both at 50 W consecutively, each for 30 min.

These hydrogen gas and plasma exposed lithium samples were heated up to near the lithium hydride decomposition temperature (~650 °C), so that the total uptake of hydrogen can be evaluated. Also, as-received samples were heated to find out the initial hydrogen contents. As shown in Fig. 4, the desorption spectra are

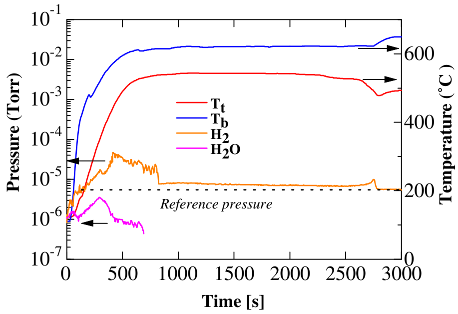

Fig. 4. Thermal desorption from lithium exposed to a hydrogen plasma at 100 °C.

generally characterized by two peaks. Surprisingly, this was true with as-received samples.

The first peak appears at around 200 °C, which is believed to be contributed from the decomposition of adsorbed water and also the desorption from the solid solution phase of the lithium–hydrogen system [11]. The second peak is observed at around 550 °C, due to the decomposition of lithium hydride and hydroxide, the latter of which, one suspects, resulted in the second peak for as-received lithium. After these two peaks were through, the crucible was always found to be empty, indicating the complete evaporation and decomposition of lithium and its compounds.

The sticking coefficient of hydrogen molecules, v_{H_2}, is defined as follows:

$$v_{H_2} = \frac{\int_0^{t_d} \{P_{H_2}(t) - P^o_{H_2}\} S_{H_2} \mathrm{d}t}{\Gamma_{H_2} t_{ex}}, \tag{3}$$

where t_d is the desorption time, $P_{H_2}(t)$ is the pressure of hydrogen, $P^o_{H_2}$ is the reference level pressure, S_{H_2} is the pumping speed,Γ_{H_2} is the arrival rate of hydrogen molecules and t_{ex} is the exposure time. Here, the reference pressure is defined as the one at which T_t drops, indicating the loss of contact pressure for the thermocouple due to lithium evaporation. Partial pressures of other hydrogen-containing molecules such as methane were negligibly small. Therefore, only $P_{H_2}(t)$ and $P^o_{H_2}$ are taken into account in Eq. (3).

The denominating factor in Eq. (3), Γ_{H_2}, is given by the following formula [12]:

$$\Gamma_{H_2} = \frac{P_{H_2}(t)}{\sqrt{2\pi m_{H_2} k_B T}}, \tag{4}$$

where m_{H_2} is the molecular weight of hydrogen, and k_B is the Boltzman constant. In the case of plasma exposure, the ion flux is estimated from the following formula [8]:

$$\Gamma_{H^+} \approx \frac{1}{2} n_e \sqrt{\frac{k_B T_e}{m_H}}, \tag{5}$$

where m_H is the atomic weight of hydrogen, n_e and T_e are the plasma density and electron temperature, respectively. Therefore, the denominating factor in Eq. (3) should be the total incoming flux: $\Gamma_{tot} = \Gamma_{H^+} + 2\Gamma_{H_2}$.

At electron temperatures around 4 eV, however, the plasma species composition cannot be 100%H^+, necessitating corrections on the total incoming flux. The following assumptions have been made. First, from the results of modeling [14], the plasma species composition is assumed to be 60%H_3^+, 30%H_2^+, and 10%H^+. Also, it is assumed that the concentration of atomic hydrogen is the same as H^+, related to Frank–Condon dissociation, and half the atomic hydrogen (H^0) flux is directed towards lithium. Finally, atomic hydrogen is assumed to have the same energy as ionic species. Therefore, the corrected total incoming flux, Γ'_{tot}, is given as follows:

$$\Gamma'_{tot} = 0.6 \times 3\Gamma_{H_3^+} + 0.3 \times 2\Gamma_{H_2^+} + 0.1\Gamma_{H^+} + 0.1 \times 0.5\Gamma_{H^0} + 2\Gamma_{H_2} = 2.55\Gamma_{H^+} + 2\Gamma_{H_2}. \tag{6}$$

The corrected data are shown in Table 1. Predictably, the correction effect is barely noticeable because, again, the total incoming flux is dominated by molecular hydrogen under the present conditions such that $100\Gamma_{H^+} \approx \Gamma_{H_2}$.

The sticking coefficients are summarized in Table 1. From the comparison between cases #1 and #2, the sticking coefficient significantly increases with helium plasma pre-bombardment, indicating the surface cleanliness effect. Comparing cases #2 between #3, the sticking coefficient of molecular hydrogen decreases with increasing temperature. Last, from the comparison between cases #4 and #5, the compound sticking coefficient in the case of plasma exposure decreases with increasing temperature. Interestingly, however, the opposite is true when the sticking coefficients are evaluated for plasma species, as will be described in detail.

These findings explain the general trend of the $1/\tau_r$ plot before the break, shown in Fig. 3(b). After the break one assumes that hydrogen recycling is either diffusion-limited or recombination-limited. However, once lithium is liquefied, the transport of LiH, obeying the Einstein–Stokes law [13], will be significantly faster than solid state diffusion. Therefore, at temperatures after the break, surface recombination is considered to be rate-limiting, as was pointed out elsewhere [6].

Using the data in Table 1, the contribution from molecular hydrogen can be subtracted from the total desorption quantities measured in plasma exposure cases. As shown in cases #6 and #7, the compound sticking coefficients for plasma species: H_3^+, H_2^+, H^+ and H^0 are 0.37 and 0.51, respectively, at 100 °C and 290 °C. Though the bombarding energy is ~10 eV, estimated from $E \approx -3kT_e$, the sticking coefficients for plasma species are significantly larger than those for molecular hydrogen.

Table 1
Summary of sticking coefficient measurements

#	Experimental conditions		Compound sticking coefficient
	Exposed gas or plasma	Temperature (°C)	
#1	As-received Li, Exposed to H_2	$T_t = 100$, $T_b = 120$	3.6×10^{-5}
#2	Exposed to He-plasma and to H_2	$T_t = 100$, $T_b = 120$	3.2×10^{-3}
#3	Exposed to He-plasma and to H_2	$T_t = 290$, $T_b = 360$	3.9×10^{-4}
#4	Exposed to He-plasma and H-plasma	$T_t = 100$, $T_b = 120$	4.2×10^{-3} (4.19×10^{-3})[a]
#5	Exposed to He-plasma and H-plasma	$T_t = 290$, $T_b = 360$	1.82×10^{-3} (1.81×10^{-3})[a]
#6	Deduced for plasma species[b]	$T_t = 100$, $T_b = 120$	0.37
#7	Deduced for plasma species[b]	$T_t = 290$, $T_b = 360$	0.51

[a] After plasma species mix corrections (see text).

[b] H_3^+, H_2^+, H^+, and H^0.

4. Summary

A new versatile facility: Vehicle-1 has been constructed and its first experiments on hydrogen recycling from solid and liquid lithium have successfully been conducted. From the kinetic analysis of hydrogen recycling, the rate-limiting step has been found to change from surface sticking to recombination at around 300 °C. Also, it has been found that molecular sticking is sensitive to surface contamination. Despite their low energies plasma species exhibit orders of magnitude higher sticking coefficients than molecular hydrogen. This allows us to predict that lithium waterfall PFCs can provide low edge recycling conditions even at steady state.

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