



Helium retention and diffusivity in flowing liquid lithium

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Abstract

The flowing liquid surface retention experiment (FLIRE) has been designed to provide fundamental data on the retention and pumping of He, H and other species in flowing liquid surfaces. The FLIRE facility currently uses an ion beam source, which injects ions into a flowing stream of liquid lithium. Its design allows the liquid lithium to flow between two vacuum chambers that become isolated from each other when the lithium flows. Flow velocities between 0.5 and 3.0 m/s down two ramps inside the upper vacuum chamber can be achieved. The ramps and lines where the liquid lithium flows are heated to temperatures ranging from 250 to 500 °C to prevent any possible freezing. A dual residual gas analyzer system monitors the partial pressure of the implanted species in both vacuum chambers. The release rate of gas atoms in the second chamber is directly related to the mechanisms of transport within the metal bulk and also the process of desorption from the surface. For the case of helium, the diffusion coefficient was calculated to be $4.5 \times 10^{-3} \text{ cm}^2/\text{s}$ at 250 °C, with an uncertainty of $\pm 2 \times 10^{-3} \text{ cm}^2/\text{s}$. Helium retention coefficients on the order of 10^{-4} were obtained based on the experimental data.

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1. Introduction

Finding materials able to withstand very high heat fluxes (5–10 MW/m² during normal operation, and up to five times more during disruptive events [1,2]) and able to extract energy efficiently is a key issue in the successful development of tokamak fusion reactors. Research in advanced plasma facing components (PFCs) is therefore of critical importance for the development of future generation tokamak reactors. One strong candidate as a PFC is liquid lithium, which has been proposed as a divertor material in a number of studies [3,4]. Some of the challenges facing this application include: effective hydrogen/helium particle retention, plasma–liquid interaction issues (evaporation, sputtering, splashing),

chemical safety issues, tritium inventory, neutron irradiation effects, power extraction, vapor shielding, and macroscopic liquid-metal (LM) removal. Recent sputtering measurements [5] and erosion/redeposition analysis for liquid lithium shows that the impurity level is within reasonable limits [6].

Understanding the interactions between free surface flowing liquids and plasmas is hence indispensable in the development of advanced PFCs for future generation fusion machines. In addition, developing techniques that advance the technology and material selection for future fusion devices is crucial. To this end, a new facility, the flowing liquid-surface retention experiment (FLIRE) at the University of Illinois has been built. FLIRE is currently designed to study the interaction of energetic bombarding ions from an ion source with free surface flowing liquid lithium at moderate surface temperatures. This interaction includes such mechanisms as impurity transport, both by sputtering and evaporation, and more importantly, injected particle retention and release

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properties. These retention properties of flowing PFCs are critical to determine fuel recycling regimes (for the case of hydrogen isotopes) and ash removal/release rate (for the case of helium), among other things [6]. This work describes the experiment and presents the first helium retention data in molten lithium.

2. Experimental design

FLIRE consists of several subsystems that work together to ultimately measure fundamental properties such as particle retention/release properties from free surface flowing liquids exposed to high intensity ion beams or low-temperature plasmas. A global schematic of the experimental setup is presented in Fig. 1. The systems include vacuum components, a LM injection and storage system (LMISS), a heating system, an ion beam source, and a dual residual gas analyzer with quadrupole mass spectrometer (RGA-QMS).

The major vacuum components of FLIRE are shown in Fig. 1. The upper chamber consists of an Osaka TG2003 turbomolecular pump along with a CTI-8 cryogenic pump. The bottom chamber consists of a Seiko Seiki STP 300 turbomolecular pump and a CTI-6F cryogenic pump. Ultimate pressures in the high 10^{-9} Torr range are routinely achieved on both chambers.

Two cylindrical LM reservoir chambers connected in series below the bottom chamber are the heart of the LMISS, they are labeled upper and lower reservoir in

Fig. 1. Between the two LM reservoirs is a LM compatible inline valve used to transfer molten lithium between the two reservoirs when the whole lithium load is in the upper reservoir. Lithium is initially melted under argon atmosphere in an external side chamber; from there, it is transferred to the lower reservoir. The liquid is pumped using argon pressure on top of the liquid level, and it is transported through heated tubing. Once the lower reservoir is fully loaded, the metal is transported to the upper chamber and onto two heated internal ramps that guide the liquid to the bottom chamber. During lithium flow, both chambers are effectively isolated from each other by the flow itself. This unique feature makes the data analysis possible, since the signal from gas carried by the metal is completely separated from the background gas needed to operate the ion source.

In its current design, FLIRE uses an ion beam to inject helium (or other) ions at near normal incidence into the flowing stream of liquid lithium. The SPECS IQE 11/35 ion source provides current densities in the order of 0.5 mA/cm^2 . The operating pressure is typically in the 10^{-6} – 10^{-5} Torr range, and ion energies up to 5000 V can be handled. The gun can also be moved to change the location of the striking point along the path of the flowing metal.

Two quadrupole RGAs monitor the partial pressure of the incident particle species in the upper and bottom vacuum chambers. The partial pressure of the injected ion is measured in the bottom chamber, allowing for the

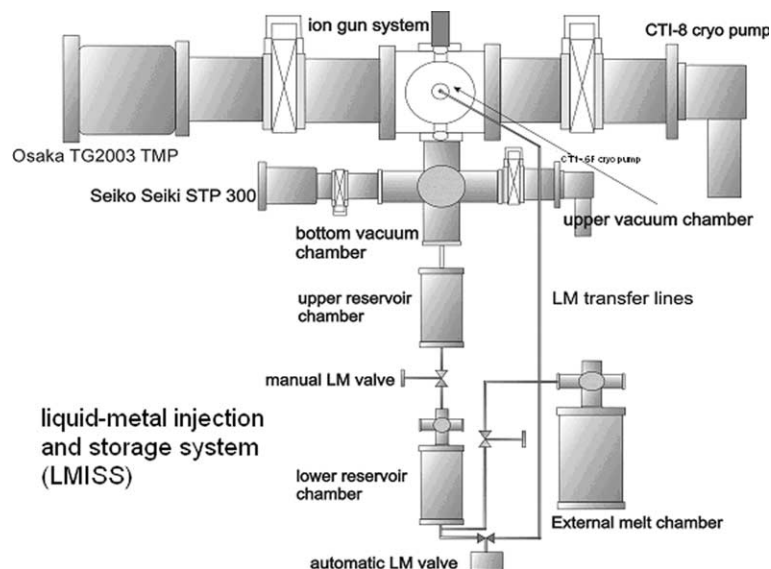


Fig. 1. FLIRE major vacuum components consist of two chambers, each with turbomolecular and cryogenic pumps and RGA-QMS systems (not shown). A LMISS is designed to inject liquid lithium at about 1 m/s flow velocities down heated ramps inside the upper vacuum chamber (not shown here).

calculation of retention characteristics. To improve the sensitivity of the RGA system, both instruments are equipped with channel electron multipliers that allow measurement of partial pressures as low as 10^{-12} Torr.

Fig. 2 shows an expanded schematic of the bottom/upper chamber interface during the experiment, as well the relevant dimensions that will be used for the data analysis. The flowing liquid lithium creates a vacuum seal at the opening between the chambers only when it is

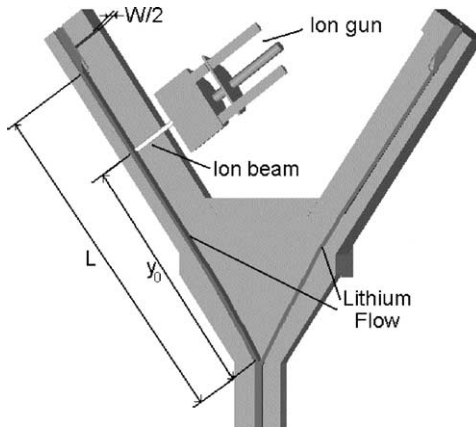


Fig. 2. View of the ramps of lithium flow inside the upper chamber. The total path of the lithium flow is L (16.9 cm) and width W of the ramp is 1 cm. The distance from the strike-point to the LM vacuum seal y_0 (10.4 cm) can be adjusted.

actually flowing down the plane. By having two flows meet at the opening, the surface layer is folded into the middle of the flow and must travel into the bottom vacuum chamber through the aperture. This concept was tested using liquid gallium on a mock-up ramp system with the same specifications as the FLIRE ramp system. After proper adjustment of the flow inlets, the experiment was a success in that the LM flowed smoothly and sealed the aperture as expected. The method used for heating the ramps employs very thin (0.25 mm diam.) tungsten wire shaped to deliver uniform heating, placed between thin (1.6 mm thick) alumina plates mounted to the underside of the ramps. Currents of 4 A (or ~ 7900 A/cm²) are passed through the heater wires to deliver 270 W of energy to the ramps, which raises the ramp temperatures from room temperature to about 400 °C.

3. Experimental determination of the diffusion and retention coefficients

Three different experiments were performed on FLIRE to measure the diffusion coefficient. Fig. 3 shows the helium traces in the lower chamber obtained for the three experiments, labeled (a), (b), and (c). During experiment (a) the upper chamber was filled with 1 mTorr of He, and a lithium flow at 250 °C with a velocity of 80 ± 20 cm/s was started. At that point, the He pressure in the lower chamber increased from 8×10^{-9} to

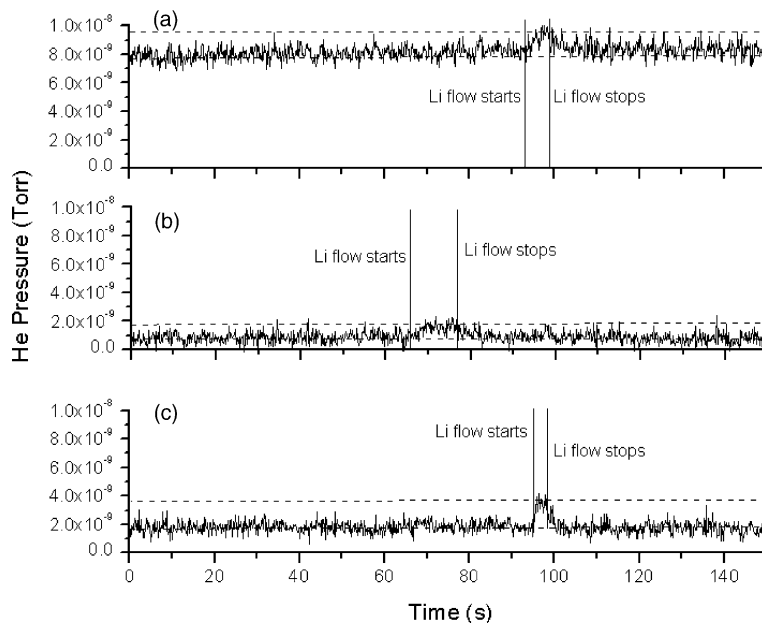


Fig. 3. He traces in the lower chamber for the three lithium flow experiments performed in FLIRE. See text for detailed explanation of each experiment. Vertical lines indicate start and end of the lithium flow on each experiment.

9.5×10^{-9} Torr (see Fig. 3(a)). The uncertainty in all partial pressure measurements is 0.5×10^{-9} Torr, since these measurements were made without turning on the channel electron multiplier on the RGA.

In experiment (b), the ion source was turned on, operating with a pressure of 2.7×10^{-5} Torr, ion energy of 1 keV and an ion current of 3×10^{-6} A. No partial pressure change in the lower chamber was detected due to the operation of the gun. A flow of lithium at 250 °C with velocity of 80 ± 20 cm/s was then run down the ramp for 10 s, and the helium partial pressure in the lower chamber rose to 1.5×10^{-9} Torr (see Fig. 3(b)). In experiment (c), the same irradiation conditions were kept, but the metal flow velocity was increased to 210 ± 70 cm/s, and this increased the pressure in the lower chamber to a value of 3.7×10^{-9} Torr (see Fig. 3(c)).

To obtain diffusion coefficient data, a model similar to that proposed by Hassanein [7] was used to interpret the data. The basic equation to describe the problem is

$$v \frac{\partial C}{\partial y} = D \frac{\partial^2 C}{\partial x^2} + G(x, y). \quad (1)$$

Here, v is the flow velocity, C is the particle concentration, G is the source term due to implantation and D is the diffusion coefficient. The x -coordinate is perpendicular to the flow direction, and y is parallel to the flow. For the case of experiment (a), the source term G vanishes. For experiments (b) and (c), the source term is given by:

$$G(x, y) = \frac{\phi}{W} \delta(x - r) \delta(y). \quad (2)$$

The ions are assumed to be implanted at the start of the path at a rate of ϕ ions per second at a range r . W is simply the width of the flow channel.

For experiment (a), the boundary conditions are

$$C(x = 0, y) = C_{\text{sat}}; \quad C(x = \infty, y) = 0; \quad C(y = 0, x) = 0. \quad (3)$$

To evaluate D from experiment (a), the solubility for helium in molten lithium at the conditions of the experiment (1 mTorr, 275 °C) is needed. Since that data is very scarce, an intermediate value (3×10^9 cm⁻³) between data extrapolated from the high pressure data in the literature [8] (3×10^7 cm⁻³) and the particle density at 1 mTorr (3×10^{13} cm⁻³) was selected. This assumption gives a D value of 3×10^{-3} cm²/s, but with an uncertainty of orders of magnitude.

For experiments (b) and (c) the solubility data is not as relevant since the pressure is much lower. The boundary conditions for these cases are

$$C(x = 0, y) = 0; \quad C(x = \infty, y) = 0; \quad C(y = 0, x) = 0. \quad (4)$$

The flow of particles into the lower chamber q , can be evaluated as

$$q = \int_0^\infty vC(x, y = y_0) dA = vW \int_0^\infty C(x, y = y_0) dx. \quad (5)$$

The expression for q is then obtained for these case:

$$q_{(a)} = C_{\text{sat}} \sqrt{\frac{4LDvW^2}{\pi}}, \quad (6a)$$

$$q_{(b),(c)} = \phi \text{erf} \left[\left(4 \frac{Dy_0}{vr^2} \right)^{-1/2} \right]. \quad (6b)$$

Here, y_0 is the distance between the striking point of the ions and the location where the two streams meet. In order to calculate D , the value of q must be measured. The relationship between q and the steady state pressure in the lower chamber P is

$$q = 3.25 \times 10^{16} PS. \quad (7)$$

S is the pumping speed in cm³/s. The pumping speed was intentionally kept low by gating the pumps mostly closed and was determined in a separate experiment to be 21.8 cm³/s. Since all the parameters are known, the diffusion coefficient D can be calculated by substituting the value of q given by Eq. (7) into Eqs. (6a) and (6b). Table 1 presents the values of all the parameters used to calculate D along with their estimated uncertainties, and Table 2 presents the obtained D values with its corresponding error.

Table 1
Parameters used for the calculation of D for experiments (b) and (c)

Parameter	Value
Pumping speed, S (cm ³ /s)	21.8 ± 10
Lithium path length from striking point, y_0 (cm)	10.4
Flow velocity, v (cm/s)	80 ± 20 (b), 215 ± 70 (c)
Lower chamber pressure, P (Torr)	9.5×10^{-9} (a), 1.9×10^{-9} (b), 3.5×10^{-9} (c)
	$\pm 0.5 \times 10^{-9}$
Implantation range, r (cm)	$4 \times 10^{-6} \pm 0.5 \times 10^{-6}$
Implanted ion current, ϕ (s ⁻¹)	$2 \times 10^{13} \pm 0.02 \times 10^{13}$

Table 2
Calculated diffusion coefficients for the two flow experiments with He implantation

Experiment	D (cm ² /s)
(b)	$5 \times 10^{-3} \pm 3.25 \times 10^{-3}$
(c)	$4 \times 10^{-3} \pm 2.5 \times 10^{-3}$
Average	$4.5 \times 10^{-3} \pm 2 \times 10^{-3}$

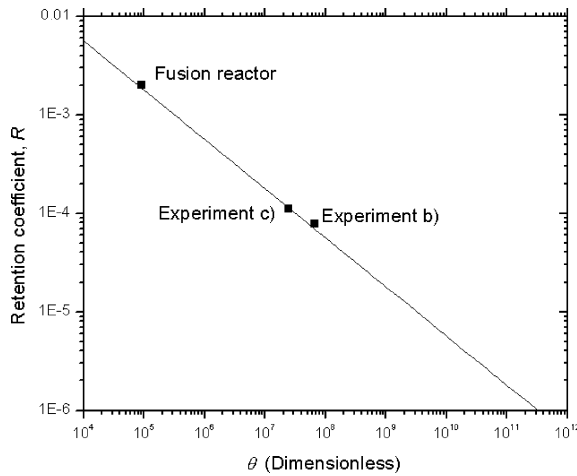


Fig. 4. Retention coefficient $R(q/\phi)$ as a function of the dimensionless parameter θ , defined as Dy_0/vr^2 . The two points corresponding to experiments (b) and (c) are marked on the figure. Operation conditions for a fusion reactor with low recycling are shown.

For experiments (b) and (c), an expression for the value q/ϕ can be obtained from Eq. (6b), which can be identified as the retention coefficient R , since it gives the ratio of transported to implanted particles. Another dimensionless parameter θ is defined as follows:

$$\theta = \frac{Dy_0}{vr^2}. \quad (8)$$

Physically, θ is the ratio of the microscopic diffusion time scale to the time it takes the lithium to flow from striking point to seal location. Upon substitution of these two dimensionless parameters, Eq. (6b) is transformed into a more compact form:

$$R = \operatorname{erf} \left[\frac{1}{\sqrt{4\theta}} \right]. \quad (9)$$

A plot of R vs. θ is presented in Fig. 4. Operational conditions for experiments (b) and (c) are indicated in the figure. A point for relevant fusion reactor conditions under a low recycling regime [7,9] ($y_0 = 50$ cm, $v = 1000$ cm/s, 10 keV He ions with $r = 5 \times 10^{-5}$ cm) is also shown in the plot. A retention coefficient of 2×10^{-3} is obtained, indicating that helium pumping by flowing liquid lithium may be possible.

4. Conclusions

With this work, FLIRE has demonstrated its potential to obtain critical measurements for gas retention

in flowing LM. For the ion implantation experiments, measurements of D were consistent for different operating conditions (experiments (b) and (c)). The large uncertainty in the solubility for experiment (a) made the estimation of the diffusion coefficient highly unreliable from that experiment. The calculated diffusion coefficient appears to be rather high, even above the self-diffusion coefficient for liquid Li, which is in the order of 10^{-4} cm²/s [10]. A possible explanation for this may be the presence of convective transport to the surface due to internal flows [11]. Steps will be taken to reduce uncertainties, especially in the flow velocity measurement. The electron multiplier will also be employed to reduce uncertainty in pressure measurements. Refinements in the analytical model will be required to account for the retention of atoms that are chemically active and require recombination at surfaces, such as hydrogen. Retention coefficient on the order of 10^{-2} – 10^{-4} indicates that He pumping is possible in future fusion devices with flowing Li divertors. Fig. 4 allows the calculation of retention for any given divertor length, lithium flow velocity and incident particle energy.

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