RETENTION/DIFFUSIVITY STUDIES IN FREE-SURFACE FLOWING LIQUID
LITHIUM

Final Report

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STTR Phase II Final Report

Project: Free Surface Flowing Liquid-Plasma Interaction Facility
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Summary

This report details work carried out in the Free Surface Flowing Liquid-Plasma Interaction Facility, which primarily houses the Flowing Liquid-surface Interaction Experiment (FLIRE), over the past two years. The report is organized in the following way: Phase II objectives for the first and second year are listed as a reference followed by a section on how these objectives have been achieved over the project period. A list of publications and conference presentations during this time period is included. An appendix with details of experimental and modeling work is included for reference.

FLIRE was designed to measure the hydrogen and helium retention and diffusivity in a flowing stream of liquid lithium, and it has accomplished these goals. Retention coefficients for helium in the flowing liquid stream were 0.1-2% for flow speeds of 44 cm/s and implantation energies between 500 and 2000 eV. The energy dependence of retention is linear for the energy range considered, as expected, and the dependence of retention on flow velocity fits the expected square-root of flow speed dependence. Estimates of the helium diffusion coefficient in the flowing lithium stream were $\sim 4 \times 10^{-7}$ cm²/s, and are independent of implantation energy. This value is much lower than expected, which could be due to several factors, such as mixing, bubble formation or surface film formation.

In the case of hydrogen, long term retention and release mechanisms are of greatest importance, since this relates to tritium inventory in flowing lithium PFCs for fusion applications. The amount of hydride formation was measured for flowing lithium exposed to neutral deuterium gas. Thermal desorption spectroscopy (TDS) measurements indicate that the hydride concentration was between 0.1 and 0.2% over a wide range of pressures (6.5×10^{-5} to 1 Torr). This result implies that the deuterium absorption rate is limited by the surface dissociation rate, since deuterium (hydrogen/tritium) is absorbed in its atomic form, not its molecular form.

Summary of FLIRE Successes

Helium retention and diffusion measurements are complete, and are outlined in the following paragraphs. Several difficulties with lithium handling, measurement techniques and data analysis were resolved over the course of this project. For example, the use of a turbo molecular pump (as opposed to a cryogenic pump) to assure a constant pumping speed during measurements. This allowed better (lower uncertainty) measurement of pumping speeds. Other improvements, such as the use of primarily iron gaskets, prevented vacuum leaks due to lithium dissolution of conventional copper gaskets and helped increase operating time and decrease lithium contamination. A new

method of measuring lithium flow rate, based on pressure evolution in the driving tank, improved measurement accuracy and made data analysis more efficient. These and other innovations allowed the successful completion of this Phase II STTR. Results from the measurements are outlined next.

- Helium Retention
 - Helium retention in flowing lithium was measured by FLIRE. The retention coefficient of helium in liquid lithium flowing at 44 cm/second down a 10-cm ramp is between 0.1% and 2%, for energies between 500eV and 2keV. The retention varies linearly with energy (as expected) over the range of energies considered, and the scaling of retention with flow velocity fits the expected square root of velocity expected by theory. This measure of retention is for helium that is quickly release from the flow (prompt release), which dominates the amount of retained helium; however, long-term trapping, which is much smaller, is also of interest.
- Long-Term Helium Retention
 - Long-term helium retention in liquid lithium was demonstrated by heating a sample of lithium after measuring its short-term helium retention. Above a threshold temperature of approximately 300°C, helium bubble bursts were detected, indicating that helium had been trapped in the liquid lithium, or on the surface within the measurement chamber. After successive thermal annealing cycles of the lithium charge, the bubbles ceased, indicating that the long-term retained helium was removed from the charge.
- Helium Diffusivity
 - A diffusion model of the helium in the flowing liquid allowed the helium diffusion coefficient in the flowing stream to be back-calculated from the measured retention coefficient. The result is a helium diffusion coefficient of $\sim 4 \times 10^{-7}$ cm²/s. This value is much lower than the expected value of $\sim 10^{-4}$ cm²/s, which may be due to mixing effects in the flow, bubble formation and/or surface film formation acting as a diffusion barrier.
- Hydride Formation
 - Thermal desorption spectroscopy (TDS) measurements indicate that deuterium is absorbed into the lithium flow directly from the background gas in the implantation chamber, even if no ion beam is present. To avoid problems with background hydrogen concentration, deuterium gas is used as a substitute. Two cases were examined: one with a high deuterium gas pressure (1 Torr) and one with a low deuterium gas pressure (5×10^{-5} Torr). In both cases, the TDS measurement indicates a hydride concentration of 0.1-0.2%. This suggests that the rate determining step for deuterium absorption is dissociation of deuterium molecules on the lithium surface, which absorbs deuterium atoms, not molecules. Because the background absorption rate is much higher than the ion current from an ion gun, a deuterium plasma source was built to expose the stream to more deuterium ions and atoms.
- MHD Capability

- MHD experiments were carried out on the FLIRE flow. Two electrical leads were placed in the path of the flow, and two permanent magnets were placed on either side of the flow ramp, forming a region where $\mathbf{J} \times \mathbf{B}$ forces on the flow could be generated and controlled by the current passing through the electrical leads. Both lifting and pressing forces were tested. The current required to exceed the gravitational force was calculated and applied to the flow, and the flow did not leave the channel. This result is likely due to two factors: 1) the high surface tension of the liquid lithium and wetting of the ramp surface provided an additional restoring force, 2) the flow “bulged” in the magnetic field region, spreading out the current density and lowering the body force on the fluid. This latter effect was not taken into account because the calculation of an equilibrium current density distribution and the resulting true body force was too complicated for the scope of the FLIRE project. The purpose of these experiments was to demonstrate FLIRE’s capability to carry out MHD studies with flowing liquid lithium, in addition to retention and diffusivity measurements.
- Modeling
 - The semianalytical model used to estimate the helium diffusion coefficient in the flowing lithium stream from retention data has been improved over the course of this project. More-detailed calculations, each focusing on a different assumption of the initial simple model, have shown that the initial model yields reasonable results when experimental error is considered. Specifically, new models have investigated the effect of a continuous implantation profile, a finite ion-beam width, finite flow depth, and pooling between the flow ramps. The results of each of these is discussed in detail in the Appendix. More detailed modeling work has been carried out using the HEIGHTs package at Argonne National Laboratory to investigate applications of flowing lithium to fusion reactor environments for this project.

Progress in Phase II research has been presented at several conferences and symposia. Research has also been published in refereed journals and is referred to for further information of technical details regarding work presented in this report (see publications section).

Phase II – Objectives

A list of Phase II objectives is shown below, together with updated objectives resulting from knowledge gained over the project period. These objectives reflect changes made to original objectives largely designed to address important issues related with diffusivity/retention measurements in FLIRE. The goals and objectives of the project are the following:

- He retention and diffusivity experiments
- H retention experiments.
- H/He desorption kinetics in liquid lithium with TDS/Purification chamber over a range of temperatures.
- H uptake and release modeling
- Coordinate with A. Hassanein (ANL) regarding plans for dc plasma experiments and modeling runs to be benchmarked against FLIRE experiments.
- Coordinate with A. Hassanein (ANL) regarding how data from FLIRE will be extrapolated to NSTX device for the possible installation of a liquid surface module (LSM).
- Prepare and deliver Final Phase II progress report to DOE.

How Goals and Objectives Have Been Met

Results and Accomplishments

- Retention coefficients for helium in liquid lithium have been quantitatively measured and behave as expected; retention goes up with increasing flow velocity and increasing beam energy. Difficulties with this measurement were resolved by reducing uncertainties in flow speed, better data analysis and a more consistent and easily measurable pumping speed.
- The diffusion coefficient of helium implanted in flowing liquid lithium has been estimated and behaves as expected, having no dependence on ion implantation energy. Diffusion coefficient estimates serve as an additional qualitative check of FLIRE data, namely, the estimated diffusion coefficient should be independent of energy and injected current.
- Single flow measurements have been carried out and compared to double-flow measurements at the same mass flow rate. These experiments confirm the expected square-root scaling of helium retention with flow speed. Comparing single-flow and double-flow results at the same mass flow rate, flow speed is changed without altering other parameters, effectively isolating the effect of changing flow speed. In previous tests with single and double flows flow-folding effects were not observed to change the measured retention coefficients.
- Deuterium absorption and hydride formation in flowing liquid lithium has been quantitatively measured. These measurements show that the rate of lithium deuteride (hydride) formation is determined by the surface dissociation rate of molecular deuterium, rather than by deuterium gas pressure in the pressure range of 10^{-5} -1 Torr.
- Modeling, used for analysis and interpretation of experimental results, has been significantly advanced, including estimates of the effects of a continuous implantation profile, finite beam width, finite flow thickness and pooling and diffusion between flow ramps. A DC plasma source was designed and built to provide a higher flux of deuterium ions and atomic deuterium to the lithium surface.

- Measurements of long-term helium retention have yielded important new results, indicating the formation of bubbles above a threshold temperature. These intermittent releases are attributed to helium trapped in voids in the lithium tank. More analysis is needed to determine this conclusively. Surface layer phenomena are another possible explanation for the apparent release of bursts of helium above a temperature threshold.
- A liquid level alarm system has been implemented to improve the reliability of the liquid metal injection system.
- A free-volume measurement system has been installed to allow more precise measurement of free volumes in the driving tank, which translates into better certainty of flow speed.
- Conventional copper gaskets have been replaced by annealed low-carbon steel gaskets in critical locations to prevent lithium-induced gasket failure (lithium dissolves copper).
- Coordination with A. Hassanein has continued; HEIGHTS modeling has been used to investigate the behavior of liquid lithium under ELM bombardment conditions in a fusion power reactor.
- Talks at various conferences and symposia have been given to inform the community of relevant results obtained in FLIRE.
- Several refereed papers have been written summarizing current measurements and analytical studies.

PUBLICATIONS: MEETING PRESENTATIONS AND JOURNALS

Meeting presentations and journal publications include:

- Allain, J.P., M. Nieto, M.D. Coventry, M.J. Neumann, E. Vargas-Lopes, D.N. Ruzic, "FLIRE—Flowing Liquid Surface Retention Experiment, Design and Testing", Fusion Engineering and Design, 61-62, 245-250 (2002).
- Talk on FLIRE progress at the ALPS semi-annual meeting in Princeton, New Jersey, November 2002.
- 44th Annual Meeting of the APS Division of Plasma Physics, November 11 – 15, 2002 in Orlando, FL. "Plasma-liquid metal interaction studies in FLIRE".
- 15th ANS Topical Meeting on the Technology of Fusion Energy, to be held November 17 - 21, 2002 in Washington, DC; "Experimental measurements of helium retention in flowing liquid lithium".
- Nieto M. Ruzic DN. Allain JP. "Experimental measurements of helium retention in flowing liquid lithium". Fusion Science & Technology, 44:1, 232-236 (2003).
- M. Nieto, D.N. Ruzic, J.P. Allain, M.D. Coventry, E. Vargas-Lopez, J. Nucl. Mater. 313-316 (2003) 646-650.
- Talk on FLIRE progress at the ALPS semi-annual meeting at Grand Canyon, Arizona, April 2003.
- D. Ruzic, J.P. Allain, D.A. Alman, M. Nieto, M.D. Coventry, T. Qiu, W. Olczak, R. Stubbers, "Plasma Material Interactions of Carbon and Liquid Lithium:

- Experiments and Modeling,” 30th EPS Conference on Controlled Fusion and Plasma Physics, St Petersburg, Russia, July 2003.
- Poster presentation for the 30th EPS Conference on Controlled Fusion and Plasma Physics, in St Petersburg, Russia, July 2003.
 - Presentation of FLIRE Progress at the 2003 US-Japan Joint Workshop on High Heat Flux Components and Plasma Surface Interaction for Next Fusion Devices & Fusion High Power Density Devices and Design at Port Townsend, Washington, July 2003.
 - J.P. Allain, M. Nieto, M.D. Coventry, R. Stubbers and D.N. Ruzic “Studies of Liquid-Metal Erosion and Free Surface Flowing Liquid-Metal Retention and Diffusivity at the University of Illinois” Fusion Engineering and Design, submitted August 2003.
 - J.P. Allain, G.H. Miley, M. Nieto, et al, “Free Surface Flowing Liquid-Plasma Interaction Facility,” Champaign-Urbana Technology Show, September 2003.
 - M. Nieto, W. Olczak, R. Stubbers and D.N. Ruzic, “Experimental measurements of He and H retention on liquid lithium on FLIRE,” 20th IEEE/NPSS Symposium on Fusion Energy, San Diego, CA October 14 – 17, 2003.
 - Poster: W. Olczak, M. Nieto, R. Stubbers, D. N. Ruzic, “Thermal Desorption Spectroscopy Studies of Hydrogen Retention on FLIRE,” 45th Annual Meeting of the APS Division of Plasma Physics, Albuquerque, New Mexico, October 2003.
 - Poster: M. Nieto, W. Olczak, R. Stubbers, D. N. Ruzic, “Particle Retention and Diffusion Measurements in FLIRE,” 45th Annual Meeting of the APS Division of Plasma Physics, Albuquerque, New Mexico, October 2003.
 - Several presentations on FLIRE progress at the ALPS semi-annual meeting at Oak Brook, Illinois, November 2003.
 - M. Nieto, “Study of Plasma-Liquid Lithium Flow Interactions in the Flowing Liquid Retention Experiment (FLIRE),” NPPE Ph.D. thesis, University of Illinois, Defended April 2004.
 - Presentations on FLIRE progress at the ALPS semi-annual meeting at Urbana, Illinois, May 2004.
 - D.N. Ruzic, M. Nieto, M.D. Coventry, W. Olczak, H. Qiu, R. Stubbers, “Plasma Liquid-Metal Interaction Experiments and Modelling,” 31st EPS Conference on Controlled Fusion and Plasma Physics, London, United Kingdom, June 2004.
 - Nieto, M., D.N. Ruzic, W. Olczak, R. Stubbers, “Measurement of Implanted Helium Particle Transport by a Flowing Lithium Film”, submitted to J. Nucl. Mater., August 2004.
 - Poster: D. N. Ruzic, M. Nieto, W. Olczak, R. Stubbers, “Measurement of helium retention and diffusivity and hydrogen absorption in flowing liquid lithium in the Flowing Lithium Retention Experiment (FLIRE),” Plasma Surface Interactions Conference, Portland, Maine, September 2004.
 - R. Stubbers, W. Olczak, M. Nieto, D. N. Ruzic, “Measurement of hydrogen absorption in flowing liquid lithium in the flowing lithium retention experiment (FLIRE),” Journal of Nuclear Materials, Submitted September 2004.
 - Poster: R. Stubbers, M. Nieto, W. Olczak, D.N. Ruzic, “The Flowing Liquid-Metal Retention Experiment (FLIRE) Results,” TOFE16, Madison, Wisconsin, September 2004.

- Poster: W. Olczak, R. Stubbers, D. N. Ruzic, “Thermal Desorption Spectroscopy Studies of Hydrogen Retention on FLIRE,” 46th Annual Meeting of the APS Division of Plasma Physics, Savannah, Georgia, November 2004.
- Invited Talk: D.N. Ruzic, “Hydrogen Retention and Helium Pumping by Flowing Liquid Lithium: A Path to a Compact (and therefore less costly) Magnetic Fusion Power Reactor”, 46th Annual Meeting of the APS Division of Plasma Physics, Savannah, George, November 2004.

During the last three years many graduate and undergraduate students have participated in the NPL FLIRE research. Notable are two PhD theses in this field:

Darren Alman	2003
Martin Nieto	2004

APPENDIX

A Background

Plasma-facing components (PFCs) on tokamak fusion reactors are exposed to a considerable amount of energy flux. Typical reactor designs consider an average heat load to the wall close to 2 MW/m^2 , but the ratio of peak to average thermal load is on the order of 50, and the peak occurs at the divertor plate¹. The heat load to internal components increases even more in the case of off-normal events, such as vertical displacements, edge-localized modes and disruptions^{2,3,4}. Under these power fluxes, most materials would vaporize, and the particles coming from the divertor plate may find their way into the plasma core. This would cause radiative losses and fuel dilution, which both increase as Z^2 , effectively reducing the fusion reaction rate. Hence, low atomic number impurities are preferable.

Most tokamaks currently in operation use solid divertor plates, typically made of graphite⁵ or tungsten⁶, which are materials that can withstand normal tokamak operating conditions. However, there are problems currently associated with the use of these solid materials:

- Solid divertors have very large exposure time to the plasma, they accumulate damage that may lead to ultimate failure after a certain period of time,
- Graphite divertors get eroded more rapidly than their metallic counterparts due to the chemical sputtering arising from the exposure to hydrogen plasmas⁷,
- Tungsten, while having a very low sputtering yield for hydrogen, has a large atomic number, which makes it an extremely undesirable impurity in the plasma since it can be sputtered by C or O readily and has a high self-sputtering coefficient. In addition, it is hard to couple with other metals that serve as heat sinks such as copper⁸.

In order to try to mitigate some of these problems, the use of materials that are already molten has been proposed. The use of liquids as divertor materials has some advantages over the use of solid components:

- (1) The exposure time can be adjusted by the use of a flowing film or jet,
- (2) Energy extraction can be done directly from the liquid outside the reactor,
- (3) The component is not permanently damaged,
- (4) Fuel trapped on the component can be extracted outside the reactor for reinjection.

There are, however, uncertainties in the use of liquid divertors. The effects of electric and magnetic field configurations on the flow of conducting liquids are not very well known. Fuel (hydrogen) and ash (helium) intake by the liquid may modify operation regimes of

the reactor. Sputtering yield from the liquid may increase due to the increased temperature, enhancing impurity transport to the plasma⁹.

Some of the liquids considered for divertor operation include molten metals and alloys, such as lithium¹⁰, tin and tin-lithium alloys^{11,12}, and gallium¹³; molten salts, such as FLiBE¹⁴, have also been proposed as liquid divertor materials. Lithium is one of the leading candidates for liquid PFCs in tokamak reactors, due to its low atomic number, excellent thermal properties, its self-sputtering being less than 1, high sputtered ion fraction (2/3), and its ability to getter hydrogen to operate the reactor in a very low-recycling mode.

The Flowing Liquid-surface Interaction Retention Experiment (FLIRE) facility at the University of Illinois at Urbana-Champaign is aimed to help resolve one of the questions regarding liquid PFCs mentioned above; that is, the problem of quantifying hydrogen and helium transport when the liquid has been bombarded with energetic ions of such gases, as it would occur in the divertor of a real fusion reactor. Hydrogen transport is important in determining fuel recycling conditions, while helium transport is crucial for ash removal from the reactor volume. Initially, an ion beam was used to provide controlled exposure of liquid lithium to a helium ion beam, allowing the quantitative measurement of helium retention and diffusivity in the liquid stream. Following helium retention and diffusivity measurements, the lithium was exposed to deuterium (an easily distinguishable hydrogen isotope), and the amount of lithium deuteride formation was measured using thermal desorption spectroscopy (TDS).

B FLIRE Experimental Setup

The FLIRE facility was designed to allow the measurement of particles retained by a flowing stream. Figure B.1 shows a diagram of FLIRE. Two vacuum chambers, one on top of the other, are independently pumped and connected by a 3mm×10 mm rectangular slit, as shown in Figure B.2. The passage connecting the two chambers becomes completely plugged when the liquid lithium flows between the two chambers and remains plugged even after the lithium flow stops, forming a vacuum seal between the two chambers that allows measurement in the lower chamber without interference from the gas in the upper chamber. In the top chamber, the flow can be exposed to a helium ion gun which usually requires backfilling with 10^{-5} Torr of neutral gas to operate. Alternatively, the flow can be exposed to deuterium gas and/or deuterium plasma, as is the case for studying hydrogen uptake in the flowing liquid stream. The bottom chamber is furnished with a magnetic sector residual gas analyzer (RGA) capable of detecting partial pressures as low as 10^{-14} Torr. For measuring hydride concentration, the flow can be directed to the TDS chamber, where the liquid sample is slowly heated (2°C/minute) and monitored with the RGA. As the sample is heated a deuterium peak will occur at a particular temperature that corresponds to the concentration of lithium deuteride in the sample, thus providing a quantitative measure of the amount of lithium deuteride formed by exposing the flowing stream to deuterium.

Figure B.1. Block diagram of the FLIRE main components.

Figure B.2. The basic setup of FLIRE, showing the upper and lower chambers separated by a small passage. The passage gets plugged by the Li flow, allowing the RGA to measure only particles carried by the metal flow. Flow direction is indicated by the small arrows.

When a good coverage of the passage is achieved during the lithium flow, only the helium being desorbed from the flowing metal can be detected in the lower chamber. To test if this condition could be achieved, experiments with the neutral pressure required for operation of the ion gun in the upper chamber were performed with the beam turned off.

The FLIRE upper chamber was filled with 10^{-5} - 10^{-4} Torr of helium, and the lithium flowed in the upper chamber for 40 – 50 seconds, with the ion source off, so that He particles were not implanted. The helium signal in the lower chamber for this experiment is shown in Figure B.3. It can be seen that the helium signal stays at background level while the lithium is flowing. This means that the chambers are adequately isolated from one another, since the neutral pressure in the upper chamber does not affect the signal in the lower chamber. The chambers remain isolated from each other even after the run is over, since the passage between the chambers is covered by lithium held there by surface tension.

Figure B.3. Helium pressure trace in the lower chamber for an experiment where the ion gun was not turned on. Flow velocity is 70 cm/s, and the flow lasts 45 seconds.

Figure B.4 shows the trace of helium in the lower chamber when a 1.5 keV, 1 μ A ion beam is incident on lithium flowing at an average velocity of 50 cm/s. Unlike Figure B.3, the helium signal in the lower chamber rises, but only when the ion gun is on, indicating that implantation is required in order to transport the helium and that the release in general is prompt. Vertical lines mark the start and the end of the flow, and the rise and fall of the helium signal is concurrent with the beginning and the end of the flow, respectively. Pressure values shown in the figures and used in the calculation of the retention coefficient have been corrected for the difference in temperature at the ion gage and for the ionization efficiency of He vs the N₂ calibration for the RGA.

Figure B.4. Helium trace observed in the lower chamber when the lithium flow is bombarded by a beam with an energy of 1.5 keV.

The next step was to measure the retained fraction with the gun on, which is discussed in the following section. The retained fraction is dimensionless and must be related to the ratio of timescales. One time scale is the average diffusion time, related to the implantation range and the diffusion coefficient. The other time scale is the transit time between the point of implantation and the exit from the upper chamber. This time scale is related to the flow velocity and the path length between the implantation point and the chamber exit. Hence, four parameters are key to determine the retained fraction:

- (1) Flow velocity (faster flow means particles have less time to escape from the material, so the retained fraction increases with increased velocity),
- (2) Path length in the upper chamber (shorter path length gives implanted particles less time to escape, so the retained fraction increases with decreased path length,
- (3) Diffusion coefficient (fast diffusing particles escape the material easily, so retained fraction increases with decreased diffusion coefficient),
- (4) Implantation range (deeper implantation means particles take longer to reach the surface, so retention fraction goes up with range).

C Helium Campaign in FLIRE

C.1 Helium Ion Beam Experiments

The amount of helium retained in the flowing metal can be directly measured in FLIRE. The retention coefficient R is defined as the ratio of the release rate in the lower chamber to the injection rate in the upper chamber, as shown in Equation C.1. From Equations C.1 and C.2, it can be seen that knowledge of the parameters j , q , and t can be used to estimate the diffusion coefficient D . Measurements of ion current from the ion gun

determine the value of j , and t is measured based on changes in driving pressure which yields the flow rate. The range of helium ions, r , is a known function of energy. The only parameter left to be determined is the injection rate into the lower chamber $q(t)$.

$$R = \frac{q}{j} \quad (\text{C.1})$$

$$q(t) = j \cdot \text{erf}\left(\frac{r}{\sqrt{4Dt}}\right) \quad (\text{C.2})$$

Here, S is the volumetric pumping speed in the chamber in, say, cm^3/s , P_{He} is the He partial pressure in the chamber, k is the Boltzman constant and T is the temperature of the helium gas. For the case of constant pumping speed and constant release rate, the system may reach a steady state where the release rate balances the pumping rate. Hence:

$$q_{\text{obs}} = \frac{P_{\text{He}} S}{kT} \quad (\text{C.3})$$

From Equation (C.3) it can be seen that three factors affect the measurement of the desorption rate: the pumping speed, the partial pressure of helium and the temperature of the helium gas. Of these three parameters, the ones that can be measured with accuracy are the pressure and the pumping speed. For temperature, it can be assumed that the helium gas is at a temperature between the chamber temperature and the temperature of the liquid metal. These two temperatures differ greatly; however, due to the low pressure in the chamber, it is believed that the temperature of the He gas is closer to the temperature of the metal than the surrounding chamber.

Of the parameters present in equation Equation (C.3), only S (and T to a certain extent) can be varied during the experiment. The value of S will determine the equilibrium pressure for a given injection rate in isothermal conditions. For a given injection rate, an increase in S will cause a decrease in steady state pressure P .

In order to find an absolute pumping speed, suppose that the injection rate disappears instantaneously. The system no longer in steady state, and the removal rate causes the pressure to decay to zero or a background value. The evolution of the pressure is given by:

$$P(t) = P_{\text{bckg}} + P_0 \cdot e^{-\frac{St}{V}} \quad (\text{C.4})$$

Figure C.1 presents the evolution of the pressure when the injection rate is removed. The exponential decay of Equation A.4 fits the observed data, and the pumping speed can be calculated from the decay constant provided the volume being pumped out is known. For the data in Figure C.1, the ratio V/S is 2 ± 0.03 s, so knowing that the volume being pumped is close to 8 liters, the pumping speed turns out to be 4 ± 0.12 liters/second.

Figure C.1. Exponential decay of pressure once the small He leak is removed.

Verification that a steady-state level can be achieved during the measurement is critical in order to measure release rate in the lower chamber, and hence determine the retention coefficient experimentally. In Figure C.2, the lithium stream in FLIRE running at 40 cm/s was bombarded with 1.5 keV He ions. The He signal in the lower chamber starts rising as soon as the flow starts, and reaches a steady state during the 20 seconds that the flow lasted. This allows the calculation of the release rate in the lower chamber from Equation C.3, and hence the retention coefficient from Equation C.1, if the incident current j , the pumping speed S and the temperature of the gas T are known. It should be noted that helium transport was not observed when the ion source was not turned on, even though the upper chamber was filled with He gas necessary for the source operation ($\sim 10^{-5}$ Torr). This means that the only He detected comes from energetic He ions implanted into the stream, and the neutral gas necessary for ion source operation does not interfere with the measurement.

Figure C.2. Helium trace in the lower chamber during a FLIRE run. The average flow velocity was 44 cm/s, and the energy of the ions was 2000 eV. Flow start and end points are marked in the figure by vertical lines.

From Equations C.1 and C.2, it can be seen that increasing the energy of the bombarding ions should increase the retention in a linear fashion, provided the mean range is linear with energy (which is the case, as is discussed in Section C.4). Hence, a linear increase with energy should be observed in the release rate measured in the lower chamber. If constant pumping, temperature and ion flux conditions are maintained for all the experiments, this means that the equilibrium pressure during the run should increase linearly as the energy of the ions is increased. Two sets of experiments were performed in FLIRE varying the energy of the beam between 500 and 4000 eV, one set with an electron emission current of 5 mA on the ion gun (set A), and another set with such current increased to 7.5 mA (set B), with at least two runs were performed for each energy in order to improve the statistics. All other experimental parameters were kept constant for both sets. Figure C.3 shows the relationship between the directly measured filament current and the injected ion current.

Figure C.3. Helium ion current as a function of energy for different electron emission currents in the gun filament. Data is corrected for secondary electron emission.

Pumping speed was reduced during the experiment by closing the cryopump gate valve completely and partially closing the turbopump gate valve. Pumping curves were obtained after taking each data set to account for small mismatches in the position of the valve. As can be seen from the exponential fits to the data in Figure C.4, the values of the time constant for the exponential pressure decay for data sets A and B are very similar: 16.5 ± 2 s for set A, and 21 ± 1 s for set B. Knowing that the volume in the lower chamber is 7 ± 0.25 liters, Equation C.4 allows the pumping speed to be found: 0.42 ± 0.06 l/s for set A and 0.33 ± 0.02 l/s for set B.

a)

b)

Figure C.4. Pumping curves for a) set A and b) set B. Time constant for the pressure exponential decay (V/S) is 16.5 ± 2 and 21 ± 1 seconds, respectively. Small mismatch in valve aperture may account for the difference.

Figure C.5 shows the helium pressure in the lower chamber as a function of energy for data sets A and B. The value of the pressure was obtained by averaging the RGA He mass signal over the length of the run. The error is obtained from the standard deviation of the RGA signal during the experiment. Notice that, as predicted by Equation C.2, the average pressure (which is proportional to the retention coefficient R) increases linearly with energy (which is proportional to the implanted particle range r).

Figure C.5. Average He pressure in the FLIRE lower chamber as a function of ion energy for a) the low current and b) the high current data sets. Pressure increases linearly with energy.

In order to make more direct comparisons, the data in Figure C.5 can be multiplied by the corresponding pumping speed and other appropriate constants in order to obtain a desorption rate from the liquid metal. For both sets of experiments the temperature of the lithium was 250 ± 10 °C, and the pumping speed for each set was determined from the pumpout data of Figure C.4. The results of this operation are shown in Figure C.6, where the desorption rate calculated from Equation C.3 is plotted as a function of energy. Fits to a straight line passing through the origin are shown on the graph also, and the values for the slope of each line are shown. Since the only difference between the two data sets is the ion current, the ratio of the slopes should be equal to the ratio of ion currents for both sets. Using Figure C.3, the ratio of ion current is $2/3$ (which also happens to be the ratio of filament emission currents for both sets). The ratio of the slopes in Figure A.6 is 0.6 ± 0.03 , which is very close to $2/3$. This demonstrates that the retention is independent of incident ion current: if the ion current increases twofold, then the desorption rate observed in the lower chamber doubles too. Upon dividing the data on Figure C.6 by the particle injection rate (i.e. ion current), it can be observed that both sets give a straight line with identical slopes within the error margin for the linear fit (see Figure C.7).

Figure C.6. Desorption rate as a function of energy for the two sets of experiments. The ratio of slopes is close to 2/3, which is close to the ratio of ion current for the two sets.

Figure C.7. Retention coefficient as a function of energy for the two data sets. Despite differences in ion current used on the two sets, retention values are very close for both of them, proving that retention is independent of flux in this case.

The other parameter that can be modified easily in FLIRE is the flow velocity. The easiest way to increase it is to have a higher driving pressure in the supply tank. However, increasing the velocity increases the mass flow rate and the chances of developing a pool between the chambers increase. So the question is: is there a way to significantly increase the flow velocity without increasing the volumetric flow rate? And the answer is yes.

The volumetric flow rate is a function of the driving pressure only. The volumetric flow rate splits into two currents such that half of the flow goes on each ramp. If the flow rate is constant, the immediate answer would be that the flow in the ramp with flow would double, compared to the flow rate that it would get if the other ramp was not blocked. If the thickness of the flowing film does not increase considerably, then the flow velocity has to double in order to get the same flow rate. This fact allows to increase the flow velocity significantly without having to increase the flow rate. The only downside of this method is that for the single ramp flow the seal between the two chambers not always develops, so some of the runs with a bad seal need to be discarded.

Figure C.8 presents results obtained for two sets of experiments with identical conditions. The only difference is that one set had lithium flowing in both channels, while the other had all the flow going through one channel only. Again, the linear increase of the plateau pressure with energy is observed, and since both sets were taken with the same pumping speed there is no need to convert to molecule flux in order to compare the two sets. If all the parameters but the velocity are equal, then from Equation C.2 the following relationship can be obtained:

$$\frac{m_1}{m_2} = \sqrt{\frac{v_1}{v_2}} \quad (\text{C.5})$$

Figure C.8. The steady state pressure values for experiments with single and double flow.

Here, m is the slope of each straight line in Figure C.8, and v is the corresponding velocity. Since one set of data is for single flow and the other one for double flow, the ratio of velocities of single to double flow should be approximately 2. The slopes obtained from a linear fit are $(2.2 \pm 0.1) \times 10^{13}$ for the single flow, and $(1.4 \pm 0.1) \times 10^{13}$ for the double flow. The ratio of slopes is then 1.57 ± 0.13 ; compare to the square root of the velocity ratio, which is 1.414, and the ratio of the slopes is very close. So the dependence of retention with velocity is confirmed.

C.2 Long-term trapping of He

Up to this point, it has been assumed that the observed release rate was equal to the rate at which implanted He enters the lower chamber. It is reasonable to raise the question about the validity of this assumption, so it is important to look at the possible mechanisms for helium desorption in the lower chamber

Two extreme cases can arise as the flow enters in the lower chamber, shown in Figure C.9. One case is to assume that the two metal streams converging at the exit of the upper chamber undergo no mixing during their transit to the lower chamber. The result of that assumption is that most of the material gets “buried” in between the two layers, and needs to travel a significant distance before it can come out. The opposite case is that of “perfect mixing”. In that case, the helium that remains inside the metal when the two streams meet spreads homogeneously through the metal, giving a stream with homogeneous concentration entering the lower chamber.

Figure C.9. The two extreme cases of helium distribution in the lithium, upon entrance into the lower chamber.

A model to describe the helium desorption from the lithium entering the chamber is a finite domain of width L (equal to $2x_2$, from Figure C.9), with an initial concentration profile C_0 :

$$\begin{aligned}
 \frac{\partial C}{\partial t} &= D \frac{\partial^2 C}{\partial x^2} \\
 C(x, 0) &= C_0(x) \\
 C(0, t) &= 0 \\
 C(L, t) &= 0
 \end{aligned}
 \tag{C.6}$$

The following dimensionless variables can be defined:

$$\begin{aligned}
 \tau &= \frac{Dt}{L^2} \\
 \xi &= \frac{x}{L}
 \end{aligned}
 \tag{C.7}$$

Then the problem in Equation C.6 can be re-written as follows:

$$\begin{aligned}
 \frac{\partial C}{\partial \tau} &= \frac{\partial^2 C}{\partial \xi^2} \\
 C(\xi, 0) &= C_0(\xi) \\
 C(0, \tau) &= 0 \\
 C(1, \tau) &= 0
 \end{aligned}
 \tag{C.8}$$

By using separation of variables on the problem, the following series solution is obtained:

$$C(\xi, \tau) = \sum_{n=1}^{\infty} A_n e^{-k_n^2 \tau} \sin(k_n \xi) \quad (\text{C.9})$$

The constants A_n and k_n in Equation C.9 are defined as:

$$k_n = n\pi$$

$$A_n = \int_0^1 C'_0(\xi) \sin(k_n \xi) d\xi \quad (\text{C.10})$$

Two forms for the function $C'_0(\xi)$, representing the two limiting cases mentioned earlier, are used to obtain the evolution of the concentration profile evolutions. The first is a delta function in the center of the sample ($\xi=0.5$), representing no He mixing due to the convergence of the two streams. The other is a flat profile ($C'_0=1$) representing perfect mixing during the transit between upper and lower chamber. The ultimate goal is to determine how the helium desorbs from the metal given these two initial conditions. Figure C.10 presents the evolution of the concentration profile in the variables ξ and τ .

Figure C.10. Concentration profile evolution in a slab domain with initial conditions given by (a) homogeneous concentration, and (b) Delta function at $\xi=0.5$

A measure of the amount of material at any certain time is the retained fraction α , which is a function of τ , defined as:

$$\alpha(\tau) = \frac{\int_0^1 C(\xi, \tau) d\xi}{\int_0^1 C'_0(\xi) d\xi} \quad (\text{C.11})$$

In the two cases treated here, the integral in the denominator of Equation C.11 evaluates to 1, so α is simply the solution integrated over the ξ variable:

$$\alpha(\tau) = \sum_{n=1}^{\infty} e^{-k_n^2 \tau} \int_0^1 A_n \sin(k_n \xi) d\xi \quad (\text{C.12})$$

For the case of the delta function initial condition, Equation C.12 yields:

$$\alpha(\tau) = \sum_{n=1}^{\infty} e^{-k_n^2 \tau} \frac{\sin(0.5k_n)}{k_n} [1 - \cos(k_n)] \quad (\text{C.13})$$

For the case of the homogeneous profile, the result is:

$$\alpha(\tau) = \sum_{n=1}^{\infty} e^{-k_n^2 \tau} \frac{[1 - \cos(k_n)]^2}{k_n^2} \quad (\text{C.14})$$

Figure C.11 presents a comparison of α obtained with Equations C.13 and C.14. Notice that for the case of the delta function initial condition, the concentration stays constant up to $\tau=0.05$, indicating that prompt release (1 second or less) does not happen unless D/L^2 is greater than 0.1.

Figure C.11. The total amount retained α as a function of dimensionless variable τ (see Eqn C.7) for the two different initial condition cases

A good measure of the speed of release is the gradient $dC/d\xi$ evaluated at $\xi=0$:

$$\left. \frac{\partial C}{\partial \xi} \right|_{\xi=0} = \sum_{n=1}^{\infty} k_n A_n e^{-k_n^2 \tau} \quad (\text{C.15})$$

Figure C.12 presents the values of the surface ($\xi=0$) gradient as a function of τ for the two cases considered. From the plot it is confirmed that the delta function case has a delayed release, while for the homogeneous profile case the release is stronger at the beginning and starts decreasing fast after the initial burst. Since experimental observations seem to suggest that release is prompt, the homogeneous concentration profile is the most likely situation. However, depending on the values of L and D , some long-term trapping of helium might be possible

Figure C.12. Evolution of the flux coming out of the surface as a function of dimensionless variable τ (see Eqn C.7).

From Figure C.2, it can be seen that the rise in He partial pressure in the lower chamber is concurrent with the flow, strongly suggesting that something closer to the mixing case might be more feasible, since the no mixing case has a delay. Assuming values for $D = 10^{-4} \text{ cm}^2/\text{s}$ and $L=0.5 \text{ cm}$, desorption of He does not occur until 25 seconds after exit from the main chamber, which doesn't match with the observed results at all, i.e. prompt release is observed in the experiment.

In order to experimentally verify the possibility of He long-term release, two experiments were performed in FLIRE. An increase in the sample temperature should accelerate desorption of the sample, so an increase in the helium release should be observed if there

is any helium left inside the lithium batch. Constant heating rate can not be achieved in this part of the machine, so the increase in temperature is not linear. After being bombarded with the He beam, the batch was heated in the lower chamber up to 350 °C over 20 minutes. It is left soaking at that temperature for about 10 to 15 minutes, and then cools down to 235 °C, which is the normal operating temperature of the upper reservoir tank, where the lithium is sitting.

An initial run was done, where the lithium was not bombarded with the ion beam, but the main chamber was backfilled with the typical operating He pressure ($\sim 2 \times 10^{-5}$ Torr). The traces of helium and lithium partial pressures during the heating are shown in Figure C.13 and Figure C.14. A completely unexpected result is observed: a series of bursts in He pressure are observed during heating, and their frequency increases when the temperature is high. It should be noted that this batch had been extensively bombarded with helium during the measurements of prompt release shown in the previous section. Could this be residual helium from all those previous runs? Is it coming solely from the exposure to He gas in the main chamber? A second experiment, keeping all experimental conditions the same (flow velocity, He gas pressure, duration of the run), was performed to see these observations were repeatable.

Figure C.13. He and Li traces during heating of the Li batch after exposure to He gas. No prompt release is observed in this case since the ion beam was not turned on.

Figure C.14. A detail of Figure A.13 showing the evidence of helium bubbles coming out once the sample is heated.

The two traces are shown in Figure C.15, and yet another interesting observation can be made. The temperature onset for bubble release remains the same; however, the strength of the signals from the bubbles weakens prematurely, compared to the previous runs. The bubbles get weaker and weaker until they disappear. This seems to suggest that all the He observed in these two experiments was probably residual helium from previous runs. On the first experiment the soak time is not long enough to get rid of all the lithium. The combined heating of both first and second heating cycles seems to eliminate the residual helium. The analysis of nucleation kinetics and its relationship to mobility inside the metal (i.e. diffusion) are another whole subject that is beyond the scope of this work. Average bubble size can be obtained from the strength of the pressure spike after bubble release, and activation parameters for the nucleation process can be found based on the temperature threshold for bubble release observations. An excellent review on the current knowledge of helium accumulation in metals can be found in Trinkaus and Singh¹⁵. Most of the work on this area has been done on solid metals, where defects and dislocations play an important role in the bubble nucleation and growth dynamics. Very little is known about nucleation of He bubbles in molten metals, so more extensive studies regarding He bubble formation and release at low concentrations are required.

Figure C.15. He and Li traces during the second heating of the Li batch after exposure to He gas.

To obtain a relative measurement of the helium left behind in the lithium after a single irradiation run, the thermal annealing treatment from the two previous experiments was performed on the lithium charge right after being bombarded with a 3 keV, 3 μ A He ion beam. The pressure trace for this experiment is shown in Figure C.16, and again release of bubbles can be seen, but the size has reduced considerably compared to the two previous experiments. This is definite proof that long-term retention of He in lithium does happen. It can be seen that the amount released promptly is large, compared to the bubble size and frequency during the late release. However, quantification might be hard due to the discrete (i.e. bubble) release from the sample upon annealing, since the model for long term release discussed in this section does not include nucleation of bubbles and would not apply.

Figure C.16. Helium release from the Li sample irradiated with a 3 keV He beam. Flow starts at 1 minute and lasts 50 seconds. Thermal annealing starts right after the axis break (three minutes). Two time scales have been used in order to show both the prompt continuous release and the delayed release upon thermal annealing.

C.3 Simple Model for Retention (diffusion controlled)

The FLIRE system can be described as a starting point by a simple model where diffusion transport from the bulk dominates the He release. Consider the system depicted in Figure C.17, with the x direction normal to the flow and the y direction parallel to the flow. All the ions are implanted at $y=0$ at a rate j , and all of them are implanted at a distance $x=r$, where r is the mean implantation range. At $y=L$, the two flows converge and the He retained up to that point is assumed to be transported into the lower chamber. It will also be assumed that the lithium stream is on laminar flow and that the dominant transport mechanism for the x direction is diffusion, while for the y direction the transport is purely convective. By making these assumptions, the equation describing the concentration in the flow simplifies to the following:

$$v \frac{\partial n}{\partial y} = D \frac{\partial^2 n}{\partial x^2} + G(x, y) \quad (\text{C.16})$$

Here, v is the flow velocity, n is the particle density in the material, D is the diffusion coefficient and G is the source term. Since helium is completely inert and not soluble,

there are no volumetric sinks, only the source term associated with the implantation. No net flux from the gas to the liquid phase is present since He is not soluble.

Figure C.17. Simple model for He retention in FLIRE.

Equation C.16 can be transformed from the Eulerian frame of reference to the Lagrangian frame of reference by making the change $t=y/v$:

$$\frac{\partial n}{\partial t} = D \frac{\partial^2 n}{\partial x^2} + G(x, t) \quad (\text{C.17})$$

The boundary conditions are:

$$\begin{aligned} n(0, t) &= 0 \\ \lim_{x \rightarrow \infty} n &= 0 \\ n(x, 0) &= 0 \end{aligned} \quad (\text{C.18})$$

The first boundary condition indicates that as soon as helium reaches the surface, it comes out. The second boundary condition establishes that the problem is solved in a semi-infinite domain. The third states that the material is free of helium before the implantation takes place. The implantation source terms has the following form, according to the initial assumptions:

$$G(x, t) = \frac{j}{Wv} \delta(x - r) \delta(t) \quad (\text{C.19})$$

W is the width of the channel, r is the mean implantation range and j is the particle injection rate. The solution to Equation B.2 subject to boundary conditions C.18 is given by ¹⁶:

$$n(x, t) = \int_0^\infty d\xi \int_0^{t_R} \frac{G(\xi, \tau)}{\sqrt{4\pi D(t-\tau)}} \left[\exp\left(-\frac{(x-\xi)^2}{4D(t-\tau)}\right) - \exp\left(-\frac{(x+\xi)^2}{4D(t-\tau)}\right) \right] d\tau \quad (C.20)$$

The integration of Equation C.20 over the τ variable yields:

$$n(x, t) = \frac{j}{Wv\sqrt{4\pi Dt}} \int_0^\infty \delta(\xi - r) \left[\exp\left(-\frac{(x-\xi)^2}{4Dt}\right) - \exp\left(-\frac{(x+\xi)^2}{4Dt}\right) \right] d\xi \quad (C.21)$$

The integration of Equation C.21 gives the concentration profile for this case:

$$n(x, t) = \frac{j}{Wv\sqrt{4\pi Dt}} \left[\exp\left(-\frac{(x-r)^2}{4Dt}\right) - \exp\left(-\frac{(x+r)^2}{4Dt}\right) \right] \quad (C.22)$$

The amount carried into the lower chamber for any time (i.e. distance along the flow) is given by:

$$q(t) = v \int_{A_\perp} n(x, t) dA_\perp = vW \int_0^\infty n(x, t) dx \quad (C.23)$$

The integral in Equation C.23 can be evaluated so the amount carried into the lower chamber per unit time is given by:

$$q(t) = j \cdot \operatorname{erf}\left(\frac{r}{\sqrt{4Dt}}\right) \quad (C.24)$$

The retention coefficient R can now be defined as the ratio of rate of injection into the lower chamber to rate of injection in the upper chamber. By defining the transit time in the upper chamber $t_R = L/v$, and the diffusion time $t_D = r^2/4D$, the expression for the diffusion coefficient is:

$$R = \frac{q}{j} = \operatorname{erf}\left(\sqrt{\frac{t_D}{t_R}}\right) = \operatorname{erf}(\sqrt{\theta}) \quad (C.25)$$

The dimensionless variable θ is defined as the ratio of diffusion to residence time. Equation C.25 is in agreement with results obtained by other authors for this same problem^{17,18}. If the argument of the error function in Equation C.25 is small, the error function can be approximated by its argument within 10% (see Figure C.18):

$$R = \sqrt{\theta} \quad (C.26)$$

Figure C.18. a) Comparison between the error function value and the value of its argument. b) Percent error between the two values as a function of the argument.

From Figure C.18b, it can be seen that for small values of the argument ($< 10^{-1}$), the error reaches a constant value, 11.4%. This means that the following expression can be used to approximate the error function even more accurately:

$$\text{erf}(x) \approx 1.114x \quad (\text{C.27})$$

Use of expression C.27 instead of C.26 reduces the error to about 1%, so more accurate values are obtained

C.4 Corrections to the Simple Model

The model for retention presented in the preceding section is simplified by making multiple assumptions. The effect of such assumptions in the overall model will be discussed in the following sections. Recall that the three main assumptions of the model were:

- The ions are not implanted to the same depth. A range distribution for the ions exists
- The implantation beam is not infinitesimal as assumed on the model: it has a finite size
- The film of lithium has a finite thickness.

Since taken into account all this effects simultaneously would complicate the model beyond the scope of this work, the effect of removing each assumption individually will be discussed here.

Effect of implantation profile

To take into account the implantation profile, the source term, Equation C.19, has the following form:

$$G(x,t) = \frac{j}{W_v} f(x) \delta(t) \quad (\text{C.28})$$

The Dirac delta function for the x coordinate has been replaced by an arbitrary distribution function $f(x)$. The concentration profile is then given by:

$$n(x,t) = \frac{j}{W_v \sqrt{4\pi Dt}} \int_0^\infty f(\xi) \left[\exp\left(-\frac{(x-\xi)^2}{4Dt}\right) - \exp\left(-\frac{(x+\xi)^2}{4Dt}\right) \right] d\xi \quad (\text{C.29})$$

In order to obtain more realistic deposition profiles, Monte Carlo simulations of He ions with different energies incident on a Li target were performed. A simple function that closely matches the profiles obtained in the simulations is a Gaussian curve multiplied by x , as can be seen from Figure C.19. Multiplication by x allows to meet the boundary conditions C.18, even for lower energies when a Gaussian may give a non-zero value at $x=0$. Each modified Gaussian curve has with three parameters: the mean value μ , the spread σ and the normalization constant A :

$$f(x,t) = \frac{A}{\sigma} x \cdot e^{-\left(\frac{x-\mu}{\sigma}\right)^2} \quad (\text{C.30})$$

Figure C.19. Deposition profiles for incident He ions on a Li target for the energies specified. Dots represent data from Monte Carlo simulation (10000 particles), solid line is the corresponding modified Gaussian fit.

Values for these three parameters for different He ion energies incident on a Li target are shown in Figure C.20. The rate of injection into the lower chamber, defined in Equation C.23, now yields:

$$q(t) = \frac{j}{\sqrt{4\pi Dt}} \int_0^\infty dx \int_0^\infty f(\xi) \left[\exp\left(-\frac{(x-\xi)^2}{4Dt}\right) - \exp\left(-\frac{(x+\xi)^2}{4Dt}\right) \right] d\xi \quad (\text{C.31})$$

Figure C.20. The three parameters for Equation C.30 as a function of energy, obtained from fits to Monte Carlo simulation results.

The retention is then calculated by just dividing Equation C.31 by j and by a normalization factor n_0 which depends only on the three fitting parameters, given by:

$$n_0 = \int_0^\infty f(\xi) d\xi = \frac{A}{2} \left\{ \sigma e^{-\mu^2} + \sqrt{\pi} \mu \left[1 + \operatorname{erf}\left(\frac{\mu}{\sigma}\right) \right] \right\} \quad (\text{C.32})$$

Replacing $f(\xi)$ by the modified Gaussian function and including the normalization factor, the retention coefficient has the following form:

$$R = \frac{\frac{2}{\sigma\sqrt{4\pi Dt_R}} \int_0^\infty dx \int_0^\infty e^{-\left(\frac{\xi-\mu}{\sigma}\right)^2} \xi \left[e^{-\left(\frac{\xi-x}{4Dt_R}\right)^2} - e^{-\left(\frac{\xi+x}{4Dt_R}\right)^2} \right] d\xi}{\sigma e^{-\mu^2} + \sqrt{\pi}\mu \left[1 + \operatorname{erf}\left(\frac{\mu}{\sigma}\right) \right]} \quad (\text{C.33})$$

Equation C.32 is a lot more complicated than the expression found for the delta function case, Equation C.25. In order to compare how important the effect of the implantation profile is, retention results from Equations C.25 and C.33 need to be compared. As an example, consider the case of 1 keV He ions incident on a Li target. From Figure C.20, the parameters from the modified Gaussian in Equation C.30 are $\mu = 280 \text{ \AA}$, $\sigma = 130 \text{ \AA}$ and $A = 40$. The mean diffusion time t_R is fixed to $0.02 \text{ }\mu\text{s}$ (obtained from the value of the mean range μ and assuming $D = 10^{-4} \text{ cm}^2/\text{s}$) for the case of the delta function model. The transit time t_R is varied between 10^{-10} and 1 second. The results of the comparison of the two models are presented in Figure C.21. Figure C.21a presents the values compared directly, while the relative percent difference between the two values is presented in Figure C.21b. For very small values of t_R , both models are very close: both predict retention very close to 100%. As t_R increases, the retention goes down and the difference between the two models becomes more important. The difference between the two models seems to reach a steady value in the range of 20 – 25 %. The model with the realistic implantation profile consistently predicts higher retention, except for low values of t_R . This is because the delta function model does not consider particles implanted beyond the mean range, and the model with the profile does. Interestingly, the profile model has 35% of the total particles beyond the mean range, which is a number close to the difference between the two models. This means that if this model is more accurate, the diffusion coefficient estimation from retention measurements based on the simple model actually underestimates the diffusion coefficient by 35%.

Figure C.21. Effect of an initial deposition profile on the retention coefficient. In a), the values obtained for both the realistic and localized (Delta function) profiles are shown. In b), the percent difference between the two is presented.

Effect of finite-size beam

Another assumption in the simple model is that all particles get implanted at the instant $t = 0$. The simplest way to remove this assumption is to assume that the incident ion beam has a width b , so in the Lagrangian frame of reference the beam is “turned on” for a finite amount of time t_I , given by:

$$t_I = \frac{b}{v} \quad (\text{C.34})$$

where v is the flow velocity. The source term is then transformed to:

$$G(x, t) = \begin{cases} \frac{j}{Wvt_I} \delta(x-r) & \text{for } t \leq t_I \\ 0 & \text{for } t > t_I \end{cases} \quad (\text{C.35})$$

The concentration profile is again obtained by performing the integration of the source term with the Green function:

$$n(x,t) = \frac{j}{Wv t_l} \int_0^t d\tau \int_0^\infty \frac{\delta(\xi - r)}{\sqrt{4\pi D(t-\tau)}} \left[\exp\left(-\frac{(x-\xi)^2}{4D(t-\tau)}\right) - \exp\left(-\frac{(x+\xi)^2}{4D(t-\tau)}\right) \right] d\xi \quad (C.36)$$

The integral with respect to ξ is immediate and yields the following expression:

$$n(x,t) = \frac{j}{Wv t_l} \int_0^t \frac{1}{\sqrt{4\pi D(t-\tau)}} \left[\exp\left(-\frac{(x-r)^2}{4D(t-\tau)}\right) - \exp\left(-\frac{(x+r)^2}{4D(t-\tau)}\right) \right] d\tau \quad (C.37)$$

The integration of Equation C.37 has an analytic form and gives the expression for the density as a function of time and depth:

$$n(x,t) = \frac{2j}{Wv t_l} \left[\frac{t}{\sqrt{4\pi Dt}} \left(e^{-A_-^2} - e^{-A_+^2} \right) + \frac{A_- \operatorname{erf}(A_-) - A_+ \operatorname{erf}(A_+)}{\sqrt{4Dt}} + \frac{H(x)}{4Dt} \right] \quad (C.38)$$

The constants A_+ and A_- in Equation C.38 are defined as:

$$A_\pm = \frac{x \pm r}{\sqrt{4Dt}} \quad (C.39)$$

And the function $H(x)$ is given by:

$$H(x) = \begin{cases} 2x & \text{for } x < r \\ 2r & \text{for } x > r \end{cases} \quad (C.40)$$

Figure C.22 shows the evolution of the concentration for times smaller than the irradiation time, set to 1 ms, normalized by the quantity j/Wv . For the calculation, the diffusion coefficient was set to $10^{-4} \text{ cm}^2/\text{s}$, and the implantation range r to 300 Å. The concentration profile after an irradiation of 1 ms and the profile obtained for the instantaneous source after 1 ms are shown in Figure C.23. The concentration profiles are different for both cases. Injection spread over time slows down desorption of implanted particles, and the peak concentration occurs closer to the surface. In Figure C.24, the evolution of the flux out of the surface for the two cases is shown. Notice that the distributed source reaches a steady state, where the flux out of the surface does not change with time. This fact may also be responsible for the higher concentration after the same amount of time for the distributed case.

Figure C.22. Evolution of concentration profile during beam irradiation. Time is given as a percent of the total irradiation time.

Figure C.23. Comparison of concentration profiles for an instantaneous ($t=0$ s) source and a source of the same strength distributed over 1 ms. Both profiles are evaluated at 1 ms.

Figure C.24. Evolution of the flux coming out of the surface for the same two cases as Figure C.23.

Another useful comparison between the two cases is the retention coefficient, defined in Equation C.25. Recall that the calculation requires integration over the x variable, as suggested by Equation C.23. The retention for the distributed case at the end of the irradiation time is given by the integration of Equation C.38 over the x variable:

$$R(t) = 2 \int_0^{\infty} \left[\frac{(e^{-A_-^2} - e^{-A_+^2})}{\sqrt{4\pi Dt}} + \frac{A_- \operatorname{erf}(A_-) - A_+ \operatorname{erf}(A_+)}{\sqrt{4Dt}} + \frac{H(x)}{4Dt} \right] dx \quad (\text{C. 41})$$

The integration yields the following expression:

$$R = 2 \left[\frac{\theta}{\sqrt{\pi}} e^{-\theta^2} + \frac{1}{2} \operatorname{erf}(\theta) - \theta^2 \operatorname{erfc}(\theta) \right] \quad (\text{C.42})$$

The variable θ is equal to the ratio of diffusion time divided by transit time (or, for the case of the distributed source, exposure time):

$$\theta = \frac{r}{\sqrt{4Dt_I}} = \sqrt{\frac{t_D}{t_I}} \quad (\text{C.43})$$

For large values of θ , corresponding to a very small beam, Equation C.42 is equivalent to Equation C.25. Figure C.25 presents the comparison between Equations C.42 and C.25.

Both give a retention coefficient of 1 for sufficiently large θ , and both expressions tend to zero as θ gets smaller. For small values of θ , the retention coefficient for the distributed source is twice as big as that for the instantaneous source. The difference decreases from 55% to 0 % between the θ values of 0.01 and 10.

Figure C.25. a) Retention coefficient values for the instantaneous and distributed ion sources as a function of the parameter θ defined in Equation C.43. b) Percent difference between the cases.

The model for the finite beam is, however, incomplete. The calculations so far describe the profile up to the point where $t = t_l$. What happens beyond that? In order to determine the behavior of the concentration profile for times larger than t_l , a scheme similar to the one presented for estimating the effect of a distributed implantation profile would be needed. The concentration would be given by Equation C.29, but the function $f(\xi)$ would be given by Equation C.38 instead of Equation C.30. Numerical integration of Equation C.29 with respect to the x variable would give the retention as a function of time for times $t > t_l$. Figure C.26 shows two profiles for the two cases. For 0.001 seconds after irradiation (Figure C.26a), the distributed source profile is higher than the one corresponding to the instantaneous source, but the difference is not as marked as it was right at the end of the irradiation (Figure C.23). If more time passes after irradiation, the concentration profiles for both cases become identical (Figure C.26b). For this case, integration of both profiles gives a retention of 0.16 %, and the error between the two models is very small (~ 1 %). It can be concluded from the calculations of the present section that the finite beam correction is important only if the size of the beam is of the same order as the path length of the film before exiting the irradiation chamber. For the case of FLIRE, however, where the size of the path length is two orders of magnitude larger than the beam width, this effect is not very important. Even though the profiles are very different right after exposure to the beam ends, the profile for the distributed

case relaxes to one very similar to the one for the instantaneous source before the flow exits the main chamber.

Figure C.26. Concentration profile for distributed and instantaneous source terms for a) 0.001 seconds and b) 0.01 seconds after end of irradiation. Irradiation time was set equal to 0.001 seconds.

Helium Diffusion Coefficient

By using the retention data and the models developed in earlier, the diffusion coefficient for He in Li can be estimated. Recall that for the simple model the retention coefficient is given by:

$$R = 1.114\sqrt{\theta} = 1.114\sqrt{\frac{\nu r^2}{4LD}} \quad (\text{C.44})$$

It has already been seen that corrections due to finite beam and implantation profile are small for the FLIRE operating conditions. The long-term trapping is a small fraction (see Figure C.15), so the diffusion coefficient can be approximated by:

$$D = 1.24 \frac{\nu r^2}{4LR^2} \quad (\text{C.45})$$

All the parameters in Equation C.45 are known: L can be determined by direct measurement, and the effect of the lithium pool in the path length can be included in the uncertainty of this measurement; ν is calculated from changes in driving pressure; r is estimated from Monte Carlo calculations, and R is measured experimentally and the values are given in Figure C.7. The parameters are summarized in Table C.1.

Table C.1. Parameters used for the calculation of the diffusion coefficient from Equation C.45

Parameter	Value
Particle range, r	$(2.5 \times 10^{-6} \text{ cm/keV}) E$
Path length, L (cm)	10 ± 0.5
Flow velocity, v (cm/s)	44 ± 6
Retention coefficient, R	$[(5.3 \pm 0.3) \times 10^{-3} \text{ keV}^{-1}]E$

Two approaches can be taken to find the diffusion coefficient from Eqn. C.45: one is to use the retention data linear fit to calculate the diffusion coefficient, the other is to use the individual measured values of the retention. Earlier, the retention coefficient R was found to follow a linear trend with energy:

$$R = K_1 \cdot E \quad (\text{C.46})$$

The mean range, as already discussed, is linear with the energy, and it is given by:

$$r = (2.5 \times 10^{-6} \text{ cm/keV})E \quad (\text{C.47})$$

Substituting Equations C.46 and C.47 into Equation C.45, the diffusion coefficient is then given by:

$$D = 1.765 \times 10^{-12} \frac{v}{K_1^2 L} \quad (\text{C.48})$$

The flow velocity v in the experiments was set to 44 ± 6 cm/s, and the distance from the beam striking point to the exit of the upper chamber L is fixed at 10.5 ± 0.5 cm. The constant $K_1 = (5.6 \pm 0.2) \times 10^{-2} \text{ keV}^{-1}$ was determined from the retention data. Use of these numbers yields a value for the diffusion coefficient of $(3.7 \pm 0.6) \times 10^{-7} \text{ cm}^2/\text{s}$. To verify how good this value is, multiple values of the diffusion coefficient were calculated using the individual values of the retained fraction obtained from the two sets of experiments described in the previous chapter. Equation C.45 was used to calculate this individual values, and the results are plotted in Figure C.27. As expected, the diffusion coefficient does not show any dependence with energy, and the average value obtained is equal to $(4.7 \pm 0.8) \times 10^{-7} \text{ cm}^2/\text{s}$, very close to the one obtained from the linear interpolation of R . The individual values of the diffusion coefficient have large error bars, and most of the uncertainty comes from the error in the retention measurement.

The value obtained for the diffusion coefficient seems very small. This may be a consequence of some mechanism that hinders the release of implanted helium, such as bubble formation or a surface film acting as a diffusion barrier. For these cases, the simple model underestimates the retention, and the consequence is that the diffusion coefficient moves up. Despite this fact, the qualitative behavior of the retention seems to agree with the model. Other more complete models that include other effects such as

flow turbulence, bubble nucleation, surface ripples, etc. can be used to obtain better estimates of the diffusion coefficient once the mechanism controlling the release is identified.

Figure C.27. Diffusion coefficient calculated with the values from Table C.1. The calculated values are the symbols, while the average is the solid line. Uncertainty in the average is indicated by the dashed lines.

The calculation of the diffusion coefficient allows for the extrapolation of the retention data from FLIRE operation parameters to any arbitrary parameters, assuming the model holds for the extrapolated system. Consider, for instance, a fusion reactor where the lithium travels 1 meter at 10 m/s, with 1 keV He particles incident on the divertor. If the lithium is operated at 250 °C, the diffusion coefficient is equal to the average value shown in Figure C.27. A plot of θ vs. R is shown in Figure C.28, with the points obtained in this study and the point for the fusion reactor operated with the parameters mentioned before. For the reactor operating parameters suggested, the retention is above 1%, close to meeting the requirement for helium pumping quoted in the literature [45]. This requirement for pumping is derived from balancing the He generation rate (proportional to the reactor power level) and data on deuterium/tritium current to the divertor. The ideal ratio of fuel confinement time to helium confinement time τ_{DT}/τ_{He} is on the order of 0.1-0.5, indicating that the helium current to the divertor is five to ten times lower than the fuel current. For a 1000 MW reactor, with typical D/T currents to the divertor of 10^{22} particles/sec, the ratio of helium generation to helium leakage is of the order of 0.01, putting a lower limit on the pumping efficiency of the liquid divertor. The retention data from this work seems to suggest that such requirement is achievable since the retention is high enough.

Figure C.28. Comparison of the data obtained in this study with the model, and the extrapolation to a fusion reactor with typical parameters.

D HYDROGEN CAMPAIGN IN FLIRE

D.1 Importance of hydrogen retention data

The issue of hydrogen transport by flowing liquid PFCs is of vital importance for fusion reactor design due to its impact on aspects such as fuel recycling operating regime, tritium trapping and subsequent recovery, among others. Extensive studies have been carried on for most of the typical solid PFC materials such as carbon, beryllium and tungsten¹⁹, but very little data exists for liquid PFC materials such as lithium, tin or FLIBE.

Unlike helium, hydrogen is soluble in lithium and hence can be trapped long term in the metal. Some authors have suggested that all implanted hydrogen stays within the lithium²⁰; this would make the liquid PFC a 100% fuel recycling surface, which would result in the suppression of the temperature pedestal between the plasma core and the edge of the device²¹. For such cases, rapid recovery of tritium fuel would be necessary.

The FLIRE facility has the capability of studying hydrogen transport by a flowing film of lithium metal. Prompt release upon entry into the lower chamber can be measured just as in the case of the helium prompt release described in earlier. In addition, the amount retained in the batch for a larger time scale can be measured by use of the thermal desorption spectroscopy (TDS) technique, which will be described in detail later in this section.

D.2 The Li – LiH thermodynamic system

The lithium-hydrogen thermodynamic system has been widely studied, both as part of the nuclear fusion program and, more recently, as part of lithium battery technology, as well as the possibility of using lithium as means of storing hydrogen for fuel cells. Figure D.1 presents the phase diagram of the lithium/hydrogen system, where five different regions can be identified. Region I is bounded by a line at approximately 180 °C (eutectic temperature): at this temperature, the lithium phase changes from solid to liquid. Region II is the region where lithium hydride in solid phase coexists with liquid lithium. A critical concentration ($\sim 19\%$) can be identified, below which a transition is made to Region III, comprised of the liquid lithium phase only; above 19% atomic H content, a transition occurs at 685 °C (monotectic temperature) when the solid hydride phase turns liquid (Region IV). Region V, comprised of pure liquid LiH, can only be reached at very high concentrations of hydrogen.

Figure D.1. The lithium-hydrogen phase diagram (from ²²).

The phase diagram of Figure D.1 is not at constant pressure. Pressure-composition isotherms for the Li/LiH system reveal a peculiar behavior of the system. In the regions to the left and right of the miscibility gap (regions III and V in the phase diagram), the equilibrium pressure increases for a given temperature; once the concentration reaches the boundary between the one-phase and two-phase regions, the pressure reaches a plateau such that increasing the concentration does not cause increase in the equilibrium pressure. After the other side of the miscibility gap is reached, further increase of the concentration in the lithium increases the equilibrium pressure. In Figure D.2a, a phase diagram similar to the one for the Li-LiH system is shown, along with four pressure concentration isotherms. Pressure goes up when $x < x_i$ and when $x > x_i'$, and stays constant between those two limits. The pressure plateau gets narrower as temperature

increases, and broader as temperature is lowered. Actual pressure-concentration isotherms for the Li-LiH system have been determined experimentally for temperatures above the monotectic, and are shown in Figure D.3b. A variation of the same type of data, pressure – temperature curves of constant concentration, for temperatures below the monotectic is shown in Figure D.3²³.

Figure D.2. a) Phase diagram for a typical 1:1 alkali hydride, showing pressure-composition isotherms for four different temperatures. Equilibrium pressure reaches a plateau for concentration values inside the two-phase region at each temperature. b) Pressure-composition isotherms for the Li/LiH system (from 24).

Figure D.3. Constant concentration curves in the P-T plane for the Li/LiH system, below monotectic temperature (data from 23).

Since the hydrogen concentration expected in FLIRE is very low, more information about the miscibility gap in the low end of the hydrogen concentration would be useful to have. Between 200 and 600 °C, the miscibility gap curve is well approximated by the following expression²⁵, plotted in Figure D.4:

$$\ln(x_{LiH}) = 3.679 - \frac{5472}{T} \quad (D.1)$$

Figure D.4. The low-concentration end of the Li-LiH phase diagram.

The fractions of hydrogen and lithium hydride are related by:

$$x_H = \frac{x_{LiH}}{1 + x_{LiH}} \quad (D.2)$$

The rise in equilibrium pressure as concentration increases in the lithium-rich phase does not obey Sievert's law, and calculations of activities and heats of solution are required to obtain the behavior of P as a function of T and x_H ²⁶. However, at low pressures (and hence low concentrations), the lithium – hydrogen equilibrium can be described adequately by Sievert's law²⁷:

$$n_s = K_s P^{1/2} \quad (D.3)$$

The following expression has been used to calculate solubility of hydrogen isotopes in lithium²⁸:

$$K_s = 1.2 \times 10^{-5} \exp\left(\frac{44400}{RT}\right) \quad (D.4)$$

K_s is given in units of % atomic/ $\text{Pa}^{1/2}$, T is in K and R in J/mol K. The expression above is valid for temperatures between 723 and 923 K (450 to 650 °C). During exposure in FLIRE, typical lithium temperature is between 250 and 300 °C, which falls outside the

range of validity for Equation D.4. However, the extrapolation of the function should give a good estimate for the concentration at the surface during exposure assuming that Sievert law still holds.

Figure D.5. Sievert constant for hydrogen – lithium equilibrium. Solid line shows the valid range, while dashed line shows extrapolated values.

By combining the data from Figure D.4 and Figure D.5, it is possible to predict if hydride precipitation will occur. The procedure to do it is as follows:

- Pick an operating temperature.
- Find the minimum concentration required to enter the two phase region from Figure D.4.
- Calculate from either equation Equation D.4 or Figure D.5 the pressure, given T and x_H from the previous two steps.
- Compare to operating pressure. If the operating pressure is below the pressure calculated in the previous step (the minimum to produce precipitation), then the hydrogen stays in solution. Otherwise, hydride has been formed and the system is in a two-phase region.

In the case of FLIRE, the operating temperature is typically between 250 and 300 °C. This means that the minimum concentration required to form the hydride phase is between 0.1 and 0.3 %. Hence, the pressure required to precipitate would be between 7×10^{-8} and 4×10^{-6} Torr. The ion source in FLIRE requires neutral gas pressure on the order of 10^{-5} Torr to produce the ion beam. This means that this backfilling pressure in

the chamber will induce hydride formation in the lithium flow if equilibrium is reached, since it is above the pressure threshold required to enter the two phase region. Evidence of this will be shown in the sections that follow.

D.3 Simple model for hydrogen retention

The model for retention presented in the preceding section is adequate to describe the release of implanted particles that are not soluble in the liquid. For the lithium/helium system, that is a very good approximation. However, that is not the case for the lithium/hydrogen system, since hydrogen is soluble in lithium and cannot leave the surface unless it recombines with another hydrogen atom to form a gas molecule.

For soluble species, like hydrogen, retention is related to two sources: 1) implantation due to the ion beam, and 2) absorption of gas at the interface. The second mechanism has been utilized over many years for separation of gases by the use of wetted wall towers²⁹. It is important to try to quantify the relative importance of both mechanisms. The implantation model is identical to the one presented for helium in the Appendix C. For the absorption, the problem can be stated as follows:

$$\frac{\partial n}{\partial t} = D \frac{\partial^2 n}{\partial x^2} \quad (\text{D.5})$$

Notice that the volumetric source term, which was present in Equation C.1, has been dropped, since no volumetric source of material is present for this case. The boundary conditions for the absorption process are:

$$\begin{aligned} n(0, t) &= n_{sat} \\ \lim_{x \rightarrow \infty} n &= 0 \\ n(x, 0) &= 0 \end{aligned} \quad (\text{D.6})$$

The only difference between boundary conditions C.3 and D.6 is the saturation condition at the surface $x = 0$. The solution of Equation D.5 subject to D.6 is given by¹⁶:

$$n(x, t) = n_{sat} \operatorname{erfc} \left(\frac{x}{\sqrt{4Dt}} \right) \quad (\text{D.7})$$

Unlike the diffusion controlled case, in the absorption case the metal keeps gaining material rather than losing it to the surface, so the average concentration in the film increases with time. Figure D.6 shows the evolution of the average concentration in a 2 mm film assuming a diffusion coefficient of $10^{-4} \text{ cm}^2/\text{s}$. The calculation never reaches saturation due to the assumption of the semi-infinite domain. However, it is accurate if all the non-zero values of the concentration are within the specified depth for calculating the average (2 mm in this case). For typical conditions in FLIRE, the time of exposure to

the gas is on the order of 100s of milliseconds, meaning that the expected ratio of average to saturation concentration is on the order of 5%. If the top layer is fully saturated (50% atomic fraction of hydrogen), then the average atomic content of the stream at the end of the path is on the order of 3%. If the top layer reaches only the solubility limit (0.1 – 0.3 %), then the average goes down to about 0.01 %, or 100 ppm.

Figure D.6. Ratio of average to saturation concentration in a 2 mm film. Diffusion coefficient is assumed to be $10^{-4} \text{ cm}^2/\text{s}$

On the other hand, consider a beam of H_2^+ ions incident on the stream. Assuming that all the implanted particles stay within the film, the amount of injected hydrogen is:

$$N_{H,b} = \frac{2I_{H_2^+}t_R}{q_e} \quad (\text{D.8})$$

The amount of lithium atoms transported during the transit in the chamber is given by:

$$N_{Li} \approx \frac{v\delta W \rho N_A t_R}{MW_{Li}} \quad (\text{D.9})$$

Using the numbers $I=10^{-5} \text{ A}$, $t_R=300 \text{ ms}$, $\delta=2 \text{ mm}$, $W=1 \text{ cm}$, $v=60 \text{ cm/s}$, $\rho=0.5 \text{ g/cm}^3$, $MW_{Li}=6.9 \text{ g/mol}$, $N_A=6.02 \times 10^{23} \text{ atoms/mol}$ and $q_e=1.602 \times 10^{-19} \text{ C/ion}$, the number of implanted hydrogen ions is on the order of 4×10^{13} , and the amount of lithium atoms is 2×10^{23} , giving a very small hydrogen/lithium ratio, about 0.1 ppb. This means that unless the saturation content at the surface is very small, the absorption of gas will contribute more to the retention of hydrogen in the system.

This model does not take into account the precipitation of hydride, which is predicted to occur as discussed in the preceding section. Hydride formation would effectively enhance hydrogen intake, since it will act as a sink and more hydrogen will be taken until the whole sample saturates. Of course, the time scales of the experiment do not allow equilibrium to be reached in the lithium overall, but only in the interface between the gas phase and the condensed phases. Reaction kinetic data would be necessary in order to estimate hydrogen intake more accurately, since that would allow to account for hydride formation.

D.4 Measurement of long-term hydrogen trapping

As mentioned earlier, the solubility of hydrogen in lithium may lead to the formation of a hydrogen solution in lithium, or even formation of a hydride phase. Although some of the hydrogen may come out promptly (see preceding sections), long-term trapping of hydrogen may occur. The hydrogen will slowly come out if no hydrogen buildup on top of the lithium is allowed, since the hydrogen comes out of the lithium trying to establish thermodynamic equilibrium between the liquid and gaseous phases.

The method used to determine the amount of long-term trapping is temperature programmed desorption (TPD), also known as thermal desorption spectroscopy (TDS). The method consists of heating the sample in a linear fashion while making vacuum on top of the liquid. The TDS technique can be used to determine activation energies for desorption, surface recombination constants, heats of solution and impurity content. The first TDS experiments in FLIRE were aimed to find deuterium content in the lithium sample after exposure to both deuterium gas and a deuterium ion beam.

D.5 The FLIRE TDS chamber

The TDS chamber in the FLIRE facility is shown in Figure D.7. A Balzers TPH-062 turbomolecular pump with pumping speed of 50 liters/sec makes the vacuum in the chamber. The magnetic sector RGA is shared with the lower chamber where the prompt release measurements are made. Two gate valves can isolate the RGA from each chamber, so the RGA can “sniff” one chamber or the other without interference from residual gases in the other chamber (see Figure D.7). Heating of the chamber (up to 1200 °C) is accomplished by a 4-kW Thermcraft cylindrical clamp furnace, shown in Figure D.8. The furnace is controlled by a combination of a fuzzy logic PID and a high power solid-state relay that can ramp up the temperature in a linear fashion. Multiple thermocouples in the outside and an internal thermocouple probe monitor the temperature at different points of the chamber during the experiment.

Figure D.7. Photograph and schematic of the TDS FLIRE chamber.

Figure D.8. The clamp furnace that heats the TDS FLIRE chamber, in open position.

The temperature program used for the TDS studies in FLIRE is a ramp of 2 °C/min, starting at 250 °C and finishing at 600 °C. The batch soaks at 600 °C for one hour to try to remove as much hydrogen as possible, and then it cools down to 250 °C again. Hence, the total treatment of the lithium batch takes about 4 hours. The traces of different species of interest during the bakeout of the empty TDS chamber are shown in Figure D.9. The main impurities in the chamber are water and hydrogen, as well as some air (nitrogen and oxygen). The presence of some residual deuterium is evident by the presence of masses 3 (HD) and 20 (D₂O).

Figure D.9. Evolution of residual gases in the empty TDS chamber during heating.

D.6 TDS data analysis

In order to analyze the data from the TDS experiments, a more careful analysis of the P-C isotherms of Figure D.2 is needed. Consider a lithium sample sitting at a temperature T_0 , with no hydrogen on it and perfect vacuum on top. Suddenly, hydrogen gas at P_0 is added, and the concentration eventually reaches the equilibrium value $x_{eq,0}$ if the pressure P_0 is kept constant, as shown in Figure D.10. If the pressure is increased again to P_1 instantaneously and kept there, the concentration increases until a new concentration equilibrium value $x_{eq,1}$ is reached. Now, what happens if the pressure is raised to a value P_2 above the corresponding plateau pressure and kept constant there? After a long time, the system would have cruised above the plateau pressure past the two-phase region, and reach the other portion of the isotherm where pressure is a function of composition, in the hydride-rich phase of the system. Since the slope of the PC isotherm is typically very steep in this region, small increases in concentration require large increases in pressure, like the jump to P_3 in order to reach $x_{eq,3}$.

Figure D.10. Isothermic formation of a hydride-rich system.

Now, what would happen if one of the pressures happens to be the plateau pressure? Assume, for example, that the pressure is raised to P_2 , but instead of being kept constant, it can relax to an equilibrium value. Then the blue line on top of the plateau is not parallel and the path intersects the equilibrium isotherm at a certain concentration. If at that point another short burst of pressure is created to raise it to the value P_2 again, then the pressure will go down and it will intercept the equilibrium concentration further down in the plateau. If we repeat this process multiple times, a series of “teeth” appear on top of the plateau, as can be seen in Figure D.11. These processes are reversible; that is, if starting with the hydride we reduce the pressure in the same way, we should go back to the original state: pure lithium with no hydrogen. The only difference would be that the blue path would pass under the equilibrium isotherm, but the same “stops” along the path would be touched.

Figure D.11. Formation of the same hydride as in Figure D.10, but with multiple equilibrium intermediate steps.

Both Figure D.10 and Figure D.11 present isothermic processes. The TDS experiments are not isothermic; in order to draw a theoretical TDS experiment multiple isotherms should be available. Since the phase diagram is available, the ends of the plateau region are known, only the composition-dependent parts of the isotherm would have to be calculated. Consider Figure D.12, where multiple isotherms have been drawn, and a red line delineates the endpoints of each isotherm's plateau (recall from Figure D.2 that these endpoints correspond to the phase boundaries, so the red line resembles the phase boundary from the phase diagram). The starting point is labeled **A**, which corresponds to a lithium sample at temperature T_0 and a known composition. The path **AB** corresponds to the heating of the sample, until the phase boundary is reached at $T=T_1$. At that point, the hydride phase no longer exists and we have hydrogen in solution with lithium. Further heating to T_2 (from **B** to **C**) removes more hydrogen, but this time the escaping hydrogen comes from the saturated solution phase since the hydride phase is not present anymore. Soaking of the sample at temperature T_2 should eventually eliminate all hydrogen from the sample (from **C** to **D** along the isotherm). The speed at which each of these steps takes place is a function of temperature and composition, since the release of hydrogen from the lithium sample is thermally activated and proportional to the concentration at the surface.

Figure D.12. Path of a theoretical TDS experiment in the pressure-composition plane

This theoretical TDS experiment establishes that something happens when temperature T_l is reached, since we move from one phase to another. Below T_l , the increases in temperature produce more hydrogen with almost no change to the composition. Once the temperature goes above T_l , then the release of hydrogen should start slowing down, since now it is required to reduce the hydrogen content in order to increase the equilibrium pressure. This seems to suggest that the maximum desorption should be observed once T_l is reached, and start decreasing after that. For the TDS experiments, this means that the hydrogen content can be estimated from the temperature at which the strongest desorption is observed.

D.7 Results from Molecular Deuterium Exposure experiments

The very first TDS experiment performed in FLIRE was done on a lithium charge that was exposed to a 1 keV, 2 μ A deuterium ion beam, and a deuterium neutral pressure of 5×10^{-5} Torr. The species being tracked in the TDS chamber are deuterium, deuterium hydride (or hydrogen deuteride), lithium deuteride, heavy water (D_2O), deuterium

hydroxide (HDO) and lithium. A strong release of all species is noted at about 50 minutes into the run, corresponding to a temperature of 350 °C, as can be seen in Figure D.13. The strongest release is HD, suggesting that both hydrogen and deuterium are present inside the metal. The increase in the HD signal at higher temperature is probably caused by re-desorption of hydrogen and deuterium from lithium condensates that get hotter as the tank as a whole gets hotter.

Figure D.13. TDS pressure traces for a lithium charge exposed to a deuterium beam and to neutral gas.

In order to confirm if the deuterium/hydrogen had been removed from the sample, the heating sequence was repeated after letting the sample cool down from 600 to 250 °C. The pressure evolution obtained from this experiment is shown in Figure D.14. Something that can be noticed right away is that the peaks at the lower temperatures are gone, indicating that indeed some hydrogen/deuterium removal has occurred. The desorption at the higher temperatures remains, probably confirming the hypothesis that lithium condensing in cold regions of the TDS chamber traps the hydrogen being released, and eventually gets heated high enough to start re-releasing the hydrogen.

Figure D.14. Pressure traces obtained upon reheating the sample from Figure D.13.

The next experiment in the TDS chamber, performed months later after the TDS chamber was dismantled for maintenance, involved lithium that was exposed to neither deuterium gas nor beam. This was done to verify the purity of the lithium. The traces for the different species during the experiment are shown in Figure D.15. The first thing that stands out is how much higher the background levels are with respect to Figure D.13 and Figure D.14. In particular, the mass 4 signal is much higher this time. Strong release of HD again occurs at 50 minutes, just as in the first experiment, with the difference that this time it is not a generalized increase as it was the case before. The release was not expected at all, since this was supposed to be clean lithium. This seems to suggest that even exposure to the residual hydrogen is enough for a significant trapping in the lithium by establishing some kind of equilibrium with it. The high mass 4 background may be responsible for the formation of species containing deuterium.

Figure D.15. Traces for the TDS experiment with clean lithium. Some hydrogen comes out indicating previous exposure, maybe to residual hydrogen.

Yet another experiment was performed in the FLIRE TDS chamber. Right after the initial TDS experiment to determine how clean the lithium really was, the batch was exposed to a deuterium gas pressure of 6.5×10^{-5} Torr, without turning on the ion source. The TDS trace for this experiment is shown in Figure D.16. Again, the absence of early release of deuterium-containing species is noticeable. In this case, the release does not start until 175 minutes have elapsed, and that corresponds to a temperature close to 600 °C. If we correlate the start of the release of HD with the change of phase, from Figure D.4 we obtain that the hydrogen content is between 6 and 8 %. If the peak release in Figure D.13 and Figure D.15 is the phase change, then the content is around 3%.

Figure D.16. TDS trace for a lithium batch exposed to deuterium gas pressure only.

The results from these TDS experiments are somewhat ambiguous, but they do demonstrate that the TDS system in FLIRE is capable of obtaining useful measurements. The release of deuterium seems to be very sensitive to the impurities present in the chamber, so a very clean environment would be required to perform the experiment. The presence of residual hydrogen in the vacuum system is especially harmful to the analysis, since it may contribute to the hydrogen content along with the deuterium to the establishment of an equilibrium state. More in-depth experiments were required to establish if indeed the main source of hydrogen in the lithium is just exposure to neutral gas, as the preliminary calculations show, and to ensure that no low-temperature emission peaks were missed.

The set of experiments consisted of performing a clean-up TDS run without exposing the lithium to deuterium, followed by exposures to neutral deuterium gas only at different pressures. Before reaching the TDS chamber, approximately 400 g of Lithium flows through the upper chamber on a set of ramps, where it is exposed to deuterium gas at a specified pressure. Two runs are discussed: (1) at 7.5×10^{-5} Torr at an average speed of 44 cm/sec for a duration of 30 sec. and (2) at 1 Torr and an average speed of 22 cm/sec for a duration of 60 seconds. The upper chamber flow path and the lower chamber are typically maintained at a temperature of 230°C.

After allowing prompt release in the lower chamber for 5 minutes, the lithium is transferred to the TDS chamber which is maintained at 250°C. The temperature program used for the TDS studies in FLIRE is a ramp of 2 °C/min, starting at 250 °C and finishing at 600 °C (a ramp time of 3 hours). The batch soaks at 600 °C for one hour after the

temperature ramp in order to remove as much hydrogen as possible. After the soaking time is over, heating is stopped, and the lithium cools to 250 °C again.

The first experiment in the TDS chamber, was performed on lithium that was not exposed to deuterium gas. This was done to verify the purity of the lithium. The traces for the different species during the experiment are shown in Figure D.17a. Figure D.17b shows background traces taken a day before with an empty chamber. Strong signal for hydrogen is observed in both cases at temperatures over 400°C. This indicates that the hydrogen and deuterium are desorbed from steel ^[6] and other colder surfaces where Li condensed from previous experiments, not necessarily from the lithium charge itself as discussed earlier, since one of these runs was without any lithium in the TDS chamber. Further supporting evidence for this is that there is no increase in the lithium signal, despite a much higher vapor pressure at elevated temperatures (~ 100 mtorr at 600°C ^[7,10]), hence the lithium condenses onto cooler surfaces before reaching the RGA.

Figure D.17. (a) TDS spectrum with lithium in TDS chamber but no exposure to deuterium. (b) TDS spectrum with no lithium in chamber.

In the second experiment, the absorption of deuterium by flowing lithium was measured by flowing lithium through the upper chamber with a background gas pressure of 7.5×10^{-5} Torr without operating the ion gun, and careful attention was paid to the initial stages of the TDS heating. At the moment of transferring Li (5 min into the scan), from the lower chamber (at 230°C) to the TDS chamber (at 250°C) there is a weak release of hydrogen and deuterium, as shown in Figure D.18. This indicates that very small amounts of deuterium were trapped during the exposure which was then released when the temperature is raised in a step-wise manner. This release may have been missed in previous runs because the shared RGA may have been monitoring the lower chamber (where prompt release from the stream is observed), rather than the TDS chamber when the transfer took place. Note that all TDS runs shown earlier also started at 250°C, and that there is an apparent peak at 550°C in Figure D.18, just as there was a peak in earlier measurements.

Figure D.18. (a) TDS spectrum of lithium exposed to neutral deuterium gas without deuterium beam. (b) Close-up of TDS spectrum of lithium exposed to neutral deuterium gas only (no beam)

At 230°C the equilibrium concentration indicated by the Li-H phase diagram is 0.1%. At 250°C it is 0.2%. The fact that D was observed upon raising the temperature implies that the fraction of LiH in the sample was greater than 0.1%, but less than 0.2%, prior to the temperature step. This deuterium concentration is close to the deuterium concentration expected from exposure to $\sim 10^{-6}$ Torr of deuterium, as calculated above; however, the actual background gas pressure was 7.5×10^{-5} Torr. This implies that the deuterium absorption rate may be too slow for the surface concentration to equilibrate within the residence time of the lithium in the upper chamber. One reason that the surface concentration of lithium deuteride may not reach equilibrium is that the formation of lithium deuteride is limited by the dissociation rate of molecular deuterium on the surface of the stream, since molecules must dissociate before they are absorbed.

To verify this conclusion, the residual lithium in the TDS chamber was reacted in a tank of flowing water, cleaned, reinstalled and baked out up to 600°C, and the dry-run measurements were repeated. Following this, the lithium charge was transferred to the TDS chamber after exposure to approximately 1 Torr of deuterium. The purpose of this experiment was to examine the influence of deuterium pressure in the upper chamber on the deuterium concentration in the lithium. Despite several orders of magnitude higher pressure, the deuterium pressure trace still exhibited a peak between 230 and 250°C, hence the absorbed deuterium concentration was still between 0.1 and 0.2%.

The results from these TDS experiments demonstrate that the TDS system in FLIRE is capable of obtaining deuterium absorption measurements in flowing lithium. The release of deuterium seems to be very sensitive to impurities present in the chamber, hence a very clean environment is necessary to perform the experiment. Compared to the amount of deuterium that can be implanted by the existing ion gun, deuterium absorption directly from the background gas dominates.

To increase the deuterium ion and atomic deuterium flux incident on the flowing lithium surface, a DC plasma source was constructed and installed in FLIRE. Figure D.19 shows

a photograph of the DC plasma source operating in FLIRE. The design of the source is simple and consists of an anode plate at one end of an alumina tube and a cathode cap on the other (near the ramp). The cathode cap is grounded to prevent the formation of a secondary discharge in FLIRE's upper chamber and has a screened hole to allow ions to pass through and strike the lithium on the ramp. A Langmuir probe is positioned between the cathode cap and the ramp to provide insitu calibration of the plasma source during experiments. This plasma ion source was tested in FLIRE, but there was not sufficient time to use this source for measurements during this Phase II project. However, experiments with the source are continuing at the University of Illinois.

Figure D.19. Photograph of FLIRE DC plasma source.

Summary

The absorption of deuterium in a flowing lithium stream was measured in FLIRE for deuterium gas pressures of 7.5×10^{-5} Torr and 1 Torr using TDS with temperatures between 250°C and 600°C. These experiments resulted in a deuterium concentration of ~0.1% to 0.2%, indicating that the absorption rate is limited by surface dissociation of the molecular deuterium.

No Lithium vapor was observed, indicating that lithium vapor condenses on cooler surfaces between the lithium charge and the RGA. This condensed layer causes some absorption and re-release of impurities during the higher-temperature portions of the TDS run, but desorption from remote condensed surfaces occurs at sufficiently high temperatures in the TDS tank itself to avoid interference with the measurement of low concentrations of deuterium.

Experimental results were compared to a wetted wall tower theoretical model. The model assumed that the surface concentration of absorbed deuterium was saturated at

50%, but over-predicted the average concentration by more than an order of magnitude (3% as opposed to 0.2%). This shows that the surface of the flowing liquid is not quickly saturated with atomic deuterium, relative to the exposure times in FLIRE (~ 0.5 s), which further indicates that dissociation of the molecular deuterium limits its absorption rate.

E MHD Experiments in FLIRE

FLIRE has also been used to study the effect of MHD forces on free-surface flowing liquid metals. This was done by adding a permanent magnet mount on either side of a lithium flow ramp and inserting current leads into the lithium flow path. The magnet holder assembly installed in FLIRE provided a magnetic field across the lithium flow channel, as shown in Figure E.1. The measured field intensity is shown in Figure E.2.

The direction of the current flow along the ramp can create either a lifting or pressing force on the flow.

Figure E.1. Setup for MHD experiments in FLIRE

Figure E.2. Magnetic field produced by the permanent magnets installed in FLIRE.

For a known magnetic field strength, the volumetric force due to an electric current flowing parallel to the liquid flow can be calculated as:

$$F_{EM}(x) = j_{Li} B(x) \quad (E.1)$$

And the gravitational force per unit volume (parallel or anti-parallel to the electromagnetic, EM, force) is given by:

$$F_g(x) = \rho g \cos \theta \quad (E.2)$$

Here, ρ is the lithium density, and θ is the inclination angle of the channel with respect to vertical. For constant volume elements, the ratio of EM to gravitational force is given by:

$$\frac{F_{EM}}{F_g} = \frac{I_{Li} B(x)}{A_{Li} \rho g \cos \theta} \quad (E.3)$$

This ratio provides an indication of what current-field combination will result in lift-off of the flow from the surface, neglecting surface tension/attraction forces. Calculating this ratio requires knowledge of the current passing through the liquid metal.

Because both the liquid and the channel are conductors, electric current will flow through both materials; however, only the fraction of the electric current that passes through the liquid metal contributes to the EM force on the flow. To determine what fraction of the total current passes through the lithium, the current was measured with the two leads touching the channel, and then again when the lithium was flowing. The percent change in current between the two measured values was assumed to be the percent of total current going through the lithium. This change was measured to be 23%, which agrees

with estimates based on resistivity data for steel and lithium and cross sectional area estimates. Calculated values of the current change range from 15 to 30%.

The force ratio perpendicular to the liquid flow as a function of distance along the channel is shown in Figure E.3 for several total current values. A flow cross sectional area of 0.1 cm^2 (Li film 1 mm thick, 1 cm wide) was assumed for these calculations. For current values under 2 A, the ratio of forces is less than 1, indicating that gravitational forces dominate. Above this value, EM forces are able to overcome the weight of the fluid (if the direction of current flow results in an EM force opposing the gravitational force), and the liquid should lift from the channel.

Figure E.3. Ratio of EM to gravitational force along the path of the flow

Experiments were performed in FLIRE for two currents: 1 A and 5 A. For the 1-A case, no visible effects were observed. For the 5-A case, the MHD effect on the flow is evident. A snapshot of the flow with current passing through the lithium along the channel to generate a lifting force on the flow is shown in Figure E.4. The white lines drawn on the photograph indicate the edges of the channel and the location of the current leads. The dashed lines indicate that the wire is immersed in the lithium flow. The flow bulges between the current leads due to the lifting MHD force, but it is not ejected from the channel. A more detailed measurement of the film thickness is necessary to calculate the total lifting force per unit volume on the stream because both the film thickness, and hence the current density passing through the film, change as the flow bulges. However, this simple experiment demonstrates FLIRE's capability to carry out MHD studies, in particular the effect of $\mathbf{J} \times \mathbf{B}$ body forces on retention and diffusivity, on flowing liquid metal PFCs.

Figure E.4. Flow with 5 A running from left to right. In this case, the flow “climbs” higher in the second (to the right in the figure) contact lead, as indicated by the dashed line

F HEIGHTS Modeling

During the normal operation of the high confinement regime (H-mode), the edge-localized modes (ELMs), are serious concern for divertor plasma-facing components (PFCs) of the next generation tokamaks. The periodic relief of edge pressure gradient results in pulses of energy and particle fluxes transported across the Separatrix to the scrape-off-layer (SOL) and eventually to the divertor surface. ELMs could, therefore, result in excessive target erosion, cyclic thermal stresses, and ultimately to shorter divertor lifetime.

To study and simulate the effect of ELMs on Lithium liquid surfaces we use a comprehensive two-fluid model that is developed to integrate SOL parameters of ELMs with divertor surface evolution (melting, vaporization, vapor cloud dynamics, and macroscopic spallation). These models are implemented in the HEIGHTS numerical simulation package. Initial results show that high-power ELMs in future tokamak machines can cause serious damage to divertor components, may terminate plasma in disruptions, and due to large contamination may affect subsequent plasma operations.

F.1 Introduction

Recently, the edge-localized modes (ELMs) are the focus of increasing attention because of potential impact of the high heat pulses on divertor design and lifetime of future tokamaks such as ITER. Importance of ELMs arises from a number of observations such

as to limit energy confinement, provide density control and limit buildup of impurities, broaden scrape-off layer (SOL) density profile, cause large heat pulses on the plasma-facing components (PFCs), and increase sputtering of divertor materials.

The spatial distribution of relative plasma parameters inside the core such as electron and ion temperatures, density, and power is shown in Figure F.1. These parameters and their spatial distribution will determine some of the main characteristics of ELMs.

Figure F.1. Typical spatial distribution of relative electron and ion temperature, density, and power in core plasma

During ELMs, part of tokamak's total plasma energy, Q_{ELM} of ≈ 0.01 - 0.1 of core plasma energy, Q_{core} , is released and deposited on the divertor surface in a duration of ≈ 0.1 - 1 ms with a frequency of ≈ 1 - 20 Hz. The incoming power from SOL to divertor plate in ITER-like devices during an ELM can then increase from ≈ 5 MW/m² to ≈ 300 - 3000 MW/m². The mass losses of divertor materials are strongly dependent on the power deposited. The internal sub-frequency of an individual ELM event should be simulated. Based on experimental data in the literature and our simulation (see below), we estimate that the ELM plasma is ~ 1 - 2.5 keV at the divertor and lasts for about 0.5 milliseconds with a subfrequency of about 100 kHz (50 10 -microsecond bursts). A summary of conditions is listed in Table F.1.

At low power deposition the surface temperature of PFC such as lithium will not exceed vaporization temperature and mass losses due to vaporization may be small. At high Q_{ELM} , however, the resulting high surface temperature causes vapor-cloud formation with similar consequences to plasma disruptions³⁰. Vapor shielding decreases energy

deposition at the surface but increases radiation flux to nearby components. In addition, liquid metal flow instabilities can occur inside lithium surface causing mass losses due to both magnetohydrodynamics and volume bubbles splashing effects and atomic vaporization³¹.

Table F.1. Desired ELM parameters for divertor simulation

ELM Parameter	Value
Power Loading	$\sim 1\text{-}3 \text{ MJ/m}^2$
ELM Event Frequency	$\sim 1\text{-}10 \text{ Hz}$
Total ELM Duration	$\sim 0.1\text{-}1 \text{ ms}$
Blob Subfrequency	$\sim 10\text{-}100 \text{ kHz}$
Blob Pulse Width	$\sim 5\text{-}20 \text{ }\mu\text{sec}$
Plasma Temperature During ELM	$1\text{-}2.5 \text{ keV}$
Plasma Density During ELM	$\sim 10^{19} \text{ m}^{-3}$
Magnetic Field Strength At Divertor	$\sim 1\text{-}5 \text{ T}$
Normal Edge-Region Plasma Temperature	$\sim 10\text{-}100 \text{ eV}$
Normal Edge-Region Plasma Density	$\sim 10^{18} \text{ m}^{-3}$

The mechanisms responsible for the rapid loss of plasma from the core edge during ELMs have not been clearly identified. Some of the mechanisms suggested include parallel convection along suddenly-opened field lines produced by island formation, turbulent radial transport due to electrostatic or electromagnetic fluctuations, or radial convection due to large scale-length potential structures^{32,33}. Therefore, to predict consequences of ELMs such as enhanced mass losses and contamination of core plasma a simplified model of plasma behavior during ELMs was developed and used taking into account features mainly inherent in most current machines.

Duration ELMs in ITER-like machines is expected to be $0.1 < \tau_{\text{ELM}} < 1 \text{ ms}$, however, usually most ELMs duration is near $\tau_{\text{ELM}} \approx 0.5\text{-}1.0 \text{ ms}$ for larger machines. During an ELM, a fraction of tokamak plasma energy, $Q_{\text{ELM}} = \eta Q_{\text{core}}$, escapes to the SOL. This energy can consist of a conduction part due to thermal conduction and a convection part carried by the diffusing particles.

Figure F.2 shows a schematic illustration of typical flowing lithium on top of the divertor plate as the plasma facing material. The liquid lithium is flowing over a structural material such as carbon, Mo, or others. The behavior of the flowing lithium during ELMs is quite important and need to be assessed and evaluated in details to make sure that such a concept is feasible and can be seriously considered in future high power tokamaks.

Figure F.2. Illustration of liquid lithium flow in reactor configurations (e.g., NSTX)

However, to understand the behavior of liquid lithium exposed to an ELM, many physical processes need to be studied in detail. In fact, four different interaction stages need to be integrated self consistently as shown in Figure F.3.

Modeling Stages of ELMs

Figure F.3. Diagram illustrating the requirement needed for ELM modeling

In our first model of the core plasma, it is assumed that the lost core plasma energy to SOL is lost within a region of radii from R_{ELM} to R_s (radius at Separatrix). Therefore, Q_{ELM} is equal to the plasma energy contained between R_{ELM} and R_s . The available experimental data indicates that the dynamics of ELMs have stochastic nature as a result of large-scale (macroscopic) plasma motion. Therefore, it is assumed in our model that the energy and particles losses across magnetic field have a diffusive character with some enhanced diffusion coefficient $D_{\perp} \approx 5 \text{ m}^2/\text{s}$, i.e., about ten times more than the diffusion coefficient during normal operation.

ELMs, most of the times, are made up of high-frequency plasma blobs injected into the edge region and are not steady plasma ejection events. Fine detailed D_{α} observations have shown that indeed each ELM event is actually a series of small plasma blobs that enter the edge-plasma region from the edge of the pedestal (see Figure F.4)³⁴. However, in this analysis, because of the high sub-frequency of the blob ELM, it is assumed that these sub ELMs are continuous with total energy and pulse duration equal to the sum of the sub ELMs.

Figure F.4. Edge probe measurements on the DIII-D reactor at General Atomics showing ELM events³⁴

F.2 Model of SOL during ELMs

According to our first assumption one can find the number of particles (DT ions), N_{ELM} , corresponding to an energy Q_{ELM} that escape to SOL during an ELM:

$$\begin{aligned}
 Q_{ELM} &= \int_{R_{ELM}}^{R_s} \frac{3}{2} k (T_i + Z_{eff} T_e) n_i(r) 2\pi R \cdot 2\pi r \cdot dr = \eta Q_{core} \\
 &= \frac{3}{2} k T_{mean} (1 + Z_{eff}) \cdot N_{ELM}, \quad T_{mean} = \frac{2 Q_{ELM}}{3 k (1 + Z_{eff}) N_{ELM}}
 \end{aligned}
 \tag{F.1}$$

where T_{mean} is the average temperature of ions and electrons in this region.

Because of the large uncertainties in ELMs physics it is more appropriate to use values that are measured directly such as τ_{ELM} rather than $D_{\perp}$ and the total energy deposited onto the divertor plate, Q_{load} .

Figure F.5 shows the predicted ELMs relative parameters as a function of radial position starting from the Separatrix ($R_s = 200$ cm) and going inward to R_{ELM} towards the center. For example, for $Q_{\text{ELM}} \approx 1\%Q_0$ (i.e., $\eta = 0.01$) it corresponds to a radius $R_{\text{ELM}} = R_s - \Delta R_{\text{ELM}} = 200 - 16 = 184$ cm, $N_{\text{ELM}} \approx 0.05N_0 = 0.2 \cdot 10^{22}$, and $T_{\text{mean}} \approx 1.0$ keV. For $Q_{\text{ELM}} \approx 10\%Q_0$ ($\eta = 0.10$), this corresponds to radius $R_{\text{ELM}} = R_s - \Delta R_{\text{ELM}} = 200 - 36 = 164$ cm, $N_{\text{ELM}} = 0.2N_0 = 0.9 \cdot 10^{22}$, and $T_{\text{mean}} \approx 2.4$ keV. The main values used for $T_0 = 10$ keV, $n_0 = 10^{14} \text{ cm}^{-3}$, $Q_0 = 0.13 \cdot 10^9$ J, and $N_0 = 4.0 \cdot 10^{22}$ for an ITER-like device. Figure F.6 shows spatial distribution of incident plasma parameters along divertor plate during an ELM of 3 MJ/m^2 energy density.

Figure F.5. Predicted ELM relative parameters as a function of radial position from Separatrix

Figure F.6. ELM relative parameters as a function of distance along diverter plate

The large increase in both particle and heat flux in ξ times higher than the normal operation, i.e., $\xi = \eta \cdot \tau_E / \tau_{ELM} \gg 1$, will result in significant increases in mass losses of diverter plate (vaporization, sputtering, brittle destruction, and liquid splashing). To predict these losses and potential contamination of core plasma, two problems must be solved, i.e., dynamics and structure of particles in SOL and the interaction of particle/heat fluxes from the SOL with diverter plate materials.

During normal operation the plasma in SOL is highly collisional, i.e., the path length, λ , of ions and electrons of $T \approx 100$ eV is less than the connection length $L_{con} = 2L_{||}$ ($L_{||}$ is the parallel distance between the two diverter plates), $\lambda \ll 2L_{||}$. However, during ELMs, $\lambda(T_{mean} > 1 \text{ keV})$ is much larger than the connection length, $\lambda \gg 2L_{||}$, and therefore, the SOL plasma during ELMs is collisionless and requires different treatment than the behavior during normal operation.

A “collisionless” plasma does not mean that collisions can always be neglected. The term is used only to indicate that the ions and electrons path length is much greater than the connection length, but large part electrons oscillating between plates located at distances much shorter than particle path length (collisionless in space) will have lifetimes determined by collisions (collisional in time). Thus, the SOL during ELMs is similar to the situation of SOL during normal operation regime with enhanced confinement^{32,33} or during a disruption³⁵.

One main feature of a collisionless SOL plasma is the edge plasma acts as an electrostatic trap for electrons, since electrons which originally have parallel energy that is lower than the wall potential energy, ϕ , will be trapped between the inner and outer diverter plates. To obtain the potential ϕ and corresponding net heat flux of ions and electrons to the

divertor plate one can make use of previous models ³⁶. For conditions of the SOL during ELMs, the negative potential, ϕ , at the boundary between the SOL and the ionized vapor cloud near the divertor surface is $|e\phi| \geq kT_e$ where T_e is electron temperature at the corresponding magnetic field line. Ions and electrons with energy parallel to magnetic field lines $E_{\parallel,e} > \phi$, leave the SOL directly (escaping particles). A simple model based on similar physical processes described in Ref. 36 is used to calculate particle fluxes from SOL to divertor surface.

F.3 Particles and Energy Fluxes

The ions escaping the SOL with increased parallel energy, $E_{\parallel,i}$, due to acceleration in potential ϕ will have increased velocity U_i , and decreased density n_i . The ion particle flux and ion heat flux, W_i , leaving the SOL is the same as in the absence of potential. However, the ion heat flux, W_{id} , reaching the evolving vapor-cloud above divertor surface increases due to acceleration in z-direction due to the potential jump. Electrons escape the SOL with a parallel energy $E_{\parallel,e} > -e\phi$, thus the average velocity U_e , density n_{ed} , total flux of escaping electrons S_e , and corresponding heat flux, W_e , are determined by the potential ϕ . The ion heat flux increases by ΔW_ϕ because of acceleration in the negative potential, ϕ , created by electrons. Because fluxes of electrons and ions are assumed equal, the density of escaping electrons is less than density of ions in the SOL. Neutralization of ions charge is achieved not only by the escaping electrons but also due to trapped electrons and cold electrons coming from the vapor cloud above divertor surface. Therefore, electron and ion densities in SOL are equal. Thermalization of the cold electrons during their flight in SOL between divertor plates can be achieved due to binary and turbulent collision ³⁶.

F.4 Interaction of incident particles with divertor plate

The power, W_{SOL} , from the SOL to the divertor plate for ITER-type machine can be estimated from the following consideration. During an ELM power of energy release from the core plasma to the SOL increases in >100 times. During an ELM duration time $\tau_{ELM} \approx 1$ ms part of total tokamak core plasma energy $\eta = (0.01-0.1)$ releases. Therefore, for regime with energy power to the divertor plate $w_E = 5 \text{ MW/m}^2 = 0.5 \text{ kW/cm}^2$, $\tau_E \approx 5.8$ s, $Q_E = 3 \text{ kJ/cm}^2$ the power, w_{ELM} , and energy load, Q_{ELM} , increase to

$$\begin{aligned}
w_{ELM} &= \eta w_d \frac{\tau_E}{\tau_{ELM}} = 0.5 \cdot 10^3 \frac{5.8}{1.0 \cdot 10^{-3}} = 3.0 \cdot 10^6 \cdot \eta \\
w_{ELM}(\eta = 0.1) &= 300 \frac{kW}{cm^2}, Q_{ELM}(\eta = 0.10) = 300 \frac{J}{cm^2}, \\
w_{ELM}(\eta = 0.01) &= 30 \frac{kW}{cm^2}, Q_{ELM}(\eta = 0.01) = 30 \frac{J}{cm^2}
\end{aligned} \tag{F.2}$$

The flow velocity, $U_0 \approx 10$ m/s, is rather small that flow time, $\tau_u = L_d/U_0 \approx 10$ ms, is much large than ELMs time, $\tau_u \gg \tau_{ELM}$, thus motion of liquid metal during ELMs can be neglected.

The target surface temperature, T_s , can be roughly estimated as

$$T_s(t) = T_0 + \delta T_{ELM}, \quad \delta T_{ELM} = \frac{Q_{ELM}}{c_p h}, \quad h = \sqrt{2\chi\tau_{ELM}}. \tag{F.3}$$

For Li with $C_p = 4.35$ J/cm³, $\kappa = 0.498$ W/cm·K, coefficient of temperature diffusion $\chi = 0.11$ cm²/s that results in skin-depth $h = 0.015$ cm for $\tau_{ELM} \approx 1$ ms. Thus, temperature increase is: $\Delta T_s(\eta = 0.01) = 460$ K, $\Delta T_s(\eta = 0.10) = 4600$ K. Therefore, at $\eta > 0.03$ detail vaporization should be taken into account even if vapor shielding occur.

The temperature distribution along divertor plate (X-direction) is inhomogeneous even during normal operation because of the inhomogeneous incoming power from the SOL, and also due to lithium flow. Incoming power $W(x,t)$ is removed by liquid flow with velocity along divertor plate surface. By neglecting of heat conduction in X-direction in comparison with heat conduction normally to divertor plate surface in Y-direction,

$\frac{\partial}{\partial x} / \frac{\partial}{\partial y} \approx \frac{L}{L_x} \ll 1$ the temperature distribution can be found from equation of heat flux

$W(y,t)$. For homogeneous heat load $W(x) = W_0$ solution has the form:

$$\frac{\partial W}{\partial t} = a^2 \frac{\partial^2 W}{\partial y^2}, \quad W(y,t) = -\kappa \frac{\partial T}{\partial y}, \quad \chi = a^2 = \frac{\kappa}{c_p}, \tag{F.4}$$

$$W(y,t) = W_0 \frac{2}{\sqrt{\pi}} \int_0^{\frac{y}{2a\sqrt{t}}} \exp(-\xi^2) d\xi = W_0 \left[1 - \Phi\left(\frac{y^2}{4\chi t}\right) \right] \tag{F.5}$$

where κ is the heat conductivity, c_p is the specific heat, $\chi = a^2$ the temperature diffusivity, $W(x)$ is the heat flux to the divertor plate surface. Temperature is then given by

$$T = T(y,0) + \Delta T = \frac{W_0}{\kappa} \frac{2a\sqrt{t}}{\sqrt{\pi}} \exp\left(-\frac{y^2}{4\chi t}\right) - \frac{W_0}{\kappa} y \left[1 - \Phi\left(\frac{y^2}{4\chi t}\right)\right] \quad (F.6)$$

where $T(y,0)=T_0$ is the initial temperature of coming flow. The surface temperature, $T_s=T(0,t)$, is

$$T_s(t) = T_0 + \delta T \sqrt{\frac{t}{\tau_u}}, \tau_u = \frac{L_x}{U_0}, \delta T = \frac{4}{\sqrt{\pi}} \frac{W_0 \tau_u}{c p \lambda_u}, \lambda_u = \sqrt{2\chi \tau_u}, \quad (F.7)$$

$$T_s(x) = T_0 + \delta T \sqrt{\frac{x}{L_x}} \quad (F.8)$$

where δT is the increase of surface temperature for flight time τ_u at heat flux W_0 , λ_u is the diffusion depth for the flight time.

For $L_x = 20$ cm and $W_0 = 5$ MW/m², $U_0 = 20$ m/s $\tau_u = 10$ ms, $\lambda_u = 0.048$ cm, $\delta T_0 = 54$ K. For ELM with $\eta = 0.01$ and $W_{ELM} = 300$ MW/m² $\delta T_{ELM} = 1029$ K and heat diffusion depth for 1 ms, $h = 0.015$ cm. After ELM, heat diffuses into the bulk of flowing liquid metal flow with decreasing of the temperature to $T_0 + \delta T_{after}$, $\delta T_{after} \approx 100$ K at $t = 10$ ms.

The decrease in surface temperature due to heat diffusion into bulk is estimated assuming that heat propagation is similar to that of instant deposition of energy $Q(x, t=0)$. For time $t > \tau_{ELM}$ distribution of temperature is given by:

$$T(x, t) = \frac{T_{s0}}{\sqrt{1 + \frac{t - \tau_{ELM}}{t_{act}}}}, \quad (F.9)$$

where T_{s0} is the surface temperature at $t = \tau_{ELM}$, and t_{act} is corresponding time at which the actual surface temperature, T_{s0} , equals to the surface temperature after instant deposition of energy on flow surface

$$T_{s0} = T_s(x, t = \tau_{ELM}) = \frac{2Q(x, t = \tau_{ELM})}{c p \sqrt{4\pi\chi t_{act}}}, t_{act} = \frac{1}{4\pi} \left(\frac{2Q}{c p T_{s0}} \right)^2. \quad (F.10)$$

Due to motion, heated liquid is renewed by fresh low temperature liquid from inlet nozzle or inlet location.

The integrated HEIGHTS package that solves detail particle energy deposition, evolution of surface materials, debris formation, vapor radiation magnetohydrodynamics, and erosion physics is enhanced and being used in this analysis. The enhancement includes development of two-fluid hydrodynamic mixing model where the incident DT plasma is treated separately from the eroded debris cloud of divertor materials (see Figure F.7). The forward-reverse radiation transport method for both line and continuum radiation with detail line resolution of vapor plasma is used. Parametric studies can be made for different ELM duration (from 0.1 to 1 ms) and different ELM intensity (1-10% Q_0). Lithium as the main potential divertor materials is analyzed in this work.

Calculations are made for each flux line that strikes the divertor plate to have two-dimensional components. Flux lines are distinguished by their distance from the strike point where the tokamak Separatrix strikes the divertor plate at $r = 0$. Input parameters are chosen assuming that the SOL width expands 10 times its size at the divertor surface. The effective size of the SOL at the mid-plane is calculated to be 1.975 cm for ions and 0.47 cm for electrons.

3.6 MW/cm², and at the strike point is 0.36 MW/cm². For electrons, the maximum energy density is 0.95 MW/cm² at the midplane and is 0.095 MW/cm² at the strike point. The ion energy at the strike point $E_i = (1+\psi) T_{\text{mean}} = 12.5$ keV and the electron energy at the strike point $E_e = T_{\text{mean}} = 2.4$ keV. The flux at the strike point $S_{\text{ELM}} = n_0 V_i(T_{\text{mean}}) = 0.67 \cdot 10^{22} \text{ cm}^{-2} \text{ s}^{-1}$. Therefore, analysis of ELMs is actually similar to disruption physics and analysis but with lower total deposited energy and lower incoming particle kinetic energy (about 2 keV instead of 10 keV). The density of incoming DT particles stopped in vapor cloud above the surface can then be comparable or exceed the vapor density of divertor surface, and therefore, two-fluid mixing model is developed. Thus, the front part of cloud can mainly consist of stopped DT particles. All other detail models and physics of plasma-material interaction during a disruption are included in this analysis³⁷.

Figure F.8 shows liquid Li response to an ELM with different % of Q_0 deposited in duration of 1 ms. The erosion from Li vaporization increases significantly with deposited energy because of exponential character of vaporization rate. At higher ELM energy, more than 90% of incident energy is absorbed by vapor cloud and 80% is radiated to nearby areas. Therefore, vapor shielding is also effective during ELMs. The temperature of cloud front is high ≈ 40 -50 eV because the front part of the cloud consists of DT ions. The plate surface temperature is rather high > 2000 K that results in high erosion rate because saturation pressure exceeds vapor cloud pressure of few atm. Vapor expansion above divertor surface also depends on deposited energy.

Figure F.8. Lithium erosion and expansion as a function of % energy in ELMs

The critical factor is net threshold power to divertor surface that determines the surface temperature at which the saturation pressure exceeds the cloud pressure of few atm. The cloud pressure is determined, however, by DT flux momentum and diffusion across the magnetic field. Because the front of the cloud consists mainly of DT with high conductivity this results in less diffusion and, consequently high cloud pressure. For an

ELM with $\eta = 0.1$ (3MJ/m^2), the erosion rate and the expansion of vapor cloud front are high. Lithium vapor can reach the X-point that can result in contamination of core plasma and terminate the plasma in a disruption. At ELM power below the threshold the erosion rate is tolerable and Li vapor expansion is <10 cm, and plasma contamination is low.

Because the temperature of divertor surface can exceed the threshold for splashing during high power ELMs, macroscopic losses in form of liquid droplets can take place. In case of well-confined vapor cloud without turbulence in the cloud, these droplets vaporize in cloud by radiation and additionally shield the divertor surface, therefore, the erosion rate may not significantly increase³⁷. However, if vapor turbulence exists, the erosion-rate can increase substantially and plasma contamination/termination is a serious problem. This problem will be studied in detail for the next report.

Figure F.9 shows lithium plate erosion and vapor expansion as a function of ELM duration for an energy of $10\%Q_0$. It can be seen that for $\tau_{\text{ELM}} \approx 0.1$ ms, the erosion rate and the DT cloud front expansion increases significantly reaching $h \approx 20$ μm and $X_{\text{DT}} \approx 50$ cm. It can also be seen that most cloud volume (60%) is contained by DT, and lithium vapor expands to distance ≈ 20 cm for powerful ELMs ($10\%Q_0$) with $\tau_{\text{ELM}} = 0.1$ ms. This may cause core plasma contamination. Therefore, lithium expansion is a very serious problem and must be studied in detail to make sure that the expanding vapor does not terminate the plasma in to a disruption.

Figure F.9. Lithium erosion and expansion as a function of ELM duration

F.5 Summary

The edge-localized modes (ELMs) may be a serious concern for plasma-facing components during normal operation of the next generation tokamaks. To study this

problem, a two-fluid model is developed to integrate SOL parameters during ELMs with divertor surface evolution (melting, vaporization, vapor cloud dynamics, and macroscopic erosion) using HEIGHTS numerical simulation package.

Results of initial simulations can be summarized as the following:

1. During ELMs mainly outer layers of the tokamak core plasma (nearby separatrix) is transported to the SOL. Therefore, plasma coming to the SOL has low temperature, T_{mean} , and larger density than the core plasma. Number of incoming particles and T_{mean} are determined by magnitude, η , of the ELM increasing with η .
2. The temperature T_{mean} of incoming plasma is close to the core plasma temperature nearby the separatrix and is about an order of magnitude less than the core plasma temperature of $T_0=20$ keV.
3. Density of SOL plasma decreases with distance from the separatrix with characteristic width, λ_i , determined by effective diffusion coefficient. Temperature of the SOL ions is constant across the SOL, but electrons temperature of the SOL decreases with distance from the separatrix more sharply than density with characteristic width, $\lambda_e \approx 0.25 \cdot \lambda_i$. Incoming power from the SOL to divertor plate decreases with characteristic width intermediately between λ_e and λ_i .
4. Effective shielding of the target surface from incoming particles takes place, particularly at higher ELM power. It is due to lower energy of incoming particles having origin near the Separatrix.
5. Due to this shielding, most incoming energy is spent in cloud formation, radiation of impurities, heating divertor plate, and work against magnetic field. At low ELM power, the total erosion becomes very small, vapor cloud expands up to distance of 10 cm or less, and contamination of core plasma becomes also low. This is due also concentration of impurities nearby the target surface.
6. At higher ELM power, large vapor expansion leads to plasma contamination and possible termination in a disruption even in renewable surface materials such as lithium where erosion is not a problem.
7. Excessive erosion from vaporization and macroscopic particles/droplets formation may affect subsequent plasma operations and lead to much shorter lifetime.

G Conclusions

Helium retention in flowing lithium was measured by FLIRE. The retention coefficient of helium in liquid lithium flowing at 44 cm/second down a 10-cm ramp is between 0.1% and 2%, for energies between 500eV and 2keV. The retention varies linearly with energy (as expected) over the range of energies considered, and the scaling of retention with flow velocity fits the expected square root of velocity expected by theory. This measure

of retention is for helium that is quickly release from the flow (prompt release), which dominates the amount of retained helium; however, long-term trapping, which is much smaller, is also of interest.

Long-term helium retention in liquid lithium was demonstrated by heating a sample of lithium after measuring its short-term helium retention. Above a threshold temperature of approximately 300°C, helium bubble bursts were detected, indicating that helium had been trapped in the liquid lithium, or on the surface within the measurement chamber. After successive thermal annealing cycles of the lithium charge, the bubbles ceased, indicating that the long-term retained helium was removed from the charge.

A diffusion model of the helium in the flowing liquid allowed the helium diffusion coefficient in the flowing stream to be back-calculated from the measured retention coefficient. The result is a helium diffusion coefficient of $\sim 4 \times 10^{-7} \text{ cm}^2/\text{s}$. This value is much lower than the expected value of $\sim 10^{-4} \text{ cm}^2/\text{s}$, which may be due to mixing effects in the flow, bubble formation and/or surface film formation acting as a diffusion barrier.

Thermal desorption spectroscopy (TDS) measurements indicate that deuterium is absorbed into the lithium flow directly from the background gas in the implantation chamber, even if no ion beam is present. To avoid problems with background hydrogen concentration, deuterium gas is used as a substitute. Two cases were examined: one with a high deuterium gas pressure (1 Torr) and one with a low deuterium gas pressure (5×10^{-5} Torr). In both cases, the TDS measurement indicates a hydride concentration of 0.1-0.2%. This suggests that the rate determining step for deuterium absorption is dissociation of deuterium molecules on the lithium surface, which absorbs deuterium atoms, not molecules. Because the background absorption rate is much higher than the ion current from an ion gun, a deuterium plasma source was built to expose the stream to more deuterium ions and atoms.

MHD experiments were carried out on the FLIRE flow. Two electrical leads were placed in the path of the flow, and two permanent magnets were placed on either side of the flow ramp, forming a region where $\mathbf{J} \times \mathbf{B}$ forces on the flow could be generated and controlled by the current passing through the electrical leads. Both lifting and pressing forces were tested. The current required to exceed the gravitational force was calculated and applied to the flow, and the flow did not leave the channel. This result is likely due to two factors: 1) the high surface tension of the liquid lithium and wetting of the ramp surface provided an additional restoring force, 2) the flow “bulged” in the magnetic field region, spreading out the current density and lowering the body force on the fluid. This latter effect was not taken into account because the calculation of an equilibrium current density distribution and the resulting true body force was too complicated for the scope of the FLIRE project. The purpose of these experiments was to demonstrate FLIRE’s capability to carry out MHD studies with flowing liquid lithium, in addition to retention and diffusivity measurements.

The semianalytical model used to estimate the helium diffusion coefficient in the flowing lithium stream from retention data has been improved over the course of this project.

More-detailed calculations, each focusing on a different assumption of the initial simple model, have shown that the initial model yields reasonable results when experimental error is considered. Specifically, new models have investigated the effect of a continuous implantation profile, a finite ion-beam width, finite flow depth, and pooling between the flow ramps. The results of each of these is discussed in detail in the Appendix. More detailed modeling work has been carried out using the HEIGHTs package at Argonne National Laboratory to investigate applications of flowing lithium to fusion reactor environments for this project.

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