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Oxidation of lithium plasma facing components and its effect on plasma performance in the lithium tokamak experiment-β

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Abstract
The characteristics of lithium-coated plasma-facing components (PFCs) have been correlated with plasma performance on the lithium tokamak experiment-β (LTX-β). Previous experiments on LTX showed that the application of lithium to PFCs was needed to achieve higher performance discharges with flat electron temperature profiles and high edge temperatures. Samples that match the LTX-β PFCs were exposed to plasmas before and after PFCs were coated with lithium and transferred under vacuum to a surface analysis station. Measurements using x-ray photoelectron spectroscopy (XPS) revealed that the primary surface constituent was lithium oxide. Earlier XPS analysis of lithium-coated PFCs on LTX was only able to show the presence of surface oxygen. The new XPS data from LTX-β have sufficient resolution to clearly identify lithium compounds for the first time, and enable them to be correlated with how lithium-coated PFCs can reduce impurities and retain hydrogen to reduce recycling.

Keywords: lithium, plasma surface interactions, plasma confinement, plasma diagnostics, tokamaks

(Some figures may appear in colour only in the online journal)

1. Introduction
Low-Z plasma facing components (PFCs) are an alternative to high-Z PFCs such as tungsten [1]. An advantage these PFCs offer is low radiative loss fraction compared to the high Z alternative [2–4]. The material erosion for low Z solid PFCs can be compensated for by replacing them with liquid metal PFCs, for example, that can be replenished by a closed loop of flowing liquid metal [5, 6]. Amongst low Z PFCs, lithium is an attractive option for reasons that include its ability to retain hydrogen to enable access to a low recycling operating regime [7–10].

The lithium tokamak experiment is the only tokamak in the world program that has the ability to operate with nearly complete coverage of lithium as the primary PFC [11]. The LTX recently demonstrated that a tokamak with evaporatively deposited lithium films can lead to low recycling operation [12]. The LTX was recently upgraded to LTX-β, which
The LTX-β vacuum vessel docks two lithium evaporators that, when fully inserted, sit at their respective poloidal midplane center. The evaporators are installed on diametrically opposite ports to provide near full coverage of lithium on the stainless steel shells [14, 17]. Lithium evaporation rate from each lithium evaporator is monitored using a Quartz Crystal Microbalance (QCM). The QCM monitoring the evaporator with a line-of-sight to the SEP recorded a lithium deposition of 140 nm thickness. Using a simple inverse square relationship, 140 nm of lithium on the QCM translates to roughly 500 nm of lithium on the SEP sample head.

The stainless steel PFCs on LTX-β were initially coated with lithium as described in earlier work [14]; following this campaign, the LTX-β vacuum vessel and the SEP were vented to dry argon for 2 weeks before they were pumped out again. This means that both the SEP sample head and the LTX-β PFCs had passivated lithium coatings on them after the vent. Following the vent, the SEP sample head was heated to 200°C for a few hours in ultra-high vacuum, allowed to cool down and then docked onto LTX-β. LTX-β shells were also baked at 200°C for about 6 h. LTX-β shells were allowed to cool down to room temperature, and the SEP sample head inserted as shown in figure 1 before a series of shots were taken (101 317–101 349). After these shots lithium was evaporated on the shells and the SEP sample head using the LTX-β lithium evaporators. Although, an XPS scan was not recorded post lithium evaporation, the state of the SEP sample head after evaporation can be inferred from earlier data [14]. Previous XPS scans show the presence of metallic lithium in XPS scans (top ~6 nm), immediately after lithium evaporation. As the sample builds residual vacuum exposure in the low water vapor environment of LTX, it is expected that the oxide layer on top of lithium grows in thickness [14, 16].

After the evaporation, the SEP sample head, along with the LTX-β vacuum vessel, saw a week of residual vacuum exposure at typical vacuum vessel pressures ($6 \times 10^{-8}$ to $9 \times 10^{-8}$ Torr). The residual vacuum exposure was followed by a sequence of about 40 shots (101 355 to 101 395). The time between lithium evaporation and plasma operation should have been sufficient to oxidize most of the top 6 nm of the evaporated lithium [14].

XPS scans of the SEP sample were taken right before the probe was docked to LTX-β, and the SEP was transferred back to the Surface Analysis Station 12 h after the last shot (101 395) to take scans again. Pre-evaporation survey spectra show a larger carbon feature compared to post-evaporation (16% of total surface composition as opposed to 4%).

Regional scans for Li(1s), O(1s) and C(1s) are shown in figure 2. Although pre-evaporation scans contained larger amounts of carbon, the region of the narrow scan that should correspond to Li$_2$CO$_3$ did not see a large feature. It is possible that a carbonate feature can be fit to the Li(1s) and O(1s) scans of the sample. This would only improve the fit marginally. Pre-evaporation, Li$_2$O and LiOH both constitute a significant fraction of the surface. The prime constituent after Li evaporation and plasma exposure, as seen earlier [14], is Li$_2$O. Pre-evaporation the O(1s) signal constitutes of 57% Li$_2$O and 42% LiOH, whereas post evaporation and plasma exposure the O(1s) signal corresponds to 85% Li$_2$O and only 14% LiOH. The choice of peak assignments and their respective binding energies are discussed elsewhere [14].

Additionally, for both pre-evaporation and post-evaporation with plasma exposure scans, the O(2s) region was scanned. This was done to look at the relative intensities of the hydroxide vs. oxide feature in O(1s) and O(2s) for

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**Figure 1.** Toroidal (top) half section view showing the SEP sample head and the Langmuir probe tip flush with the shells at the low field side. The image also shows the filterscope fiber-view to the SEP.
Figure 2. Regional XPS scans of Li(1s), O(1s) and C(1s) peaks; first, second and third column from the left, respectively, of the SEP sample head before and after fresh Li evaporation and plasma exposure in LTX-β. The carbon levels shown in the third column from left, for example, are reduced by a factor of four after lithium evaporation. In the pre-lithium evaporation XPS data shown in top row, primary surface constituents are Li 54%, O 29%, C 16%; bottom row surface constituents are Li 65%, O 31%, C 4%.

3. Dependence of plasma performance on PFC conditions

Based on the results of the PFC surface analysis, the dependence of plasma performance on PFCs conditions becomes a comparison of discharges with PFCs that differ significantly in their ratios of lithium hydroxide to lithium oxide. On LTX-β a plasma current Rogowski coil measures the plasma current; a 1 mm interferometer measures the plasma current; a 1 mm interferometer measures line-integrated plasma densities along the poloidal midplane. The integrated emission intensities are measured by a filterscope fiber looking at the SEP. The fiber was made to focus on the SEP sample head such that the view through the fiber was smaller than the probe area (figure 1). This was done to sample emissions primarily from the surface of the probe. The fiber was split into three 3 channels, looking at Hβ = 486 nm, LiII = 548.6 nm and OII = 441.6 nm. Li and O emission lines were chosen since they are the prime surface constituents, and Hβ was chosen instead of Hα because its filterscope channel had a better signal-to-noise ratio. The line integrated-emission intensity is directly proportional to the signal measured by the filterscope. The constant of proportionality, i.e. the ratio of line integrated-emission intensity to measured signal was determined by recording the measured signal while exposing the filterscope to light from a source with known spectral radiance.

Figure 4 shows key plasma performance indicators for two representative shots, 101 346 and 101 395. 101 346 was a day before lithium evaporation and 101 395 was 8 days after lithium evaporation. These shots were chosen in particular since they had closely matching line integrated electron densities. Fueling was terminated for both shots at about 455 ms, this is followed by a reduction in plasma density and some evidence of hotter edge temperatures for 101 395. The flat temperature profiles measured on LTX earlier [12]
Figure 4. A comparison of key plasma parameters between shots representative of before—101 346, represented by the red trace, and after lithium evaporation—101 395, represented by the blue trace (a) plasma current, (b) line integrated electron density, (c) ion saturation flux to the single Langmuir probe, (d) line integrated H\(_{\beta}\) emission from the SEP sample head, (e) electron temperature at the single Langmuir probe head, (f) line integrated O\(_{\beta}\) emission from the SEP sample head, (g) electron density measured by the single Langmuir probe, (h) line integrated Li\(_{\beta}\) emission from the SEP sample head. Additionally, the duration for which the gas puff valve is opened for each shot is denoted by vertical bands, blue—101 395 and red—101 346.

were similarly observed after fueling was terminated. This is likely because the plasma is fueled by a high field side gas puffer, which is a source of cold neutral gas at the edge of the plasma. Terminating the gas puff under low recycling conditions is expected to produce a hot edge \[7\].

Plasma equilibrium reconstructions using PSI-TRI \[18\] show little difference in the way the plasma geometry evolves. Both 101 346 and 101 395 start with a large plasma volume and limiting on the low field side, and eventually moving towards the high field side and collapsing on the high field side midplane limiter. A key difference between the two run days was that 2.5–3 times more gas had to be pumped in after lithium evaporation to match the same line integrated electron density. In fact, gas puff programming identical to shot 101 346 was unsuccessful in initiating a break-down post lithium evaporation. Figure 5 compares gas puffs for two run days with gas calibration shots, or shots that had the same gas puff programming as a plasma shot but a breakdown was not initiated.

Gas calibration shots for 101 346 and 101 395 were not available; two corresponding shots were chosen for the same run. Gas puff programming for pre lithium shot 101 328 and its gas calibration shot 101 330 are shown in figure 5 on the left. The plasma on 101 328 quenched at 0.465 seconds. At the instant the fast ion gauge read a difference of less than a percent between 101 328 and 101 330. The plots for the hydrogen pressures thus nearly overlay, and the small difference between the two is reflected in the purple trace (with legend designation ‘Retained’) in figure 5. However, for post lithium (8 days old) shot 101 382 and its corresponding gas calibration shot 101 381, the difference at the end of the shot at 483 ms is \(\sim 85\%\). This higher retention of hydrogen is shown on the right side of figure 5.

4. Discussion

The O\(_{\beta}\) and Li\(_{\beta}\) line integrated emission intensities were tracked for all the shots for the two run days corresponding to before lithium and 8-day-old lithium. Figure 6 represents the trends for the fiber focused on the SEP. The line integrated emission intensities are normalized by line-integrated electron densities to normalize the effect of emission from the plasma volume. A strong reduction in the O\(_{\beta}\) emission was seen in shots after lithium evaporation, and an increase by an order of magnitude was observed in the Li\(_{\beta}\) intensities. Additionally, a further reduction in the O\(_{\beta}\) emission through shots after initial lithium evaporation can also be seen in figure 6, possibly due to the continued retention of oxygen from water vapor in the lithium films; this is supported by a suppressed water vapor peak (18 amu) as recorded by the LTX vessel Residual Gas Analyzer (RGA). The weakened O\(_{\beta}\) plasma emission was observed along with a large Li\(_2\)O peak in XPS scans. This suggests that the presence of Li\(_2\)O on the surface leads to reduced oxygen emission, when compared to passivated lithium that contains other compounds. The H\(_{\beta}\) signal over the same period for the two classes of shots normalized by the line integrated
Figure 5. Hydrogen gas evolution following gas puffs with (orange trace) and without (red trace) plasma and their difference (purple traces). The timing and duration of the gas puffs are indicated by the short vertical bars. The duration of the plasma shot is highlighted by the width of the vertical red band. The left plots compare shots with gas only (101 330) and plasma (101 328) before lithium evaporation, and they are nearly identical. The right plots compare shots with gas only (101 381) and plasma (101 382) after lithium evaporation, and their difference is consistent with hydrogen pumping during a discharge.

Figure 6. \( \text{O}^{\text{II}} \) and \( \text{Li}^{\text{II}} \) line integrated emission intensities normalized by line integrated electron densities (in S.I. units), show a persistent reduction in oxygen emission after lithium evaporation, for a lithium surface that is mostly \( \text{Li}_2\text{O} \). The values are averaged over first 10 ms of each discharge, since these discharges evolved similarly for the first few milliseconds.

electron density was relatively flat. Strong \( \text{O}^{\text{II}} \) emission is also evident in figure 4(f), (column 2, row 3) before lithium evaporation [19].

The simplest assumption for hydrogen retention is that H ions incident on \( \text{Li}_2\text{O} \) should be retained as LiOH [20]. But there are multiple competing processes that complicate this simple picture. For instance, there seems to be some time dependence to hydrogen retention in LTX [15]. Hydrogen is seen to outgas on the timescale of seconds after a shot. This could be related to hydrogen diffusivity in Li and \( \text{Li}_2\text{O} \). Since it takes a few hours to physically move the SEP to the Surface Analysis Station [13], it is difficult to ascertain the effect of this time dependence using the SEP. It has been shown, using Quantum Classical Molecular Dynamics simulations, that the diffusion coefficient of H in Li reduces as the H concentration in Li increases [21]. Modeling of H diffusivity in \( \text{Li}_2\text{O} \) and LiOH using Density Functional Theory based molecular dynamics simulations is in progress and will help shed light on this behaviour.

If we assume the LTX high temperature edge extends all the way to the wall, it would imply ion impact energies anywhere between a few hundred eV to about a keV (3 times the sheath potential – edge electron temperature). At these energies H ions may be penetrating the thin \( \text{Li}_2\text{O} \) overlayer and interacting with pure Li forming LiH. LiH is unresolvable with XPS but can be quantified for bulk samples using temperature programmed desorption (TPD). The SEP is designed to enable TPD studies of its sample head. Efforts to commission this capability for SEP on LTX-β are under way.

It is also possible that the H plasma is reducing Li in \( \text{Li}_2\text{O} \) and LiOH, thereby freeing up more Li metal sites that bind H in Li-H. To diagnose such behavior, we need a PFC characterization diagnostic that can measure elemental and chemical abundances, in-situ, preferably during a discharge with high temporal and spatial resolution. This presents an opportunity, and the invention of a diagnostic that can do all these things simultaneously will likely make it possible to help identify all the processes responsible for improving plasma performance.

Earlier surface science experiments have shown that \( \text{Li}_2\text{O} \) can retain hydrogen at rates comparable to pure lithium films in vacuum [20]. The retention data presented in figure 5 is consistent with that picture. LTX-β is presently developing capabilities for shot-averaged global recycling measurements, based on neutral particle modeling and Ly-\( \alpha \) emissions, since Ly-\( \alpha \) is less prone to reflections from PFC surfaces compared to visible light lines such as H-\( \beta \) [22, 23]. Based on this information, it is hypothesized that global particle recycling will not change substantially as \( \text{Li}_2\text{O} \) grows on fresh lithium surfaces. Rather, a more significant increase in recycling is expected...
when sufficient time passes after lithium evaporation for sufficient LiOH to accumulate on the surface.

We observe by direct measurement that Li$_2$O is the primary surface constituent for both the passivated and relatively fresh lithium coatings, but with the former having a higher concentration of LiOH. Additionally, the retention of oxygen in lithium oxide is consistent with spectroscopic measurements indicating reduced oxygen emission in plasma following lithium evaporation. The data presented here support the possibility that PFCs which are predominantly lithium oxide on the surface can reduce recycling and improve plasma performance; this is demonstrated using comparisons of hydrogen retention and discharge behaviour before and after lithium evaporation.

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