



Measurements and modeling of D, He and Li sputtering of liquid lithium

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Abstract

The absolute sputtering yields of D^+ , He^+ and Li^+ on solid lithium have been measured and modeled at low energies in the ion-surface interaction experiment (IIAX). The experiment has been extended to measure physical sputtering from liquid lithium surfaces bombarded by D^+ , He^+ and Li^+ . A Colutron ion source is used to create and accelerate gaseous or metal ions onto the liquid metal target. A plasma cup removes any oxides and saturates the surface with deuterium. A small high-temperature, HV substrate heater is used to heat the 0.76 g lithium sample past its melting point to 200°C. Upon melting, a thin oxide layer is formed on the exposed lithium surface, which is cleaved by an in situ arm rotated in front of the target. Results suggest that the absolute sputtering yield of lithium is less than unity. In addition, the behavior of liquid lithium self-sputtering suggests stratification of the top liquid metal surface. This is consistent with VFTRIM-3D modeling, where D atoms migrate into the bulk while the first few monolayers remain mostly lithium. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

The use of solid lithium as a first wall or divertor plasma-facing material has been researched in previous work [1]. Modeling and measurements of solid lithium result in less-than-unity physical sputtering yields [2], which prevent runaway events [3]. Not only is lithium advantageous due to its low Z and relatively good thermophysical properties, but also due to its ability to absorb impinging species. The ability of lithium to strongly absorb impinging D–T ions may lead to high-temperature/low density or ‘low recycling’ regime [3].

If lithium is used in its liquid state, it allows continuous recovery of damaged surfaces exposed to the large heat fluxes in the reactor wall and divertor regions. Results of lithium physical sputtering in the solid state [2] show lower yields than previously predicted [4]. The

present study attempts to determine whether the absolute sputtering yield of liquid lithium is found to be too large for applicable use as a plasma-facing material in tokamak first wall and/or divertor regions.

2. Experiment

The ion-surface interaction experiment (IIAX) is designed to measure the absolute, angular resolved and self-sputtering yields of many particle/target combinations (see Fig. 1). A Colutron ion source is used to create and accelerate gaseous or metal ions onto a 0.018 cm² liquid metal target. Gaseous ions are obtained by means of electron-impact ionization while the lithium metal ions are obtained by thermionic emission from a LiCl powder. The bombarding ions are mass-selected through an EXB filter and decelerated near the target by a five-element cylindrical electrostatic lens system. Complete details of the system can be found in earlier papers [5,6]. A 0.75 mm thick and 100 mm² lithium target is inserted in the main chamber containing argon gas at just above an atmosphere. The target can be rotated in order to

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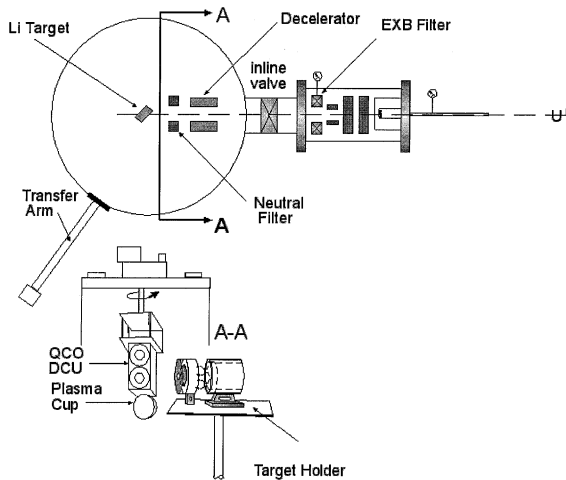


Fig. 1. The IIAX. The experimental device is shown with two differentially pumped chambers. On the right, the ion gun chamber and on the left, the main chamber, where the lithium target sample is located. Cross-section A–A of the main chamber shows the (QCM-DCU) quartz crystal microbalance dual-control unit with respect to the target holder.

provide variation in the angle of incidence. 45° incidence was used based on the average angle of incidence a gyrating particle makes, where the magnetic field lines cross the divertor plates at oblique incident angles.

A plasma cup is used to provide plasma cleaning of the target leading to the removal of any oxides or other impurities from the surface. This method also allows for the saturation of deuterium within the top surface of lithium in the liquid-phase, simulating plasma-facing wall conditions within a fusion reactor. A dual quartz crystal oscillator unit is rotated in front of the target to collect the sputtered flux, measuring the absolute sputtering yield. The QCO unit is mounted on a manipulator and thus its spatial and angular position with respect to the target is known. The solid angle of the QCO which subtends the sputtered flux is measured 0.16 ± 0.02 steradians, assuming a cosine distribution for the angular distribution of sputtered atoms. A small high-temperature, HV substrate heater is used to heat the 0.76 g lithium sample past its melting point to 200°C. Upon melting a thin oxide layer is found on the exposed lithium surface, which is scraped off by an in situ arm rotated in front of the target. A tantalum evaporative shield is placed on the sample to reduce the evaporative flux component during erosion of the sample. The evaporation flux of lithium atoms at 200°C was measured to be 2.8×10^{15} atom/cm². The partial pressure of the system is monitored with a quadrupole gas analyzer. Typical total base pressures before the beam is switched on are 10^{-6} – 10^{-5} Pa. An incident ion flux of the order of 10^{14} ions/cm²/s can be achieved with an average beam spot diameter of 1.0 mm. During the course of a 4 h

exposure the total dose to the target is of the order of 10^{16} ions.

The dual quartz crystal oscillator technique has increased the signal to noise ratio by a factor of 25 over previously used techniques. The mechanism works by using a dual QCM unit which consists of two gold-coated quartz AT cut crystals, in thermal contact, with a fundamental frequency of the order of 6 MHz. One sensor acts as the deposition crystal, the other as a reference crystal. Deposition of the sputtered and evaporative fluxes are measured by a decrease in frequency as mass is collected on the crystal when the QCM is rotated in front of the target. A background trace monitors the evaporative flux using both crystals for a period of 15–24 h. One crystal collects the evaporative flux, while the other remains a reference without deposition. Since the sticking coefficient is 25% for sputtered lithium atoms on the gold-covered QCM crystal, a thin carbon layer is evaporated on both the deposition and reference crystals increasing the sticking coefficient from 25% to 85%. The liquid metal target is then irradiated with the beam and after the proper dose is obtained, the QCM dual unit is rotated away from the target and both crystals are kept running until the change in frequency due to the evaporative flux alone is obtained again. The frequency variation of the deposition crystal is correlated with the mass loss of the sample as will be shown quantitatively in Section 3.

3. Data analysis and modeling

Analysis of the absolute sputtering yield of lithium is done by correlating the frequency variation in the crystal signal with the time period of ion beam dose. The calculation of the absolute sputtering yield is then possible after accounting for sputtering of QCM deposited material by highly incident energetic reflected particles, the sticking coefficient of sputtered atoms onto the QCM crystal and the ion fraction of sputtered atoms. A mass balance is then used to set up the analytical expression for the absolute sputtering yield. The mass deposited on the crystal, which corresponds to the mass loss from the lithium sample, M_d due to both evaporation and physical sputtering is shown to be

$$M_d = D f_i S^{\text{QCM}} Y \Omega m_{\text{Li}_2\text{O}}, \quad (1)$$

where D is the total ion dose, S^{QCM} the sticking coefficient defined as $1 - R_j^{\text{QCM}}$, which corresponds to the reflection coefficient for sputtered species j off the QCM crystal surface and are calculated by VFTRIM-3D, Y the absolute sputtering yield, Ω the solid angle subtended by the QCM crystal surface, f_i a factor accounting for the ion fraction of sputtered species ranging from 1.5 to 1.8 and $m_{\text{Li}_2\text{O}}$ is the mass of lithium oxide

deposited on the QCM deposition crystal. The mass deposited on the QCM crystal as measured by the Leybold Inficon XTC/2 monitor is defined as

$$M_{\text{QCM}} = \frac{\Delta f}{f} M_{\text{crystal}}, \quad (2)$$

where Δf is the frequency change measured from the raw frequency difference between the deposition and reference crystal data, M_{crystal} the mass of the crystal given by the manufacturer, and f is the initial frequency of the QCM crystal. The M_{QCM} term incorporates the mass loss due to sputtering from reflected deuterium or helium neutrals from the lithium surface. The reflection coefficient of deuterium or helium atoms, R_j (j for the species type), the sputtering coefficient of energetic neutrals impinging on the QCM crystal surface, Y_j^{QCM} , and the corresponding solid angle subtending these reflected neutrals, Ω_j , are factored into the mass variation of the QCM in this manner

$$M_{\text{QCM}} = \frac{\Delta f}{f} M_{\text{crystal}} (1 + R_j Y_j^{\text{QCM}} \Omega_j). \quad (3)$$

A mass balance between the mass loss from the lithium sample and the mass gained on the QCM deposition crystal results in the expression for the absolute sputtering yield:

$$Y = \frac{1}{D f_i S^{\text{QCM}} \Omega_{\text{mLi}_2\text{O}}} \frac{\Delta f}{f} M_{\text{crystal}} (1 + R_j Y_j^{\text{QCM}} \Omega_j). \quad (4)$$

For the self-sputtering case we would have the expression:

$$Y_{\text{self-sputtering}} = \frac{1}{D f_i S^{\text{QCM}} \Omega_{\text{mLi}_2\text{O}}} \frac{\Delta f}{f} M_{\text{crystal}}, \quad (5)$$

which differs from (4) because reflected incident lithium atoms are still part of the self-sputtered signal.

Modeling is done with the VFTRIM-3D model which simulates surface roughness [7] based on TRIM-SP [8]. VFTRIM-3D includes an improved low-energy, multiple collision in a binary collision approximation model. Inelastic energy loss is calculated using an equipartition between the local Oen–Robinson inelastic energy loss model and a non-local Lindhard–Sharff inelastic energy loss model. To determine the surface composition to use we note that experimental data from Sugai [9] shows that the hydrogen solubility in lithium is one to one. As the temperature is increased and lithium reaches the liquid state, deuterium atoms will begin diffusing to the bulk [10]. These atoms will form Li–D bonds in subsequent layers below the surface [9]. Therefore, computational runs are modeled with the first layer composed of 100% Li. For solid Li, the surface comprises of a one to one mixture of D and Li. This model is confirmed by comparing the D-saturated solid and liquid lithium modeling results to the experimental

sputtering results. Using a 100% Li layer for both, or using a 50–50% layer for both do not reproduce the data.

In VFTRIM-3D the surface binding energy (SBE) is a key parameter at these low energies. Usually the heat of sublimation (1.68 eV for Li) is used. The bond energy (BE) – the energy to break a bond in the bulk is typically taken as $\text{BE} = 0.1 \text{ SBE}$. For a liquid surface the Guggenheim model [11] empirically fits the surface tension and thus the SBE for liquids as a function of temperature. From this model, at 200°C, the SBE for lithium is calculated to be 1.40 eV. Therefore, an adjusted surface binding energy of 1.40 eV from 1.68 eV is used for the liquid case.

Bonding of bulk atoms play a crucial role in the transfer of energy to recoils in the bulk of the liquid metal. For Li and He bombardment, the bond energies used in the model are the heats of enthalpy for Li–Li and Li–D bonds, 1.1 and 2.49 eV, respectively [12,13]. For deuterium bombardment, free D can be released from bulk layers, where D is present. Therefore, the BE for D atoms coming from the bulk is 0.0 eV.

4. Results

Tables 1–3 summarize and compare solid [2] and liquid lithium sputtering data. Fig. 2 shows experimental data at energies between 200 and 1000 eV for D^+ bombardment of D-saturated liquid and solid lithium at 45° incidence along with VFTRIM-3D modeling. The simulation gives a reasonable fit to the experimental data. The maximum Li sputtered is found near 400–500 eV region for the liquid lithium and 200 eV for solid lithium. Fig. 3 shows experimental data for self-sputtering at energies between 200 and 1000 eV for both liquid and solid lithium. The data is also shown along with the VFTRIM-3D simulation. The self-sputtering yield reaches a maximum near 700 eV for both liquid and solid lithium. Fig. 4 shows experimental data for He^+ bombardment of D-saturated liquid and solid lithium for energies between 200 and 1000 eV. A maximum of the yield from helium bombardment is noted

Table 1
 D^+ on solid and liquid lithium, low-energy sputtering data

E_i (eV) ^a	Lithium $Yn \pm \Delta Yn$	
	D^+ on D-sat.	D^+ on D-sat. liq.
100	0.064 ± 0.016	
200	0.104 ± 0.026	0.117 ± 0.029
450	0.098 ± 0.024	
500		0.126 ± 0.031
700	0.084 ± 0.021	0.121 ± 0.030
1000		0.093 ± 0.023

^a Incident particle energy.

Table 2
Li⁺ on solid and liquid lithium, low-energy sputtering data

E_i (eV) ^a	Lithium $\bar{Y}_n \pm \Delta\bar{Y}_n$	
	Li ⁺ on D-sat.	Li ⁺ on D-sat. liq.
200	0.159 ± 0.040	0.203 ± 0.051
450	0.182 ± 0.046	
500		0.329 ± 0.082
700	0.218 ± 0.055	0.376 ± 0.094
1000	0.135 ± 0.034	0.264 ± 0.066

^a Incident particle energy.

Table 3
He⁺ on solid and liquid lithium, low-energy sputtering data

E_i (eV) ^a	Lithium $\bar{Y}_n \pm \Delta\bar{Y}_n$	
	He ⁺ on D-sat.	He ⁺ on D-sat. liq.
200	0.117 ± 0.029	0.126 ± 0.031
500	0.162 ± 0.041	0.199 ± 0.050
700	0.145 ± 0.036	0.170 ± 0.043
1000	0.133 ± 0.033	0.150 ± 0.038

^a Incident particle energy.

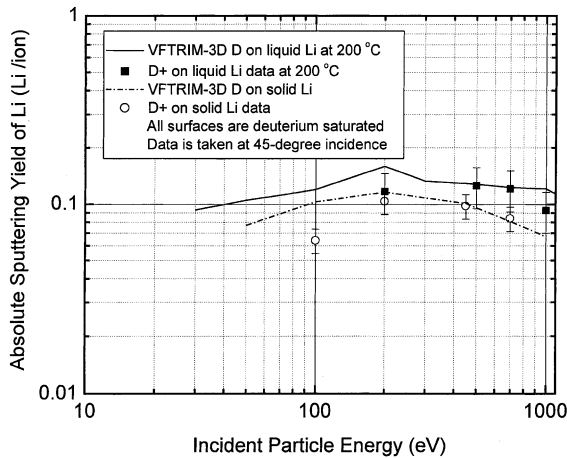


Fig. 2. D⁺ bombardment of D-saturated liquid and solid lithium at 45° incidence.

near 500 eV for both liquid and solid lithium. The fraction of Li atoms sputtered in the ionic state from liquid-phase lithium is 0.65 ± 0.1 compared to 0.55 ± 0.1 for solid-phase lithium [2] at 700 eV He⁺ bombardment.

5. Discussion and conclusions

All sputtering yields for liquid lithium are greater than sputtering from lithium in the solid state. In both cases the relative low absolute sputtering yield of Li is directly related to the saturation of the surface by deu-

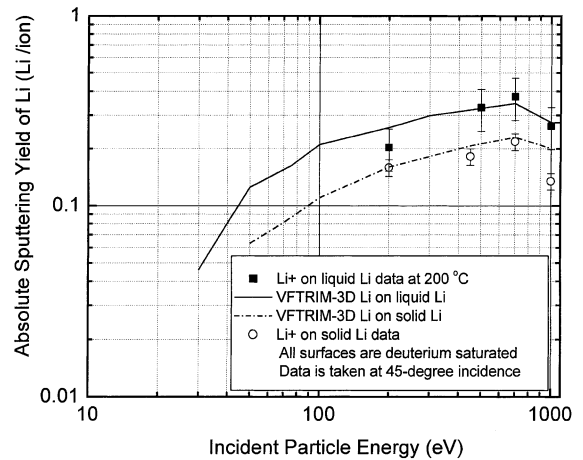


Fig. 3. Li⁺ bombardment of D-saturated liquid and solid lithium at 45° incidence.

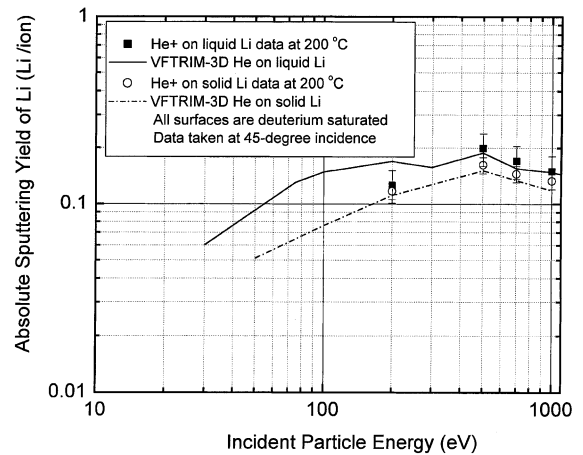


Fig. 4. He⁺ bombardment of D-saturated liquid and solid lithium at 45° incidence.

terium atoms. In the case of the solid-phase, preferential sputtering mechanisms dominate the physical sputtering yield. In the case of lithium in the liquid-phase, the relative bonding between atoms dominates the absolute sputtering of Li atoms.

Deuterium bombardment exhibits different maxima depending on the state of lithium. This suggests the enrichment of deuterium atoms in the bulk of liquid lithium shifting the maximum of the nuclear stopping cross-section to higher energies.

The effect of deuterium saturation varies from solid to liquid-phase. Experimental data show that for solid lithium the sputtering yield of Li atoms is significantly decreased by saturation of the surface [2]. In contrast, although there is a strong solubility of deuterium in liquid-phase lithium, D atoms readily migrate to the

bulk and thus a larger lithium yield is obtained. In addition, the surface binding energy is effectively decreased by an increase in temperature. Therefore, the role of D atoms on the surface of liquid lithium is minor compared to lithium in the solid-phase. For example, the sputtering yield of Li at 700 eV He⁺ bombardment for liquid lithium saturated with deuterium is 0.170 ± 0.034 atoms/ion. While the sputtering yield of Li atoms with the same bombardment conditions on non-saturated liquid lithium is 0.200 ± 0.045 atoms/ion.

Comparing the experimental results for liquid lithium to VFTRIM-3D modeling one notes the significance of Li–D bonding. Using other surface binding and BE models are not able to predict the experimental data. Use of the heat of enthalpy of Li–Li and Li–D bonds and the temperature adjusted SBE gives the proper sputtering yield behavior. As the temperature is increased, we expect the sputtering yield of liquid Li to increase. Experimental results as a function of temperature will be the subject of a paper to be submitted in the near future.

Note that the ion fraction of sputtered atoms plays an important role on erosion characteristics of liquid lithium. Sputtered Li ions will never penetrate to the core plasma due to the sheath at the edge. In the case of liquid-phase lithium, 65% of the sputtered Li atoms come off as an ion.

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