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D, He and Li sputtering of liquid eutectic Sn–Li

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Abstract

The absolute sputtering yields from bombardment of D^+ , He^+ and Li^+ on liquid tin–lithium eutectic have been measured and modeled at energies between 200 and 1000 eV. The Ion-surface InterAction Experiment (IIAX) has been optimized to reliably measure the absolute sputtering yield of many ion-target combinations including solid and liquid lithium. A Colutron ion source is used to create and accelerate gaseous or metal ions onto a liquid metal target. The bombarding ions are mass-selected through an E X B filter and decelerated near the target. The target can be rotated in order to provide variation in the angle of incidence. Deuterium plasma from a hollow cathode source is used to remove any remaining oxides. Upon melting of the sample a thin oxide layer forms and is cleaved by an in situ arm. Results show that sputtering yields from liquid tin–lithium are larger than pure lithium. In addition, modeling with VFTRIM-3D confirms that Li atoms segregate to the surface of liquid tin–lithium. This is consistent with results of ion fraction of sputtered atoms, which show a sputtered-atom ion fraction of 65% for liquid tin–lithium, equal to pure liquid lithium, and <10% for solid tin–lithium. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Liquid metal; Plasma–material interaction; Sputtering

1. Introduction

The use of lithium as a first wall or divertor plasma-facing material has been the object of recent study [1]. Lithium in both solid and liquid phase has also been examined as a fusion first-wall material and demonstrates less-than-unity absolute sputtering yields for light ion and self-sputtering [2]. Lithium is advantageous due to its low Z , good thermal conductivity, and its ability to absorb impinging species. The ability of lithium to strongly absorb impinging D–T ions may lead to a high temperature/low density or ‘low recycling’ regime [3,4]. Lithium’s liquid state allows continuous recovery of damaged surfaces exposed to the large heat fluxes in the reactor wall and divertor regions.

Pure liquid lithium however suffers from high evaporation rates which limits the plasma-facing wall temperature. Therefore a new material, eutectic 80%

Sn–20% Li (0.8 Sn–Li) has been suggested as the next possible alternative to liquid lithium for plasma-facing applications [4]. The new eutectic is attractive due to its low evaporation rate. The evaporative flux for pure liquid lithium is at least three orders of magnitude greater than eutectic liquid 0.8 Sn–Li. For example at 500°C the evaporative flux for 0.8 Sn–Li is $8 \times 10^{14}/\text{cm}^2/\text{s}$ compared to the evaporative flux for pure liquid lithium at the same temperature, $1 \times 10^{18}/\text{cm}^2/\text{s}$. This leads to a wall temperature limit of about 400°C for pure liquid lithium and 700°C for eutectic 0.8 Sn–Li [3]. Due to low solubility of deuterium in liquid tin–lithium [4], a high-recycling regime is expected [3].

Vapor from evaporated eutectic liquid Sn–Li is dominated by the lithium component. In addition Bastasz and Eckstein [5] found that lithium will segregate to the surface upon melting of the eutectic Sn–Li. Therefore lithium would be the plasma-facing material in eutectic Sn–Li in the liquid phase. The present study attempts to determine whether the absolute sputtering yield of eutectic liquid tin–lithium is found to be too large for applicable use as a plasma-facing material in tokamak first wall and/or divertor regions.

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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 including liquid metal targets [2] (see Fig. 1). A Colutron ion source is used to create and accelerate gaseous or metal ions onto a 0.018 cm^2 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 E X B 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 [6,7]. A 0.75 mm thick and 12.5 mm diameter tin–lithium target is inserted in the main chamber. The target can be rotated in order to 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 [10].

A small high-temperature, HV substrate heater is used to heat the tin–lithium sample past its melting point to 380°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. The arm also has a thermocouple, which measures the liquid surface temperature as it is immersed in the liquid metal. A stainless steel evaporative shield is floated on

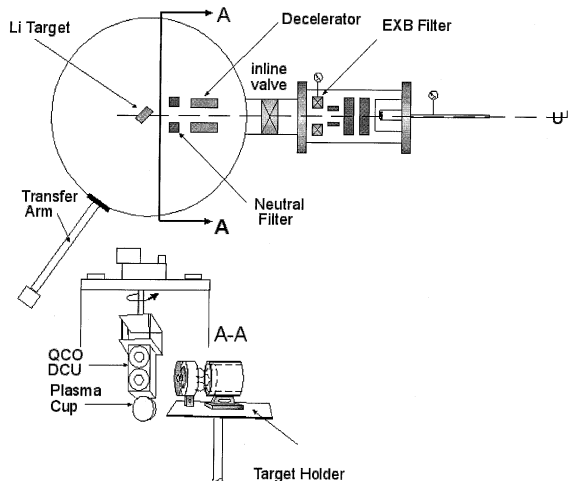


Fig. 1. The Ion-surface InterAction Experiment (IIAX). The experimental device is shown with two differentially pumped chambers. On the right, the ion gun chamber and on the left, the main chamber where the lithium target sample is located. Cross section A-A of the main chamber shows the (QCM-DCU) quartz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

the sample, vertically to reduce the evaporative flux component during erosion of the sample. The evaporation flux of lithium atoms at 380°C was measured to be $2.0 \times 10^{16} \text{ m}^{-2}/\text{s}$. The stainless steel evaporation shield has a 1.5 mm diameter hole, which allows the beam to clear the shield and strike the melted Sn–Li metal. 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. The partial pressure of the system is monitored with a quadrupole gas analyzer. The partial pressure of oxygen before and after melting was found to be relatively constant at $2.0 \times 10^{-7} \text{ Pa}$. Upon sputtering of the sample, the partial pressure of oxygen was noted to rise slightly to $5.0 \times 10^{-7} \text{ Pa}$. Typical total ultimate pressures before the beam is switched on are 10^{-6} – 10^{-5} Pa . An incident ion flux of the order of $10^{18} \text{ ions/m}^2/\text{s}$ can be achieved with an average beam spot diameter of 1.0 mm . A dual quartz crystal oscillator unit is rotated in front of the target to collect the sputtered flux, measuring the absolute sputtering yield. During the course of a four hour exposure the total dose 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 [6]. 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. Details of this measurement and how it is incorporated in the analysis of the absolute sputtering yield is given in a paper of liquid lithium sputtering measurements [2].

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 expression derived in liquid lithium sputtering work [2] for the absolute sputtering yield of lithium is:

$$Y = \frac{1}{Df_i S^{\text{QCM}} \Omega m_{\text{Li}_2\text{O}}} \frac{\Delta f}{f} M_{\text{crystal}} (1 + R_j Y_j^{\text{QCM}} \Omega_j), \quad (1)$$

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, Y the absolute sputtering yield, Ω the solid angle subtended by the QCM crystal surface, f_i is a factor accounting for the ion fraction of sputtered species and $m_{\text{Li}_2\text{O}}$, the mass of lithium oxide deposited on the QCM deposition crystal.

Expression (1) does not include the partial sputtering yield of Sn for two reasons. The sputtering threshold for pure Sn is high near 200–300 eV due to its high elemental surface binding energy (SBE). At 45° incidence the sputtering yield of Sn tops 10–20% at energies of 500–1000 eV, according to simulations using TRIM-SP. In addition, if the beam-facing surface was truly a mixture of Sn and Li atoms, the amount of Sn sputtering calculated above would lead to sufficient Sn on the QCM deposition crystal. This amount would be measurable by X-ray photoelectron microscopy (XPS). XPS measurements show that 99.6% of the mass deposited on the QCM crystal is lithium. This confirms observations that in eutectic tin–lithium in liquid phase, Li atoms migrate to the surface [5]. Thus, the surface sputtered is mostly lithium atoms and not a mixture of Sn and Li atoms. In addition, measurements in IIAX of the fraction of sputtered atoms which emerge as ions of solid 0.8 Sn–Li and liquid 0.8 Sn–Li compared to pure liquid lithium conclude that the beam-facing surface during bombardment is pure Li. The sputtered atom ion fraction of liquid 0.8 Sn–Li is 65%, equal to pure liquid lithium compared to solid phase 0.8 Sn–Li at 10%.

Modeling is done with the VFTRIM-3D model [8] which simulates surface roughness under the basic TRIM framework. VFTRIM-3D includes an improved low energy, non-binary collision 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 be modeled for liquid tin–lithium, we note the experimental results of Bastasz and Eckstein [5]. Li atoms migrate to the surface upon melting. Therefore, computational runs modeling liquid tin–lithium are modeled with the first layer composed of 100% Li. Subsequent layers are modeled using the 80% Sn and 20% Li composition.

In VFTRIM-3D the SBE applies the heat of sublimation of the material as a key parameter at these low energies (usually 1.68 eV for lithium and 3.12 eV for Sn). 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 [9] empirically fits the surface tension and thus the SBE for liquids as a function of temperature. From this model at 380°C, the

SBE for lithium is calculated to be 1.30 eV. However, use of the previously calculated [2] temperature-adjusted SBE of 1.40 eV at 380°C, better predicts the experimental data. The Guggenheim model demonstrates that the SBE is a weak function of the temperature once in the liquid state. Use of the 1.40 eV value implies that the functionality of the SBE with temperature for lithium in the liquid state is even weaker. Therefore, an adjusted SBE from 1.68 eV is used for the first layer composed of liquid Li. For subsequent layers below the model used an effective heat of sublimation of 2.83 eV. This value uses the heats of sublimation of Li and Sn, 1.68 and 3.12 eV, respectively, weighed by their composition.

Bonding of surface and bulk atoms also play a crucial role in the transfer of energy to recoils in the bulk of liquid tin–lithium as in pure liquid lithium [2]. For D, He and Li bombardment, the bond energy used for the pure lithium surface is the heat of enthalpy for Li–Li bonds of 1.1 eV. For subsequent layers below, 10% of the effective SBE for 0.8 Sn–Li is used. Note that as opposed to deuterium bombardment of pure liquid lithium, no free D atoms exist due to low deuterium solubility in eutectic liquid tin–lithium, consistent with our model and with hydrogen solubility measurements of liquid tin–lithium [4]. Deuteration of the liquid tin–lithium surface is found to play no role in the absolute sputtering yield of Li. This is consistent with results showing low deuterium solubility in liquid tin–lithium [4]. For example, measurements in IIAX show that the sputtering yield of Li from a deuterium-treated liquid tin–lithium surface from He⁺ bombardment at 700 eV is 0.297 ± 0.083 compared to a non-deuterium-treated surface with a yield of 0.350 ± 0.098 . In addition, this model correlates with the model used for liquid tin–lithium divertor systems that operate in high-recycling regimes due to low hydrogen solubility [3].

4. Results

Table 1 summarizes measured and computational results for D⁺, Li⁺ and He⁺ bombardment of liquid tin–lithium. Fig. 2 shows experimental data for the absolute sputtering yield of Li at energies between 200 and 1000 eV for D⁺, He⁺ and Li⁺ bombardment of liquid tin–lithium at 45° incidence, along with VFTRIM-3D modeling.

The simulation gives a very good fit to the experimental data. The maximum Li sputtered for deuterium bombardment is found near 400 eV region, similar to pure liquid lithium results [2]. Experimental data for helium bombardment show a maximum of the yield near 400 eV, although this is only suggested by the model since no data below 400 eV was obtained for helium bombardment. This sputtering yield maximum is also close to those obtained for helium bombardment of pure

Table 1
Liquid tin–lithium low-energy sputtering data

Incident particle energy E_0 (eV)	D^+ on liquid tin–lithium $Y_n \pm \Delta Y_n$	Li^+ on liquid tin–lithium $Y_n \pm \Delta Y_n$	He^+ on liquid tin–lithium $Y_n \pm \Delta Y_n$
200	0.130 ± 0.026		
300		0.536 ± 0.104	
400	0.156 ± 0.040		0.312 ± 0.093
500		0.598 ± 0.142	
700	0.120 ± 0.024	0.662 ± 0.148	0.297 ± 0.083
1000	0.087 ± 0.017	0.592 ± 0.162	0.256 ± 0.073

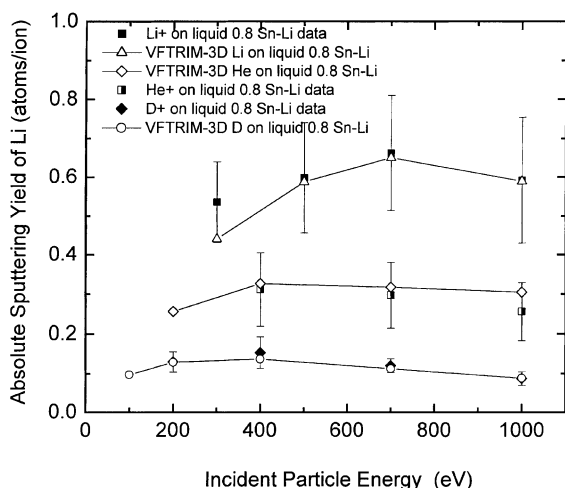


Fig. 2. D^+ , He^+ and Li^+ bombardment of liquid tin–lithium data and VFTRIM-3D simulation at 45° incidence.

solid and liquid lithium. Data for lithium sputtering at energies between 300 and 1000 eV are also shown along with VFTRIM-3D simulation in Fig. 2. The Li sputtering yield reaches a maximum near 700 eV, similar to results of pure solid and liquid lithium sputtering [2]. The measured fraction of sputtered particles that emerged as ions for D^+ , He^+ and Li^+ bombardment was less than 10% for solid 0.8 Sn–Li, but around 65% for liquid 0.8 Sn–Li.

5. Discussion and conclusions

The largest contribution to the absolute sputtering of lithium comes from bombardment by lithium ions onto liquid 0.8 Sn–Li. The maximum absolute sputtering yield of Li by Li bombardment is about a factor of two greater for helium bombardment and a factor of four greater for deuterium bombardment. This is due to a greater transfer of energy between Li bombarding ions and Li atoms on the surface, compared to D and He bombarding ions. In addition subsurface layers containing 80 a/o Sn act as a reflective wall thus leaving

incident bombarding particles as highly-energetic back-scattered particles, leading to more lithium sputtering. For example, incident Li atoms transfer about 16% of their energy to sub-surface Sn atoms and incident He atoms, 10%. This leaves backscattered Li and He atoms with sufficient energy to cause significant sputtering of surface Li atoms. Greater sputtering results from lithium bombardment due to their larger mass.

All sputtering yields for liquid tin–lithium are larger than those on pure lithium in liquid phase. This is primarily due to the fact that lithium is preferentially sputtered from the subsurface layers containing 80% Sn and 20% Li. In the case of pure liquid lithium, subsurface layers contribute very little lithium to the sputtering yield since in that case, deuterium is preferentially sputtered. Since solubility of deuterium in liquid tin–lithium is relatively low its contribution in decreasing the absolute sputtering yield of Li is also very low. Furthermore, the subsurface Sn–Li reflective wall as discussed above leads to greater lithium sputtering on the basis of energy transfer mechanisms. For example the maximum energy transfer factor for D atoms on Li is 0.70 compared to 0.066 for Sn.

In addition to preferential sputtering mechanisms, the relative bonding between atoms in liquid tin–lithium is critical. As in liquid lithium, in tin–lithium the SBE is effectively decreased by an increase in temperature. However, for subsurface layers the effective SBE is higher which, accounts in part for the almost non-existent sputtering of Sn from the surface.

Again as in the sputtering of pure liquid lithium the ion fraction of sputtered atoms is important. Particles that sputter as ions will be quickly returned to the surface because of the plasma sheath. For the case of liquid tin–lithium, the ion fraction is measured to be 65% compared to <10% for solid tin–lithium. This relatively large ion fraction coincides with those measured in pure liquid lithium [2]. This provides further evidence that Li atoms segregate to the surface of liquid tin–lithium and behave similar to pure liquid lithium.

These results indicate that 0.8 Sn–Li may be an ideal plasma-facing component for future high-heat flux fusion devices. The desirable low Z and high ion fraction of the sputtered flux is present due to the Li segregating

to the surface. However, the disadvantages of a Li surface – high evaporation rate, low boiling point, and high tritium retention – are absent.

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