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Temperature dependence of liquid-lithium sputtering from oblique 700 eV He ions

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Abstract

The lithium-sputtering yield of liquid lithium as a function of sample temperature has been measured in the ion-surface interaction experiment (IIAX). Lithium sputtering is measured for D^+, He^+ and Li^+ bombardment at energies between 100 and 1000 eV at 45° incidence. In this work VFTRIM-3D is used to provide a qualitative physical picture of mechanisms responsible for the temperature dependence of liquid-lithium sputtering. The present study is done for 700 eV He^+ bombardment of liquid lithium at 45° incidence with respect to the target normal. The lithium-sputtering yield, after evaporation is taken into account, is found to increase almost an order of magnitude when the target temperature is increased from the melting point up to roughly 410 °C. The deposited energy distribution near the liquid-lithium surface is found to play a significant role in explaining the observed enhanced lithium sputtering as well as the temperature dependence of the surface binding energy.

Keywords: Plasma-wall interactions; Liquid lithium; Sputtering; Thermal sputtering; VFTRIM-3D; Evaporation

1. Introduction

The use of liquid lithium as a plasma-facing component material is currently being studied. Liquid facing materials can provide existing and future fusion devices an innovative solution to the technological challenge of the coupling between fusion plasmas and the materials they interact with [1]. Liquid systems can enhance the performance of existing fusion devices by innovative particle control techniques. For example, liquid lithium has a very high-hydrogenic pumping capability thus resulting in a low-recycling regime with steep pedestal edge temperatures characteristic of high-confinement regimes [2]. Liquid systems can also provide plasma-facing surfaces able to withstand the high heat-flux (>10 MW/m^2) environment of a next-step option machine.

Measurements of ion-surface interactions have been conducted in the ion-surface interaction experiment (IIAX) and the flowing liquid-surface retention experiment (FLIRE) at the University of Illinois [3–8]. Recently measurements of lithium erosion as a function of target temperature from static liquids have been completed. An enhancement in lithium erosion is measured as the surface temperature is raised from 200 to about 410 °C. This phenomenon has also been measured in the linear plasma device, PISCES-B [9]. The temperature dependence of sputtering has been studied to a large extent and a strong, temperature dependence is not expected for physical sputtering of materials [10,11], although some studies suggest that for high-near-surface energy densities such enhancements may exist [12–14]. However, sputtering studies of liquids is limited especially for light particle bombardment at low-incident particle energies, typical of conditions found in a fusion device.

An understanding of the temperature dependence of lithium sputtering is important since a temperature window in which to operate a fusion device must be established and enhanced erosion mechanisms can

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lead to prohibitive impurity levels. In this work He^+ bombardment is only considered to avoid the complications from chemical activity of other bombarding particles such as H^+ or D^+ on liquid lithium.

Motivation for this work is to investigate mechanisms believed to be responsible for lithium erosion enhancement with target temperature with the binary collision approximation (BCA) – based simulation code VFTRIM-3D, which simulates surface roughness under the basic TRIM framework [15]. VFTRIM-3D cannot simulate the temperature dependence of sputtering. Instead a number of parameters used in VFTRIM-3D will be varied with target temperature to elucidate mechanisms responsible for lithium-sputtering enhancement with target temperature.

2. Experimental measurement of lithium-sputtering temperature dependence

Liquid-lithium sputtering is measured with a quartz crystal microbalance. Details of the experimental technique and analysis are described in earlier references [4,6,7]. The incident particle fluxes range from 10^{13}–10^{14} ions/cm^2/s. A background trace of liquid-lithium evaporation is taken before and after beam irradiation at each respective temperature. Before irradiation with helium ions, the liquid-lithium surface is treated with a 10^{16}–10^{17} D/cm^2 fluence of deuterium ions to simulate conditions in a fusion device. In addition, 45° incidence was used based on the average angle of incidence a sheath-accelerated, gyrating particle makes where the magnetic field lines cross the divertor plates of a fusion device at oblique incident angles [16]. The lithium-sputtering yield is obtained from the QCO (quartz crystal oscillator) frequency data after subtracting the background evaporation data. During ion irradiation, therefore, any ion-induced evaporation is not accounted for in the data.

3. Simulation and modeling

Computational analysis is done with VFTRIM-3D (vectorized fractal Transport in Matter 3D) code, which simulates surface roughness under the basic TRIM framework [15] for an amorphous target. In VFTRIM-3D the surface binding energy (SBE) applies the heat of sublimation of the material as a key parameter at these low energies. For liquid lithium at 200 °C, a 'smooth' surface with fractal dimension of 2.00 is used. The model used in VFTRIM-3D is described in earlier work [4].

For temperatures greater than 200 °C, a systematic model was developed (Model 1, described in sections to follow and tabulated in Table 1) to predict the measured sputtering yield of lithium from liquid lithium with an increase in target temperature. This model is then compared to three other models (Models 2–4) to determine which physical mechanism(s) is important in the sputtering of liquid lithium as target temperature is increased.

Table 1
VFTRIM-3D physical models describing key mechanisms in the lithium-sputtering temperature dependence for 700 He^+ bombardment at 45° incidence

Model number	SBE (eV)	D at.%–Li (near surface)	Surface energy distribution altered
1	Sbe(T)	$C_D(T)$	Yes
2	1.40	$C_D(T)$	Yes
3	Sbe(T)	50% D–Li	Yes
4	Sbe(T)	$C_D(T)$	No

For Models 2–4, one parameter/mechanism is kept constant with temperature. For example, Model 2 keeps the SBE constant at 1.40 eV with target temperature.

Four main mechanisms are hypothesized in Model 1 as responsible for lithium erosion enhancement and depend on target temperature: (a) the SBE, (b) the binding energy of lithium and deuterium atoms, (c) the near-surface concentration of deuterium-implanted particles and (d) the deposited energy density distribution near the liquid surface.

The second model, Model 2, keeps the SBE constant with temperature varying the other parameters with temperature as tabulated in Table 1. Model 3 keeps the near-surface concentration of deuterium atoms constant with temperature and Model 4 does not alter the deposited energy density distribution. The mass density for lithium is a very weak function of temperature and thus kept constant at 0.513 g/cm^3 for all models.

3.1. Surface binding energy as a function of temperature

The first parameter accounted for in the models (except Model 2) is the SBE. For temperatures greater than 200 °C, the temperature-dependent surface tension Guggenheim Model utilized by Grossman is not used [17]. Rather, the SBE is calibrated to spectroscopy measurements of temperature-dependent sputtered-energy spectra from liquid-lithium erosion in PISCES-B [9], assuming a Thompson distribution [18]. The SBE dependence on target temperature, Sbe(T), is shown in Fig. 1 along with the temperature dependence of the binding energies for the Li–Li and Li–D bonds.

3.2. Bond energy as a function of temperature

The second parameter/mechanism accounted for in the models is the bond energy between lithium atoms

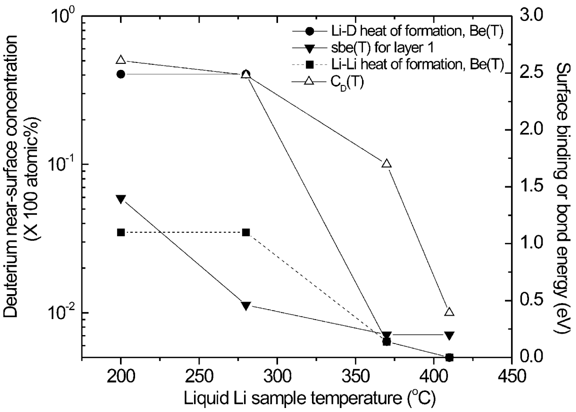

Fig. 1. Temperature-dependent parameters for Model 1 used in VFTRIM-3D simulation of liquid-lithium-sputtering temperature dependence. The SBE, Sbe(T) calibrated to data from PISCES-B [9], The binding energy Be(T) using the heat of formation, and the deuterium near-surface composition $C_D(T)$ are plotted here against target temperature.

and between lithium atoms and deuterium-implanted atoms. The binding energy or dissociation energy is the energy required to break a bond between two atoms and results in mobile atoms. The heats of formation for these bonds are used, similar to the model at 200 °C. The heat of formation for lithium bonds is 1.1 eV and the heat of formation for Li–D bonds is 2.49 eV.

The Li–Li and Li–D bond energies are dependent on temperature and are plotted in Fig. 1. The heats of formation used are discussed in earlier work [4]. At higher temperatures the coordination number of atoms in the liquid state will decrease. This is simulated by a decrease of the respective bond energies for Li–Li bonds and Li–D bonds [19] with an increase in temperature.

3.3. Near-surface deuterium concentration as a function of temperature

The third parameter/mechanism accounted for in the models (except Model 3) is the net concentration of deuterium atoms below the pure lithium top layer. Liquid lithium is known to be an effective pump of hydrogen isotopes and this has been recently measured for liquid lithium at low-deuterium implantation energies [20] and free-surface flowing liquid lithium [5]. More detail on the implantation of hydrogen and hydrogen isotopes in liquid lithium and its alloys is given in other work [8,21–23].

The deuterium near-surface concentration below the pure lithium top layer is plotted against target temperature, $C_D(T)$ in Fig. 1. As the temperature is raised the net amount of deuterium atoms below the pure lithium top layer decreases and thus the number of lithium atoms near the surface increases. Deuterium atoms are preferentially sputtered reducing the amount of lithium atoms sputtered [8]. As the lithium sample temperature is increased the net amount of deuterium atoms near the surface is decreased and thus the lithium-sputtering yield increases.

3.4. Transfer of kinetic energy from subsurface layers to the surface

The fourth parameter/mechanism in the models (except Model 4) accounts for the energy from soft-collisions to the top layer. The amount of energy deposited near the surface can be significant for a low mass, low-atomic number material such as lithium bombarded by helium ions. Much of this energy is dissipated in low-energy non-binary 'soft' collisions. An increase in the local surface temperature would lead to more mobile lithium atoms whose bonds are weaker, thus energy deposited near the surface can result in a higher probability to sputter (or evaporate) surface lithium atoms.

Since VFTRIM-3D only simulates binary collisions of energetic particles with stationary particles, the energy imparted near the surface is underestimated. Non-binary collisions normally are irrelevant in VFTRIM-3D in the solid state because of the magnitudes of surface binding energy and binding energy compared to the amount of energy the soft-collision partners receive. In the liquid state non-binary collisions are more important since even small amounts of imparted energy may be able to liberate an atom. Therefore, a mechanism is added in VFTRIM-3D to transfer momentum to the non-binary collision partners near the surface. This is done by juxtaposing the secondary knock-on atoms portion of the collision cascade along the surface as shown schematically in Fig. 2. A more sophisticated model to simulate this mechanism is planned for future work [24].

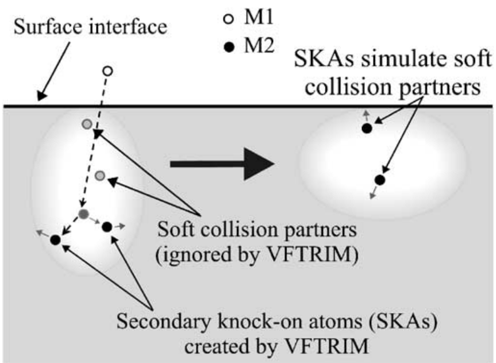

Fig. 2. Schematic showing the juxtaposition of secondary knock-on atoms' portion of the collision cascade along the surface to account for energy transfer from soft-collisions to the top layer.

4. Results and conclusions

The results for 700 eV He^+ bombardment at 45° incidence as a function of temperature for the four models described is shown in Fig. 3. As shown in Fig. 3, the experimental data is well predicted by Model 1. If the variation in SBE with temperature is ignored and kept constant (Model 2), the model underestimates the yield. If the net deuterium atom concentration is kept constant (Model 3), the model also underestimates the yield. If one accounts for both these variations but no additional energy is transferred from subsurface atoms to topmost atoms (Model 4), the yield is again underestimated.

The key mechanism resulting in a significant change in the lithium-sputtering yield as a function of temperature is the additional momentum imparted to the surface from soft-collisions. However, the mechanism behind this transport, whether an enhanced evaporative component from a thermal spike, or enhanced recoil density at the surface due to a collisional spike is still in dispute. The results from Model 1 conclude that spike-like conditions leading to enhanced erosion can exist for light-particle bombardment at low-incident particle energies contrary to the existing region of applicability of non-linear sputtering events [25].

An equally important mechanism is the SBE variation with temperature resulting in a significant effect on the lithium-sputtering yield. When the SBE is kept constant with temperature (Model 2) the liquid-lithium-sputtering yield is strongly underestimated. The compositional variation of implanted deuterium atoms near the surface does not show a strong effect on the liquid-lithium-sputtering yield with temperature. This is expected since a significant change in the lithium-sputtering yield is expected less from a variation in the net concentration of deuterium atoms than a dramatic change in SBE.

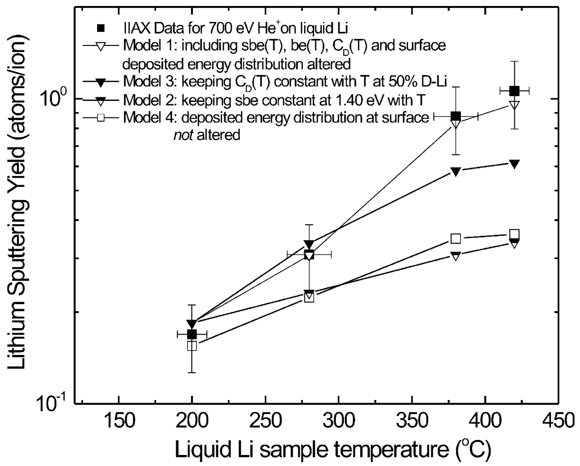

Fig. 3. IIAX results for He^+ bombardment of D-treated liquid lithium at 45° incidence plotted versus liquid-lithium sample temperature. VFTRIM-3D models (Models 1–4) are compared to experimental data.

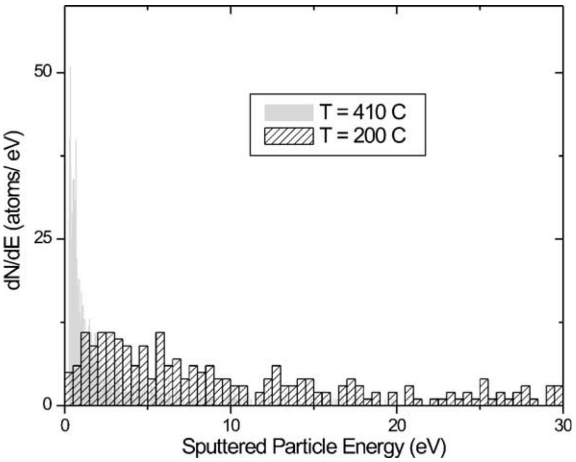

Fig. 4. VFTRIM-3D sputtered energy distributions for lithium particles sputtered from liquid lithium at 200 and 410 °C.

Since the SBE is directly proportional to the mean ejected energy of sputtered atoms, this decrease in energy could also be related to either enhanced evaporation mechanisms or enhanced damage near the liquid surface [13,26]. The energy distribution of sputtered particles is shown in Fig. 4 for liquid-lithium temperatures 200 and 410 °C calculated from Model 1 in VFTRIM-3D. A larger number of particles are sputtered at lower energies when the temperature is increased from 200 to 410 °C. This effect is measured in IIAX from the distribution of sputtered lithium ions for temperatures up to 410 °C [27]. About 60–65% of the sputtered lithium particles are in an ionized state [4,6]. The ion distributions show a ‘shift’ to lower energies implying thermal sputtering mechanisms such as ion-induced evaporation [13,26]. This result is significant since the experimental data subtracts evaporation of lithium without beam irradiation and any ‘ion-induced’ evaporation is not accounted for in the data.

Taking into account the effect of additional energy near the surface, the effective decrease of SBE and binding energy with temperature makes a significant difference to the lithium yield. In turn, variation of near-surface deuterium concentration for lithium in the liquid state does not lead to a significant change in lithium sputtering. In addition, although an enhancement of erosion is noted the lithium-sputtering yields shown in Fig. 3 are for both neutrals and ions. Lithium erosion remains tolerable even at higher target temperatures since in a fusion device only the neutral lithium yield is relevant due to the magnetic sheath.

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