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Molecular dynamics modeling of deuterium in liquid lithium surfaces

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Abstract

Sputtering yields and reflection coefficients from liquid lithium surfaces under low-energy, light-particle bombardment have been modeled for this work. An extended molecular dynamics (MD) simulation has been developed using an improved singlet ab initio Li–D potential splined with the universal potential at small distance for higher energy interactions. The results show a temperature effect. For example, 100-eV deuterium incident at 45° on liquid Li surfaces at 473 K and 653 K give sputtering yields of 0.196 ± 0.040 and 0.315 ± 0.060 respectively, and reflection coefficients of 0.165 ± 0.040 and 0.234 ± 0.060. The effects of surface temperature, incident energy and incident angle on sputtering yields and reflection coefficients, together with the \pertinent energies of the sputtered and reflected atoms, are shown and compared with the results of the standard binary collision code, TRIM.SP.

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Keywords: Molecular effects; Liquid metal; Lithium; Sputtering; Reflectivity

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1. Introduction

Liquid lithium has been proposed as a plasma facing component (PFC) material [1] in fusion devices because of its low atomic number and good thermal properties. Compared with solid surfaces, the structural and thermodynamic properties of liquid Li surfaces are different, which leads to significant effects on the erosion and reflection. Recent experiments, IIAX at UIUC [2] and PICSES at UCSD [3–5], show enhanced erosion with increasing surface temperature. Codes based on the binary collision approximation (BCA) (e.g. TRIM) cannot model these temperature effects. Therefore, molecular dynamic (MD) modeling is widely used to study issues related to PFC materials, since MD includes temperature effects, as well as cascade effects. This paper shows sputtering yields and reflection coefficients calculated with MD simulations, as well as the energies of sputtered and reflected atoms, for two different angles of incidence and two different temperatures.

2. MD simulation method

A MD code, MolDyn [6], was improved to handle the lithium/deuterium system. First, the liquid Li–Li potential was obtained with the neutral pseudoatom (NPA) method [7] by considering both structural and dynamical properties. This potential was developed for

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liquid lithium MD calculations and is effective for high temperatures, ranging from 470 K to 843 K. We used the singlet ab initio potential [8] splined with the universal potential at small distances for the D–Li interatomic potential. The weight function is given by [9]

$$f_c(r) = \frac{1}{2} + \frac{1}{2}\cos\left[\frac{\pi(r - r_1)}{r_2 - r_1}\right]. \quad (1)$$

This potential enables accurate predictions of the scattering of the alkali metal atoms by deuterium atoms [1,10]. There have been recent attempts to include surface effects in the potential [11] but they are not included in this work. The Berendsen velocity scaling method [12] is used to control temperature in the calculations. All Li atoms within the surface are considered as bulk plasma but are subject to different heat baths. Basically, the top layer is free while the bottom layers within 2 Å of the boundary are fixed. The sides of the surface have periodic boundary conditions. The Li surface started as a BCC structure 42.2 Å × 42.2 Å and 59.8 Å deep, containing 5040 Li atoms. The determinations of the surface dimensions used in the MD calculations are described as below.

2.1. Surface preparation

In order to simulate a surface consisting of liquid PFC material, the original surface (BCC) was gradually heated to 473 K or 653 K over a long time until equilibrium was reached. Fig. 1 shows the oscillating radial distribution function [13] curves characteristic of liquid properties compared with the discrete peaks seen for a solid surface.

To insure the correct interaction between atoms, the surface (x, y) dimensions should be greater than twice the cutoff distance. Otherwise, an atom might interact with an image of itself and the images of the other atoms due to the periodic boundary condition. Increasing x and y dimensions beyond 42.2 Å did not affect incident ion trajectories. The cut-off distance for the potential is 5.75 Å. The depth of the surface plays a key role for including the necessary cascade effects. Sputtering yields have been investigated by two other codes: MAIDS [14] and VFTRIM [15]. According to the results of MAIDS, the deepest sputtered atoms come from ~40 Å. Only 1.7% of the sputtered atoms come from initial positions deeper than 40 Å. In addition, several VFTRIM runs were done, each with a different maximum depth of the material being simulated. The results show no effect on total sputtering yield once the depth is larger than ~40 Å. Hence, in order to account for extended cascade effects, 60 Å was chosen for the surface depth.

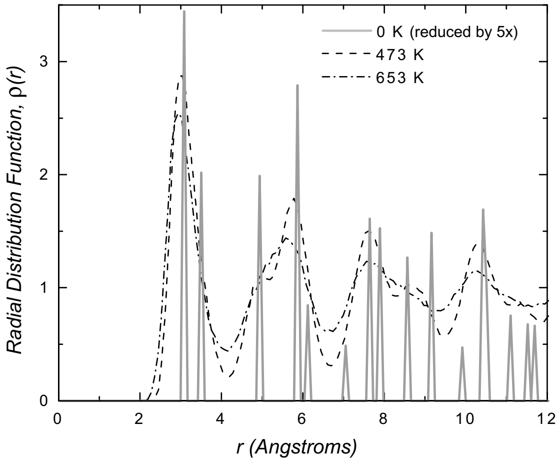

Fig. 1. The radial distribution functions of the created Li surfaces. The discrete spectrum lines correspond to the solid BCC Li surface at 0 K.

2.2. Calculation configuration

To pursue a more accurate simulation, the time step needs to be made small enough to avoid any unphysical results. The trajectories for 300-eV (the maximum incident energy we used in this work) deuterium atoms incident at 45° on a 473-K Li surface at different time steps were investigated. When time step was greater than 0.1 fs, the cascade collisions in deep layers (~40 Å) were missed which resulted in an incorrect trajectories. The model gave similar backscatter and sputtering coefficient results when time step was less than or equal to 0.1 fs. As a tradeoff between accuracy and CPU time, 0.1 fs was chosen as the time step in present work.

The pre-prepared liquid Li surfaces were used for the following MD calculations. Deuterium atoms, with energies ranging from 50 eV to 300 eV, were incident at 20° and 45° on the liquid lithium surfaces at 473 K and 653 K, respectively. A distributed computation system among 15 PCs was employed to speed up the calculations. We benefit from such a multiprocessor-like system since it takes about 7–10 h of CPU time for a single flight on one machine. The effects of incident energy, incident angle and surface temperature on the sputtering yield and reflection coefficient have been investigated and are discussed below.

3. Numerical results

The sputtering yield for deuterium incident at 20° and 45° on liquid lithium surfaces with temperatures of 473 K and 653 K are shown in Fig. 2. The sputtering yield tends to be higher in the case of the higher surface temperature. As described in earlier work [16], the higher surface temperature allows the surface binding energy to be temporarily reduced during the cascade-collision with the sputtered atom due to non-binary collisions

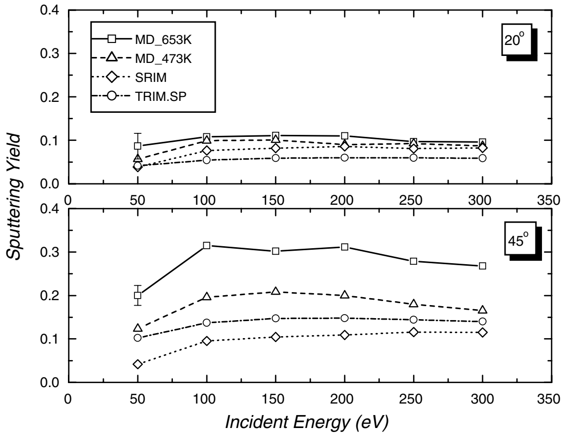

Fig. 2. The sputtering yield as the function of incident energy. The error bars shown are representative of the statistical error for every point.

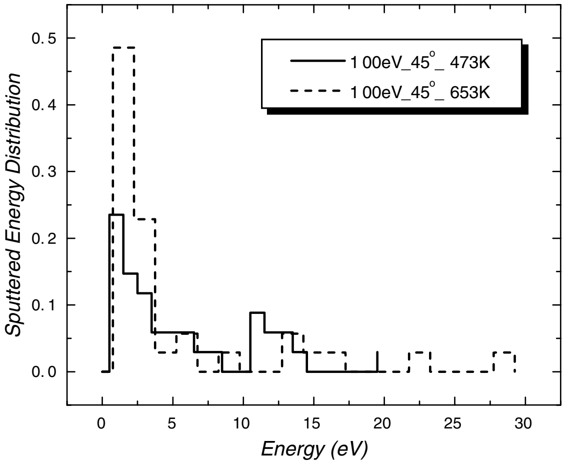

Fig. 4. The energy distribution of the sputtered atoms at different surface temperature. The low energy peak comes from the thermal sputtering due to evaporation flux on the surface.

so that the erosion is enhanced. Larger incident angles from normal enhance sputtering too. It is shown that the surface temperature has a significant effect on sputtering, especially at higher incident angles. TRIM.SP [17] and SRIM [18] results are also shown. As expected, at the highest energies where the collision cascades are better approximated by the BCA, the MD and BCA codes tend to converge.

In order to understand this temperature effect, the average energy and the energy distribution of the sputtered atoms is shown in Figs. 3 and 4, respectively. According to Fig. 3, surface temperature has little effect on the average energy of the sputtered atoms, but the energy distribution of the sputtered atoms is affected, as seen in Fig. 4. There are two peaks in the energy distribution: a low-energy stimulated-evaporation component and the more typical Thompson energy distribution contribution. Higher surface temperature increases the stimulated-evaporation low-energy peak. This has also been seen experimentally [2–5].

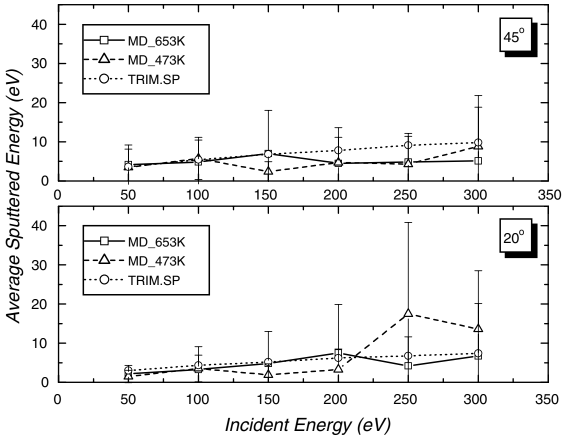

Fig. 3. The average energy of the sputtered atoms. The error bars show the standard deviation in the sputtered energy distribution.

Furthermore, the vapor pressure due to evaporation at high temperature has been calculated by integrating the Clausius–Clapeyron equation to see if it could be seen in the MD simulations. Vapor pressures of 4.8×10^{-7} mTorr and 0.027 mTorr were found for 473 K and 653 K, respectively. Based on the assumption of no ambient pressure limitation, the equivalent evaporation rates are 0.05 1/s and 2509.10 1/s correspondingly. Even this huge vapor pressure at 653 K only represents one atom leaving a 42 Å × 42 Å section of surface area every 0.40 ms. However, the time step and time scale in the MD calculation are in the order of fs and ps respectively, which are much less than the evaporation time scale. Hence, the evaporation due to the high temperature of the surface cannot be seen in MD.

The reflection coefficients for deuterium incident at 20° and 45° on liquid lithium surfaces maintained at 473 K and 653 K are shown in Fig. 5. As expected, the reflection coefficient becomes higher at the more glancing incident angle. The surface temperature has an effect only at low incident energies and shows good agreement with TRIM.SP results at high incident energies.

As shown in Fig. 6, the normalized average energy of the reflected atoms is independent of surface temperature. Similarly with the reflection coefficient, larger incident angles lead to larger average reflected energies, as expected. Basically, MD and TRIM.SP show a range of 20–50% for $< E_{\text{ref}} > / E_{\text{inc}}$.

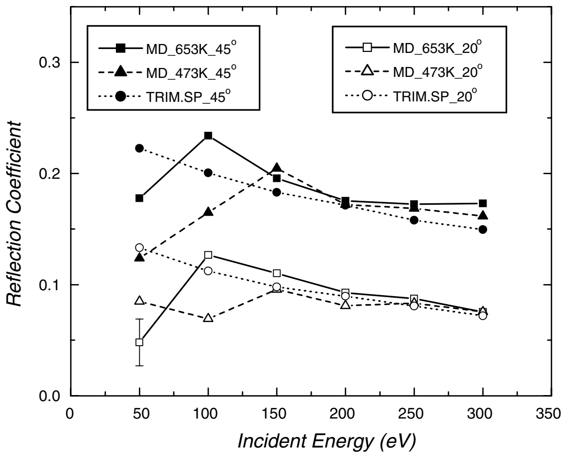

Fig. 5. The reflection coefficient as the function of incident energy. The error bar shown is representative of the statistical error for every point.

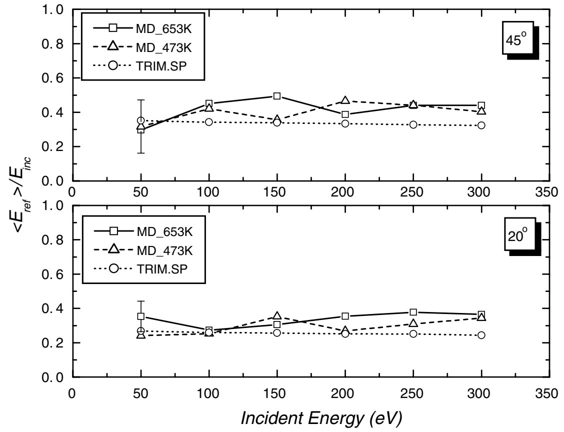

Fig. 6. The ratio of average reflected energy and incident energy. The error bars show the standard deviation in the reflected energy distribution.

4. Conclusions

Liquid Li–Li and D–Li potentials were added to the MolDyn code, which was modified to look at liquid metal erosion. Sputtering and reflection data for deuterium incident on liquid lithium have been calculated using this improved MD code. The surface temperature has a key effect on sputtering yield. Sputtering yield increases with surface temperature and incident angle, as observed in Li^+ on liquid Li experiments [2]. Furthermore, surface temperature has a significant effect on the thermal sputtering contribution from the liquid surface. The average energy of the sputtered atoms is independent of surface temperature and incident angle; however, the energy distribution is responsive to surface temperature changes. The reflection coefficient is slightly affected by surface temperature only at low incident energies and agrees well with TRIM.SP at higher energies. The reflection coefficient and average energy of reflected atoms increases with incident angle, as expected.

MD and TRIM.SP results have been compared in the results above. MD results differ most from TRIM.SP results for the sputtering yield at lower incident energies. TRIM.SP is based on the binary collision approximation, which is not valid at low incident energies. Moreover, the liquid state and temperatures effect on the surface cannot be modeled by TRIM codes. Therefore, the MD results presented here are more valid for liquid PFCs, especially at low incident energies when surface bonding and cascade collisions are important.

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