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Behavior of high temperature liquid surfaces in contact with plasma

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Abstract

The erosion rate of liquid lithium samples exposed to plasma bombardment has been studied in PISCES-B. The temperature dependence of the erosion yield from liquid samples exhibits an enhancement beyond that expected from a combination of physical sputtering and evaporation. The enhancement is observed during either deuterium or helium plasma exposure and is observed to depend linearly on the incident ion flux. The onset of the enhanced erosion coincides with a decrease in the ejection energy of particles from the sample, indicating that the enhancement is due to an increase in evaporation from the sample. The implications of these results on the implementation of a liquid plasma-facing component are discussed.

Keywords: Plasma-wall interactions; Erosion; Liquid metals; Sputtering

1. Introduction

The concept of flowing liquid plasma-facing components has been proposed as a potential solution to the critical plasma-material interaction issues faced by magnetically confined fusion plasma experiments. A flowing wall would remove operational constraints based on wall erosion determined lifetime, material damage induced during off-normal events and damage/radioactivity due to neutron loading, thereby increasing the economic attractiveness of nuclear fusion as an energy source. However, little work has been done in the area of how plasma interacts with a free liquid surface and so these potential benefits could be outweighed by unforeseen difficulties.

The operational limit of a liquid plasma-facing surface is typically based on the tolerable amount of impurity atoms in the core plasma. This calculation usually involves a maximum operating temperature of the liquid directly derived from the equilibrium vapor pressure of the material. Measurements obtained from the PISCES-B device indicate that there is a material loss mechanism further restricting the maximum operating temperature of a free liquid surface exposed to plasma bombardment.

The dependencies of the enhanced loss rate mechanism are examined. The enhanced loss rate scales linearly with incident plasma ion flux. The enhanced loss rate is independent of incident plasma energy. This phenomenon occurs regardless of the bombarding plasma species and has been observed from both lithium [1] and gallium [2] samples. The implications of these results must be incorporated into any design effort to implement a liquid plasma-facing component.

2. Experimental setup

The PISCES-B device [3,4] is a steady-state plasma facility used to investigate fusion plasma-material interactions (Fig. 1). The device consists of a plasma source region, a main chamber and a sample-loading chamber. A 6-cm diameter plasma column is formed along the magnetic axis of the vacuum chamber. A fast scanning

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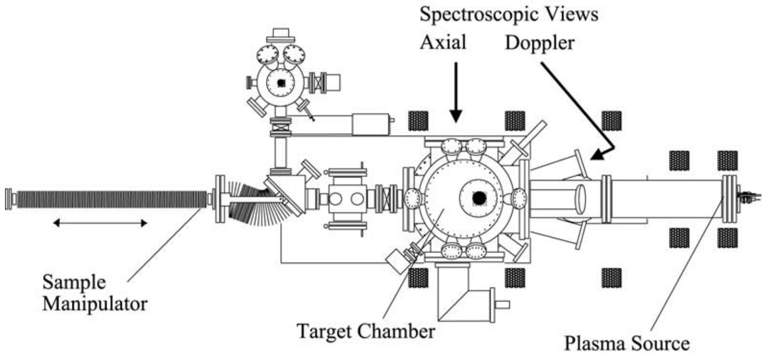

Fig. 1. Schematic of PISCES-B device showing the orientation of 'axial' and 'Doppler' spectroscopic views of plasma interaction region.

double-Langmuir probe is used to measure the plasma temperatures and densities in the radial direction. The facility produces steady plasmas for several hours of continuous exposure. The base pressure of the system is 2.0×10^{-8} Torr. The plasma parameters, T_e (5–40 eV) and N_e ($1–100 \times 10^{17}$ m^{-3}), near the target region can be adjusted by varying the gas flow rate and input discharge power to the cathode. Applying a negative bias to the samples provides the ion bombarding energy. A sample manipulator holds the biased samples in the center of the plasma column such that the magnetic field (and therefore the ion incidence angle) is perpendicular to the sample surface. The samples consist of a molybdenum cup mounted vertically on the end of the sample manipulator. The lithium sample (1 cm diameter × 2 mm deep) sits inside the well of the molybdenum cup. Surface tension prevents the lithium sample from flowing out of the holder when the sample liquefies.

The sample manipulator is also used to control the samples' temperature by varying the flow rate of the coolant (either water or high-pressure gas) through the manipulator. The sample temperature is typically measured in two ways, by directly measuring a thermocouple inserted through the back of the molybdenum cup into the liquid pool and by an infrared pyrometer. However, the very low emissivity of liquid metal surfaces makes the interpretation of the pyrometer data difficult and, therefore, in these measurements temperature is monitored only using the thermocouple technique. Because the thermocouple is immersed within the liquid pool, the temperature measurement is fairly accurate. Visual observation reveals motion of the surface (i.e. melting) within 10 degrees of the melting temperature of the material, a value consistent with the thermal gradient expected between the thermocouple and the sample surface.

The erosion behavior of the lithium is measured using line emission (at 670.7 nm) from neutral lithium atoms in the plasma column. The atomic physics reaction rate calculations for lithium have been extensively investigated and verified [5]. An absolute calibration of the spectrometer makes this an accurate measurement of the absolute number of lithium atoms in the plasma. Sputtering from any material is composed of an ionic fraction and a neutral atom fraction. Typically the ionic fraction (or so-called secondary ion yield) is small [6], however in the alkaline metals this fraction can become large. The secondary ion yield for solid lithium has been measured to be 66% [7]. In a plasma environment, where a negative potential is established on the wall material (due to the sheath potential), only the neutral atoms will contribute to the sputtering yield. Throughout this paper when we refer to the sputtering yield, we will always be referring to the neutral atom fraction.

The spectroscopic detection system can be operated in two different modes (see Fig. 1). The first, called 'axial mode', measures perpendicular to the magnetic field (i.e. parallel to the sample surface) at different distances along the plasma column from the sample. This method allows a direct measurement of the ionization mean-free path of neutral lithium atoms ejected from the sample surface. The second mode, called 'Doppler mode', switches the viewing direction to be parallel to the magnetic field (i.e. viewing perpendicular to the sample surface). This method allows a measurement of the Doppler shift of the lithium line emission and thereby a direct measurement of the velocity of atoms ejected from the sample surface.

3. Erosion measurements

A temperature dependent material loss rate is expected from a liquid surface exposed to plasma bombardment. Fig. 2 shows the anticipated behavior of the loss rate of a lithium sample as the temperature of the material is increased. At temperatures below the melting temperature, the sample should exhibit a constant loss rate determined by the physical sputtering yield and the

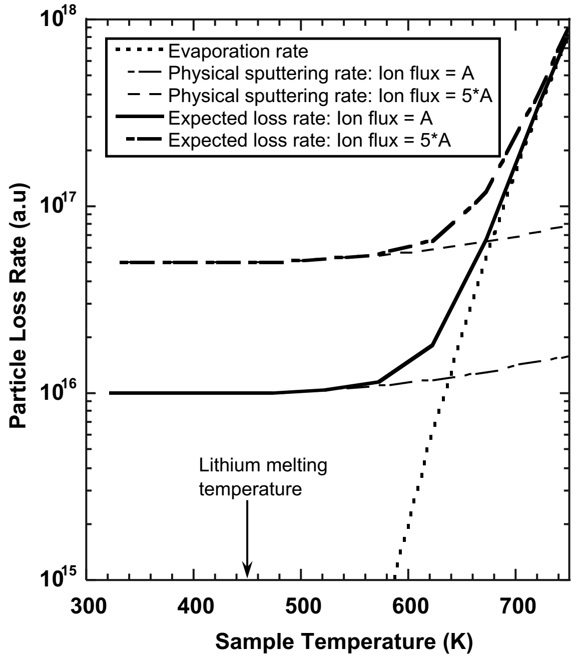

Fig. 2. Erosion rate expected from a lithium sample as the sample temperature increases. Also shown are the expected loss rates at two different ion flux conditions (1 and 5 A). Note that at high temperature the loss rate should become dominated by surface vaporization and become independent of the incident ion flux.

incident ion flux. As the temperature is increased, the sample melts (in the case of lithium at approximately 453 K) and becomes a liquid. The erosion rate increases slightly once the sample melts because the surface binding energy is reduced by the heat of fusion of the material [8] (the heat of fusion is typically only a few percent of the surface binding energy). In these measurements, the change in sputtering due to the solid-liquid phase change would not be observable. As the liquid is then heated further the sputtering rate is empirically predicted to increase slightly according to the Guggenheim relation for the surface binding energy [9]. As the liquid's temperature continues to increase, the evaporation rate will become comparable to and then eventually dominate the material loss rate from the sample. At high temperature, when the evaporation rate is the dominant loss mechanism of material from the sample, the material loss rate will be independent of the incident ion flux to the material.

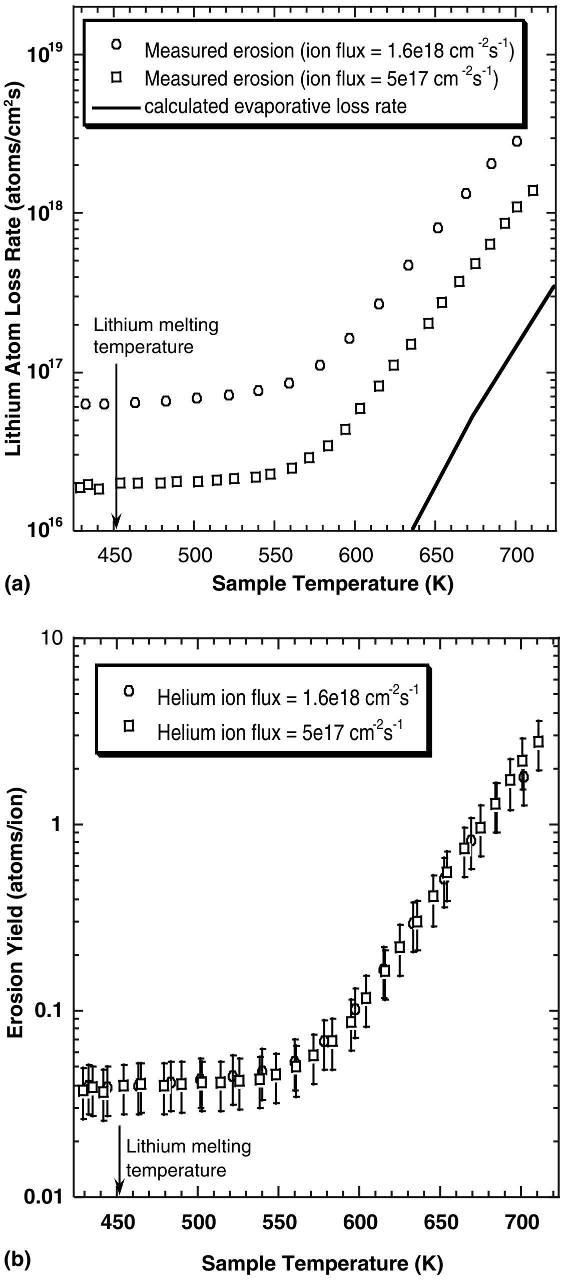

Fig. 3. Measured erosion rate (a) and yield (b) of lithium samples exposed to plasma with 175 eV helium ion bombarding energy at two different ion fluxes. Note that at high temperature the erosion rate is dependent on the incident ion flux.

Before measurements are made, the plasma is used to clean the native oxide/hydroxide layer off the lithium samples. This procedure has been described previously [1]. In addition, to minimize the impact of chemical interactions in the surface of the lithium during the plasma bombardment, helium plasma can be used. Fig. 3(a) shows the measured loss rate of lithium from a sample exposed to two different helium plasma fluxes both with 175 eV incident ion energy. During the solid phase of the measurement, the erosion rate is constant in temperature and varies with the incident ion flux as expected, similar to Fig. 2. As the sample melts little change is observed in

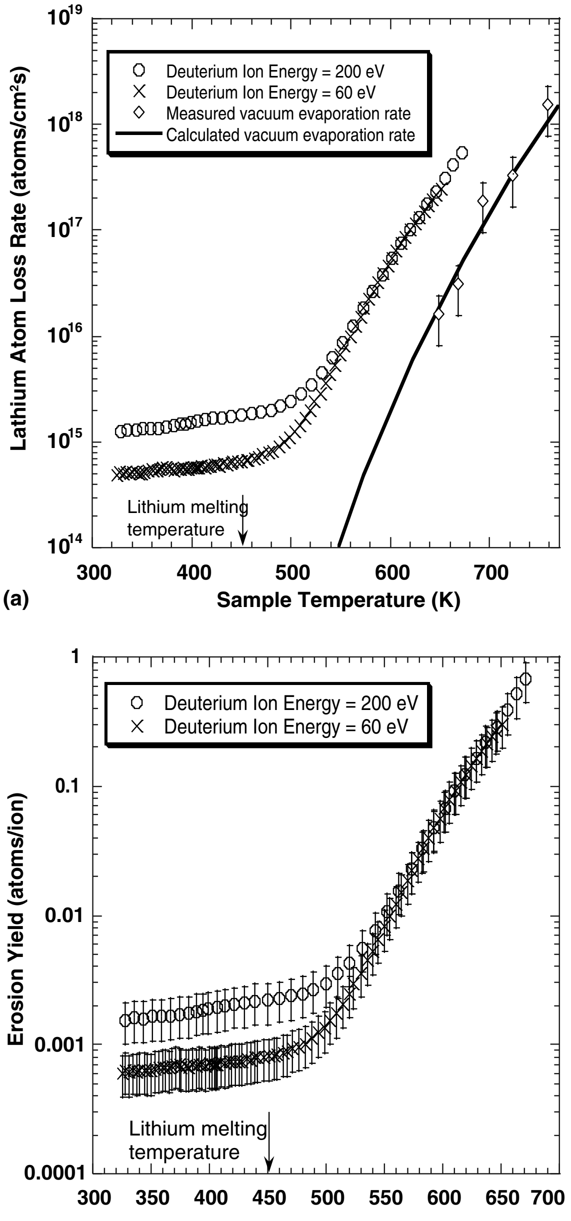

Fig. 4. Measured erosion rate (a) and yield (b) of lithium samples exposed to deuterium plasma bombardment at a constant ion flux, but with different incident ion energy. At high temperature the erosion rate is independent of the incident ion energy, or power flux, indicating that thermal gradients in the liquid samples are not responsible for the enhanced erosion.

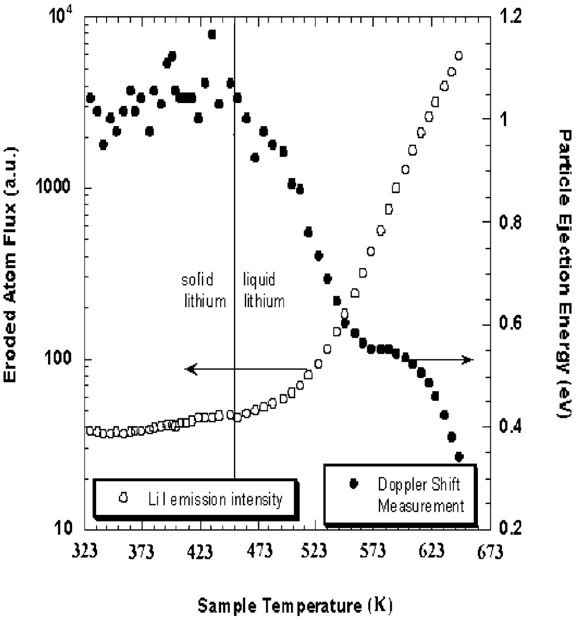

Fig. 5. Average ejection energy of atoms from the lithium sample surface decreases as the number of atoms being removed from the surface increases. At high surface temperature the ejection energy approaches evaporation energies.

the erosion rate, again as expected from Fig. 2. However, as the temperature of the liquid sample increases, the erosion rate begins to increase exponentially at a temperature much lower than that expected from the equilibrium vapor pressure of lithium. In addition, at high temperature the material loss rate from the surface still exhibits a dependence on the incident ion flux, in contrast to the behavior expected from Fig. 2. This fact indicates that the mechanism involved is due to an increase in yield associated with each individual ion striking the surface, as can easily be seen when the data is presented in the standard yield graph (Fig. 3(b)).

Based on the exposure conditions in Fig. 3, the maximum temperature difference across the 2 mm depth of the sample is 3 K (ion flux $= 5 \times 10^{17}$ ions/cm^2 s) and 10 K (ion flux $= 1.6 \times 10^{18}$ ions/cm^2 s). The thermal conductivity of lithium is approximately 0.8 W cm^{-1} K^{-1} [10]. One obvious explanation for these results might be a larger than expected temperature variation between the surface of the sample and the location of the thermocouple within the bulk of the liquid pool during these exposures. However, Fig. 4(a) shows a similar measurement made (in this case with deuterium plasma bombardment of a lithium sample) while the ion flux to the sample is kept constant and the incident ion energy is varied by adjusting the bias voltage of the sample. This measurement clearly shows that the variation of the incident ion energy/power flux, and thereby the ΔT across the thickness of the sample, is not responsible for the discrepancy in the material loss rate behavior. Fig. 4(b) replots the same data in the customary yield format using the experimentally measured flux of 1×10^{18} deuterium ions/cm^2 s. Also shown in Fig. 4(a), is the evaporation rate of one of our lithium samples measured using a quartz crystal oscillator in the absence of plasma. This data agrees with the calculated evaporation rate, providing convincing evidence that our samples are exceptionally clean lithium surfaces and that surface impurities cannot be the cause of this effect.

While the value of the equilibrium vapor pressure fails to account for the magnitude of the material loss rate from these samples, the enhancement is due to an increase in the amount of material evaporating from the surface. Monitoring the ejection energy of atoms emitted from the surface, using the spectroscopic Doppler view mode described in Section 2, shows the average velocity of material being removed from the surface drops as the material loss rate begins its exponential increase. Fig. 5 shows there is a marked decrease in the average ejection energy of atoms from the surface as the temperature increases, indicating a larger fraction of particles are being thermally released.

4. Discussion

Measurements of the atom loss rate from plasma bombarded liquid lithium show enhanced erosion at high surface temperature. The enhancement exceeds that expected from a combination of physical sputtering and vaporization from the surface. The enhanced erosion is observed during either helium or deuterium plasma bombardment of the surface. While these measurements have focussed on the liquid lithium experiments, similar results were also obtained during plasma bombardment of liquid gallium surfaces [2]. Similar, although smaller, increases have also been observed during ion beam measurements on liquid lithium [11]. These results lead us to believe that this phenomenon is generic to all liquid metal plasma-facing components. The implication of these results is that the operational temperature window for any liquid metal plasma-facing component will be smaller than expected based solely on a calculation of the equilibrium vapor pressure of the material.

For example, the maximum operating temperature of a liquid lithium wall, based of the vacuum evaporation rate, has been calculated, for a given plasma configuration, to be 650 K [12]. This is equivalent to a lithium atom flux of 2×10^{16} atoms/cm^2 s (see Fig. 4). Assuming a similar flux (1×10^{18} cm^{-2} s^{-1} in Fig. 4) of deuterium charge exchange and ion flux to the wall, one can see from Fig. 4(a) that a similar lithium atom loss rate is obtained at a temperature of only 575 K, a reduction of 75 K. Practically, what this means is that for a given heat flux to the flowing material, one would have to increase the flow velocity of the liquid metal to keep the temperature rise to an acceptable value.

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