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Section 3. Advanced plasma facing materials

Measurements of erosion mechanisms from solid and liquid materials in PISCES-B

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Abstract

PISCES-B is a steady-state linear-plasma simulator facility dedicated to the study of plasma–material interactions. This paper will summarize recent experiments conducted in PISCES in the areas of: plasma–liquid-metal interactions, the flux dependence of the chemical erosion of graphite, and surface modification to tungsten samples exposed to high-fluence plasmas. Enhanced erosion of liquid-metal samples exposed to plasma is observed for both lithium and gallium samples. The surface stratification of liquid-metals is discussed as a possible explanation. The importance of correction terms in the interpretation of the chemical erosion measurements from graphite samples is then emphasized. And finally, surface analysis of tungsten samples, after deuterium plasma bombardment, has revealed microscopic damage in the surface at ion energies below the sputtering threshold energy. This damage appears to be due to pockets of deuterium gas forming within the tungsten and subsequently rupturing. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

The PISCES devices [1] are steady-state, linear-plasma simulator devices that use a reflex arc to initiate and sustain the plasma discharge. PISCES was the first in a class of high-power plasma simulators used for plasma–material interaction research that has expanded to include simulators in most of the international fusion research programs. Due to its longevity, the PISCES Program has developed an extensive, and redundant, set of both plasma and material diagnostics. These diagnostics provide a high level of confidence in interpreting our experimental results. In addition, a sealed safety enclosure [2] surrounds PISCES-B allowing experiments on toxic materials.

This paper will briefly describe the available diagnostic set before summarizing some of the recent re-

sults from plasma–material interaction studies. The first experimental results discussed will involve the interaction of plasma with free-boundary (i.e. liquid) materials. The PISCES Program has been investigating liquid-lithium and liquid-gallium interactions with both deuterium and helium plasmas. The investigation of free surfaces in contact with intense plasma flux offers several unique challenges. After addressing these challenges and interpreting the results obtained from experiments to date, several more conventional scenarios of plasma interactions with solid surfaces will be discussed.

Carbon has long been the subject of investigations for possible use as a plasma-facing material. However, even though its interactions with plasma have been extensively studied, controversy still surrounds certain aspects of its use. The dependence of its chemical erosion rate on the flux of incident plasma is one such issue [3,4]. An extensive series of flux-dependence experiments were performed on POCO graphite samples in PISCES [5]. The interpretation of these experimental results will be summarized.

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Tungsten also has an extensive history as the subject of plasma–material interaction investigations. As the fluence of the incident plasma has increased during tungsten experiments, new effects have begun to be observed. One such effect is the formation of blisters and pits in the plasma-facing surface [6,7]. Recent results focussing on the behavior of the tungsten surface are also summarized.

2. Experimental apparatus

Of the utmost importance in any plasma physics experiment is the ability to properly interpret diagnostic signals to determine the conditions within the plasma. In PISCES, a set of somewhat redundant diagnostics is used to provide confidence in the interpretation of the signals from these diagnostics. A reciprocating, double-Langmuir probe is used to determine the radial profiles of the plasma density, flux and electron temperature. A least-squares fit to the data using the probe collection calculations of Lafambroise [8] is used. The measurements of plasma conditions from the probe are compared to an absolutely calibrated spectrometer and also compared to the current collected by the sample when biased into ion saturation [5].

Another quantity that is crucial to know in any plasma–material interaction experiment is the impurity concentration in the plasma. Although typically small, these impurity concentrations can dominate such interactions under certain conditions [9]. Again, the absolutely calibrated-spectrometer is used to determine the density of impurities in the PISCES plasma column [10]. Background impurity levels obtained spectroscopically are compared to measurements using a magnetically shielded, high-pressure residual gas analyzer (RGA) during the plasma discharge.

Sample temperature, which often can effect the plasma–material interactions, is measured with optical pyrometers; both a single color (at low sample temperature) and a two-color pyrometers (at sample temperature above 650–750°C). These measurements of the sample temperature are compared to a direct measurement of the temperature of the back surface of the sample using a thermocouple. In the case of the liquid measurements, the thermocouple is inserted through the sample holder into the liquid itself.

Erosion measurements are often based on a straightforward weight-loss measurement. However, even these measurements are cross-checked against erosion-yield measurements calculated from the optical line emission of sputtered sample material which forms an impurity in the plasma column. Spectroscopic views perpendicular to the plasma column, as well as parallel to the column allow for e-folding length measurements of the impurity concentration and Doppler-shift mea-

surements of the ejected target material. Full three-dimensional modeling of the plasma exposure is used to interpret these results [5,11].

In almost all cases the approach in PISCES [12] is to obtain confirmation of the interpretation of a diagnostic measurement by using an entirely different diagnostic technique. This approach allows for an exceptionally high level of confidence in determining the plasma exposure conditions in PISCES.

3. Liquid–plasma interactions

The concept of a flowing-liquid plasma-facing component offers many advantages; power is removed along with the flowing material, the surface is self-healing from an erosion and disruption viewpoint, and certain materials may be used as a pump to remove incident hydrogen isotopes, impurities and perhaps even helium. However, the behavior of such a free surface in contact with plasma is largely an unexplored area. Therefore, the PISCES Program has initiated a systematic exploration of plasma/free surface interactions. The results involving experiments with liquid-gallium are described elsewhere in these proceedings [13]. This section describes measurements made on both solid- and liquid-lithium samples.

A sample preparation technique has been developed to minimize surface contamination of the lithium samples before exposure to the plasma environment. The samples are prepared and then transported in an argon atmosphere to the PISCES-B device, whose samples interlock chamber is purged with argon during sample installation. However, in spite of these precautions, each virgin sample inserted into PISCES-B unavoidably contains a native oxide/hydroxide layer [14] prior to plasma exposure. This oxide/hydroxide layer is often difficult to remove from the lithium in its solid state. For this reason samples are routinely liquefied during exposure to plasma as a means of preparing a clean surface. Fig. 1 shows the neutral oxygen line radiation (from the distinctive triplet near 777 nm) emanating from the sample during such a cleaning run. The OI radiation is not detectable during the plasma bombardment of the solid sample, but is seen to peak as the sample temperature rises and then is observed to decrease as the oxygen is depleted from the surface. Prior to these cleaning exposures the samples are visibly observed to be gray in color, whereas after plasma cleaning the samples appear a metallic, silvery color.

During plasma exposure, line emission from neutral lithium atoms (670.8 nm) in the plasma can be used to provide information on the erosion behavior of the sample. By viewing normal to the magnetic field (i.e. across the plasma column) the axial profile of the emission from the plasma is used to obtain the ionization rate

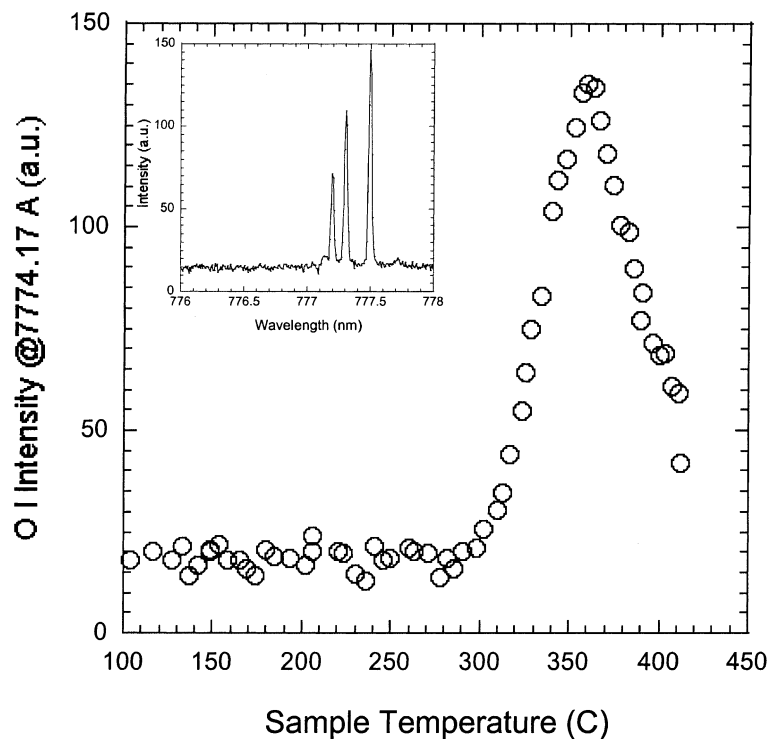


Fig. 1. Variation of the OI line radiation from a lithium sample exposed to helium plasma during a 'cleaning' run. OI emission peaks and then decreases as the surface hydroxide layer is removed. Subsequent plasma exposures show no detectable OI emission from the sample. Inset shows the distinctive three line series of OI emission from the plasma at 777.2, 777.4 and 777.5 nm.

of lithium atoms. These measurements are in agreement with rate calculations based on the Belfast atomic physics database [15]. In addition, by altering the viewing geometry to be parallel to the magnetic field (i.e. along the plasma column) the Doppler shift of the neutral lithium line emission can be used to provide a direct measurement of the ejection velocity of eroded material. For low-temperature measurements, where the lithium sample is solid, the ejection velocity agrees with that calculated from an analytical Thompson model [16] using a surface binding energy of 1.7 eV. However, at higher-temperature when the lithium surface liquefies during the plasma exposure, the average ejection energy of lithium atoms leaving the surface decreases. Fig. 2 is a plot of the average ejection velocity of lithium atoms leaving the sample surface as a function of the sample temperature. (The melting temperature of lithium is 180°C.) An interesting feature of this data is that there does not appear to be abrupt change in the ejection velocity immediately upon the change in state of the sample. Also, the ejection energy during the liquid-interaction phase of the exposure is larger than expected if a thermal release process (0.05 eV) was the dominant loss mechanism.

Since the spectroscopic system is absolutely calibrated, we have also performed measurements of the

erosion yield of lithium atoms from the sample. One expects the material loss rate to equal the sputtering yield at low temperature [17]. As the temperature of the sample increases the vapor pressure of the material will rise and eventually the material loss rate from thermal evaporation will become comparable to, and then dominate, the loss rate from sputtering. However, spectroscopic measurements of the lithium atom loss rate from plasma bombarded samples in PISCES-B do not exhibit this behavior. Fig. 3(a) shows the lithium atom flux leaving the surface of a liquid-lithium sample exposed to deuterium plasma as a function of the sample temperature. In the intermediate temperature regime the material loss rate can exceed, by a factor of 10 or more, the predicted loss rate from the combination of physical sputtering and thermal evaporation. One possible explanation of these results could be the influence of chemical reactions between the incoming deuterium ion flux and lithium atoms on the surface. In effect, a chemical erosion term that increases with temperature could be adding to physical sputtering to give enhanced loss rates. However, similar measurements with helium plasma bombardment of lithium show very similar results, see Fig. 3(b) ([17] uses the TRIM code, but presents no data on helium sputtering of Li, so similar

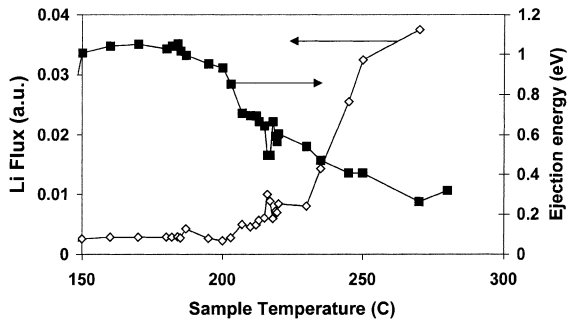


Fig. 2. Spectroscopic measurements of the temperature dependence of the eroded lithium flux from a sample, and the eroded lithium atom ejection energy.

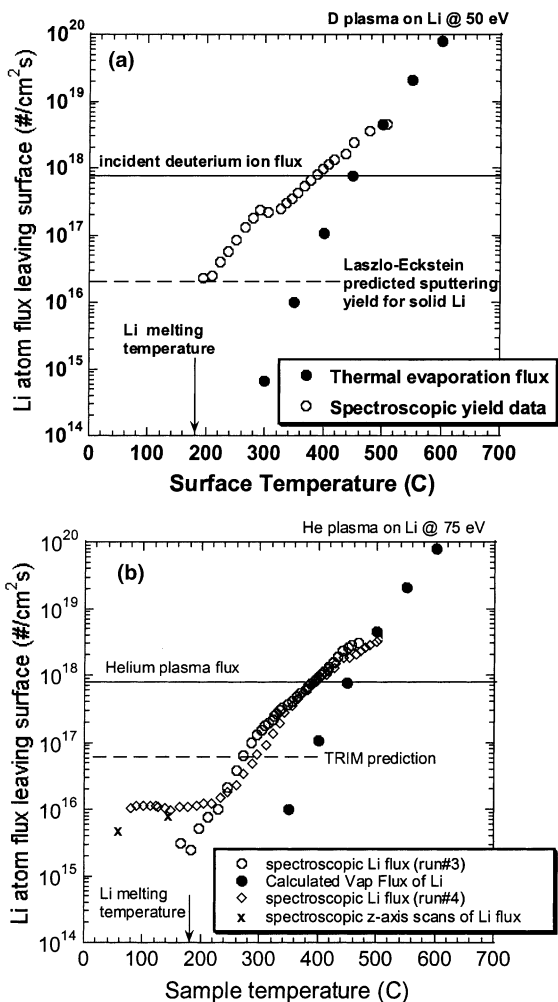


Fig. 3. Enhanced erosion of lithium samples are observed during both deuterium (a) and helium (b) plasma bombardment. The vapor pressure of lithium (dark circles) is not sufficient to explain the loss rates.

calculations were performed for this case). Helium should not form stable molecules with lithium and, therefore, no chemical activity is expected in this case, but the enhanced material loss rate is still observed. Similar enhanced loss rates are also observed during helium plasma bombardment of liquid-gallium samples [13].

At issue here is the shape of the expected erosion curve. Due to space restrictions, the magnitude of the erosion yield at low temperature will be discussed in detail in subsequent publications. However, here it should be noted that these erosion measurements are performed in a plasma environment and therefore only measure the fraction of particles sputtered as neutral atoms. It has been predicted [18] and recently measured [19] that a large fraction ($\approx 2/3$) of the sputtered particles from an alkali metal surface leave as ions. In a plasma sputtered ions will be immediately returned to the surface due to the sheath potential and will therefore not contribute to the sputtering yield. The predictions of the solid Li sputtering rates shown in Fig. 3 are included only to give a point of reference for the reader and to provide some indication of what the shape of the temperature dependence of the sputtering yield might be expected to have.

It has been known that a liquid-metal surface in contact with vacuum will form a striated surface layer extending several monolayers into the liquid bulk [20]. Although first predicted by D’Evelyn and Rice [21] and subsequently observed experimentally in a variety of liquid-metals [22], little is available in the published literature concerning the temperature dependence of this surface structure. In addition, Rice and co-workers have investigated all the alkaline metals, and many other liquid-metals, with the notable exception of lithium [23]. A striated surface layer in contact with plasma could drastically alter the expected erosion rate of the liquid material. As an example of a striated liquid-metal surface, Fig. 4 is a plot of the surface density profile for liquid sodium [24].

Presently, plasma erosion is typically modeled using the TRIM [25] Monte Carlo simulation code. This code

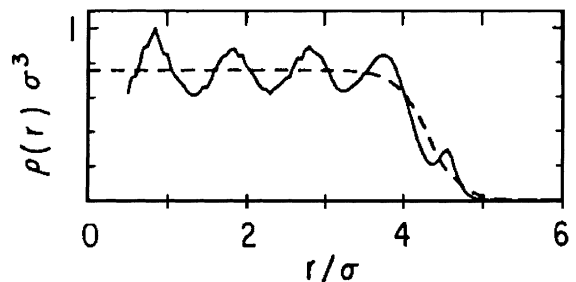


Fig. 4. Surface density profile for liquid sodium. (σ is the atomic diameter.)

assumes an amorphous solid and a binary collision approximation. A comparison has been made of TRIM Monte Carlo calculation results to a molecular dynamics simulation (including a stratified liquid-metal surface) and an increase in sputtering yield at low energy (i.e. below 200 eV when the range of the incident ions is comparable to the stratification layer thickness) has been documented [26]. Unfortunately, these calculations were performed for only a single temperature, so any dependence as the sample temperature increases is still unclear.

We presently hypothesize the following argument to explain our observations of enhanced erosion rates as the sample temperature increases. The density profile of a liquid-metal/vapor interface (seen in Fig. 4 at $r/\sigma \approx 4.5$) shows a small density peak located just beyond the vapor/liquid interface. It is atoms in this peak that may account for the increase in sputtering yield from stratified liquid surfaces. The attractive force to the bulk/surface on a displaced atom goes as r^{-3} (where r is the distance from the surface) and as this peak is shifted away from the surface, the binding force to the surface drops. In addition, the attractive force is proportional to the number of atoms a given distance away, so the attractive force felt from the other atoms in this outermost density layer is also smaller. As the temperature of the liquid increases, this peak shifts further away from the atoms in the bulk liquid (since, by definition, at the critical temperature there is no distinction between the liquid and vapor atoms), reducing still more the binding energy of these outermost atoms. As the binding energy falls, the sputtering yield increases.

This implies there may be a fundamental property of plasma interactions with liquid-metal surfaces that is causing the enhanced material loss rate. If this is true, it could impose a severe restriction on the use of flowing liquid-metals as plasma-facing components in confinement devices. This enhanced loss will limit the operational-temperature window of the material in contact with power flux leaving the device.

4. Flux dependence of carbon chemical erosion

The issue of any dependence of the chemical erosion rate of graphite has been a long debated topic [27–29]. Recent experiments in tokamaks have reignited this debate [30,31]. One of the primary difficulties in any experimental investigation of a flux dependence is the fact that it is not possible to independently vary the flux in any plasma experiment. A variation in flux involves a variation of either the plasma density, or the electron temperature, or both. Changes in either the density, or temperature, can have unanticipated effects on other quantities that in turn effect the interpretation of the experimental results (i.e. incident ion energy, molecular

dissociation chain, redeposited fraction, etc.). Recent experiments in PISCES have performed a flux scan by primarily changing the plasma density [5]. The changes in density on other factors have been both documented and modeled using the WBC code [32] to aid in the interpretation of these contentious results.

Weight loss and CD band spectroscopy (430 nm band head) were used to measure the chemical erosion of graphite samples [5]. The incident ion energy during each exposure was kept constant at 30 eV by applying a negative bias to the samples. The impact of physical sputtering at these ion energies is negligible. Radial and axial plasma profiles were obtained and used for modeling the interaction of the eroded molecules with the PISCES plasma column to determine the appropriate correction for the redeposition fraction, R , to each of the weight-loss measurements. The spectrometer was calibrated with a known deuterated-methane puff (through a small hole drilled in a non-graphite sample) at each flux condition. This calibration procedure allowed for the determination of the corrections needed for the background CD band emission level (contribution from the vessel walls, that changes with plasma condition) to be subtracted from the experimental data. In addition, the variation in the dissociations per photon ratio (the so-called D/XB ratio) with plasma density was also measured. The redeposited fraction and the dissociations per CD band photon are shown in Fig. 5 over the range of the flux scan performed.

The inclusion of these correction terms in the PISCES data is crucial to the correct interpretation of

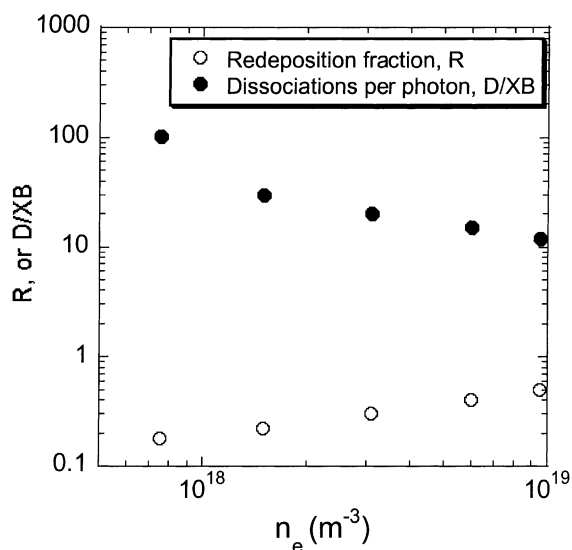


Fig. 5. Redeposition fraction, and dissociations per CD band photon, terms needed to calculate the chemical erosion yield measured by weight loss, and CD band spectroscopy, respectively.

these results. Fig. 6 shows both the raw and the corrected data. Before including the correction terms, clear trends toward decreasing yield with increasing flux are noted on both the weight loss and CD band data (open symbols). However, once the corrections are applied to the data these trends disappear in both set of data (closed symbols). The larger yield measured using the weight loss technique compared to the CD band spectroscopy is interpreted as the chemical erosion of higher order hydrocarbons contributing to the weight loss. We

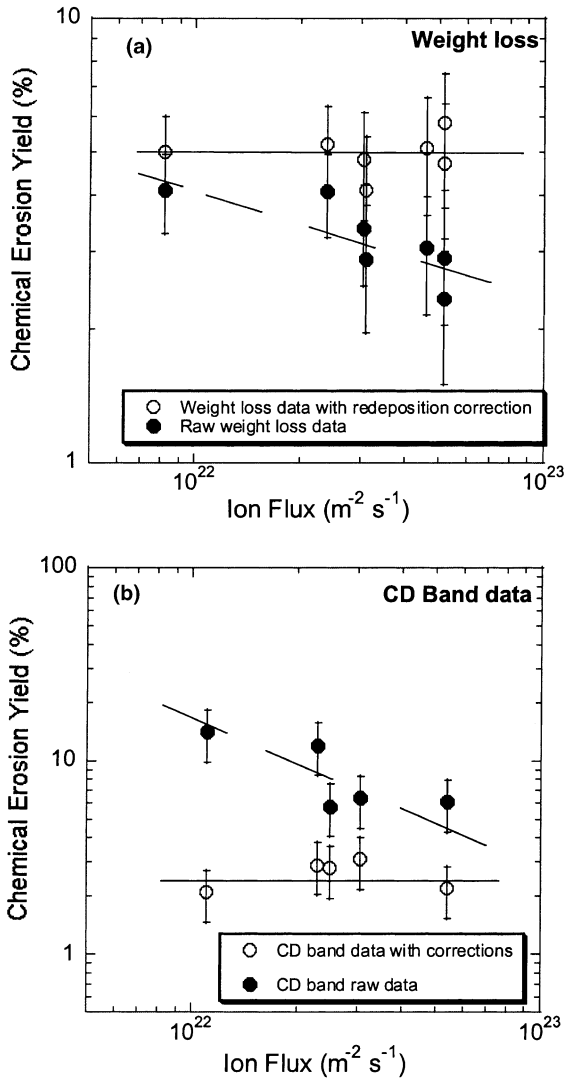


Fig. 6. Raw, uncorrected, chemical erosion yield from (a) weight loss and (b) CD band spectroscopy compared to the corrected data. Correction for (a) is the redeposited fraction from Fig. 5, and for (b) is the appropriate background subtraction for contributions from the vacuum vessel walls. Inclusion of the correction terms eliminates any apparent flux dependence of the chemical erosion yield.

conclude that we observed no flux dependence on the chemical erosion of graphite over the range of flux examined in PISCES.

5. Plasma-induced surface modification of tungsten

Recent observations of modifications to tungsten surfaces exposed to low-energy, high-flux plasma [6] and ion beam [7] bombardment have shown the formation of pits and blisters. Experiments are underway to try to systematically investigate and identify the mechanisms responsible for this behavior.

In the first series of measurements, blister formation during low-temperature sample exposures is being documented. It is believed that the rupturing of blisters, and/or the formation of microcracks in the tungsten surface, may be responsible for a decrease in deuterium retention during low-temperature exposure [33]. The role of defects within the material is being investigated

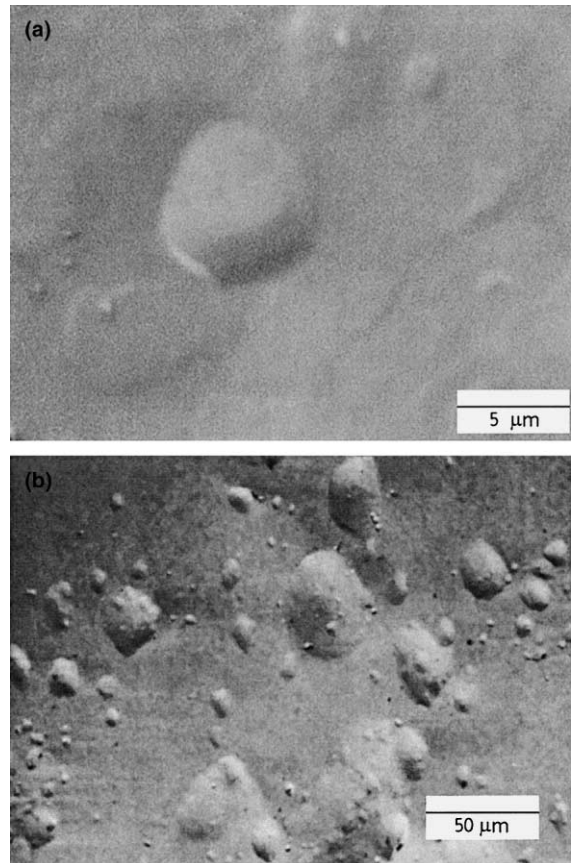


Fig. 7. Blister formation on tungsten samples exposed to deuterium plasma fluence of (a) 7×10^{25} ions/ m^2 and (b) 3×10^{26} ions/ m^2 . The number and average size of the blisters increases with increasing fluence.

by annealing identical cold-rolled tungsten-foil samples at different temperatures before plasma exposure. Although it has been found that unannealed samples (i.e. samples with higher defect densities) tend to form an increased number of blisters during plasma exposure, it is also true that samples annealed to 1200°C have shown blister formation during plasma bombardment. The rate, or amount of blisters formed in the surface, seems to be related to the defect density within the material.

Studies are also being conducted at different plasma fluences to understand the blistering mechanisms. All samples are annealed to 1000°C prior to plasma exposure and exposed to plasma at temperatures close to 550°C. Samples exposed to a deuterium plasma fluence of 7×10^{25} ions/m² exhibited surface blistering, whereas identical samples exposed to a fluence of 7×10^{24} ions/m² did not exhibit any blisters [34]. Exposures at higher fluence tend to result in the formation of a larger number of blisters, many with larger areas. Fig. 7 compares blisters formed at a fluence of 7×10^{25} ions/m² (a) to those formed at a fluence of 3×10^{26} ions/m² (b). Another interesting result of these measurements was that no blistering of tungsten samples exposed to low-energy, high-flux helium plasma bombardment (100 eV ions, 550°C, fluence = 6×10^{25} ions/m²) was observed.

6. Summary and acknowledgments

Three areas of work from PISCES are described in this paper. First, the temperature dependence of the atom flux removed from liquid-metal surfaces during plasma bombardment has been measured to be up to a factor of 10 higher than might be expected from physical sputtering and evaporation losses. Second, no evidence for a flux dependence of the chemical erosion of graphite has been observed over the flux range of 10^{22} – 10^{23} ions/m²s. And, third, a threshold in the fluence (at about 7×10^{25} ions/m²) of 100 eV deuterium plasma ions has been observed for the onset of blister formation on the surface of tungsten samples exposed at 550°C.

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