Available online at www.sciencedirect.com
SCIENCE DIRECT®
Journal of Nuclear Materials 337–339 (2005) 877–881
journal of nuclear materials
www.elsevier.com/locate/jnucmat

High temperature erosion of beryllium

R.P. Doerner [a,*], M.J. Baldwin [a], S.I. Krasheninnikov [a], K. Schmid [b]

[a] *Fusion Energy Research Program, University of California – San Diego (UCSD), Mail Code 0417, Gillman Drive 9500, La Jolla, CA 92093-0417, USA*
[b] *Max-Plank Institut für Plasmaphysik, D-85748 Garching, Germany*

Abstract

The erosion behavior of solid beryllium surfaces exposed to plasma bombardment at high temperature is investigated. The experimentally measured erosion rate of surfaces exposed to energetic particle bombardment at elevated temperature exceeds that predicted by a summation of the physical sputtering rate and the thermodynamic sublimation rate. A model based on the creation of surface adatoms, due to energetic projectile bombardment of surfaces, and their subsequent sublimation is used to explain the enhanced erosion. Molecular dynamics (MD) simulations of beryllium confirm that the experimentally measured evaporation energy (~2 eV) is consistent with the binding energy of adatoms. Of critical importance in this model is the creation rate of adatoms during the surface bombardment. The experimental dependence of the increased erosion on the incident particle flux, particle species and incident particle energy is presented and discussed in relation to the adatom model.

PACS: 52.40.Hf
Keywords: PISCES-B; Beryllium; Evaporation; Erosion and deposition; Ion surface interactions

1. Introduction

Experiments in PISCES-B [1] at UC San Diego, in collaboration with EFDA, are investigating the plasma interaction behavior of beryllium targets as the temperature of the sample approaches its melting temperature. The behavior of beryllium surfaces as a plasma facing material will be critical in determining the actual performance of next step experimental device designs, such as ITER [2] and FIRE [3]. Of a more global nature, the plasma interaction behavior of surfaces exposed at elevated temperatures is fundamental to many branches of technology and surface science.

The generally accepted picture of erosion from a chemically inert surface is one of physical sputtering dominating the erosion rate at lower temperature, until the thermodynamic loss of particles (sublimation from solids and evaporation from liquids) eventually equals and finally exceeds the sputtering rate. Physical sputtering is believed to be independent of the temperature of the surface and thermal sublimation is believed to be independent of the flux of incident energetic particles. However, experimental evidence from a variety of materials [4–8] has shown an enhancement in the erosion rate at elevated temperature. We have proposed an erosion mechanism which depends on both the incident particle flux, as well as the surface temperature [9]. We believe

* Corresponding author. Tel.: +1 858 534 7830; fax: +1 858 534 7716.
E-mail address: rdoerner@ucsd.edu (R.P. Doerner).

0022-3115/$ - see front matter © 2004 Elsevier B.V. All rights reserved.
doi:10.1016/j.jnucmat.2004.09.025

that the erosion mechanism is a fundamental and universal property of materials exposed to energetic particle bombardment at elevated temperature.

2. Model description

Our model of adatom production and subsequent sublimation has been described in detail elsewhere [9], we will briefly summarize the key points of the model to elucidate the improvements to the model that we report on here.

A surface adatom is an atom that acquires sufficient energy during the slowing of an incident energetic particle to become liberated from its lattice location near the surface of a material. The liberated atom, however, does not acquire sufficient energy to be sputtered from the surface and, therefore, creates a surface vacancy and a dislocated atom on the surface itself. As shown in both published MD simulations [10] and our own MD simulations, a certain amount of the created surface adatoms (due to particle bombardment, not condensation) recombine quickly with the surface. Some adatoms generated during such bombardment, however, do not recombine quickly into the surface vacancy they leave behind. Once they are mobile on the surface and leave the vicinity of their associated vacancy, the energy with which they are bound to the surface is reduced. An adatom will diffuse across the surface until it encounters a recombination site (i.e. surface vacancy or other lattice imperfection) or at elevated temperatures, until it sublimates from the surface. Since adatoms are less strongly bound to the surface they will sublimate at lower surface temperatures than the surface atoms.

The time rate of change of the areal density of adatoms, dn_{ad}/dt, can be written as

$$\frac{dn_{ad}}{dt} = Y_{ad}J_{in} - \frac{n_{ad}}{t_{rec}} - \frac{n_{ad}}{t_{sub}}, \tag{1}$$

where the creation rate of adatoms is the adatom yield, Y_{ad}, times the incident energetic particle flux, J_{in} and the loss rates of adatoms are from recombination with surface sites, t_{rec}, and from sublimation, t_{sub}. In equilibrium this means that the surface density of adatoms, n_{ad}, is

$$n_{ad} = \frac{Y_{ad}J_{in}}{\frac{1}{t_{rec}} + \frac{1}{t_{sub}}}. \tag{2}$$

The total atom loss rate, J_{out}, from a surface bombarded by energetic particles is written:

$$J_{out} = Y_{ps}J_{in} + n_{ad}/t_{sub} + n_0/t_0, \tag{3}$$

where Y_{ps}, is the physical sputtering yield and n_0/t_0 is the standard thermodynamic sublimation rate from the material. Eq (3) can be rewritten as

$$J_{out} = Y_{ps}J_{in} + \frac{Y_{ad}J_{in}}{1 + \frac{t_{sub}}{t_{rec}}} + K_0 n_0 \exp\left(\frac{-E_0}{T}\right), \tag{4}$$

The time constant associated with sublimation of an adatom, t_{sub} = constant * $\exp(E_{ad}/T)$ and that associated with recombination, t_{rec} = constant * $\exp(E_D/T)$. Here E_{ad} is the binding energy of an adatom to the surface and E_D is the activation energy for surface diffusion to a recombination site. Eq. (4) can then be rewritten in the form, where $E_{eff} = E_{ad} - E_D$:

$$J_{out} = Y_{ps}J_{in} + \frac{Y_{ad}J_{in}}{1 + A\exp\left(\frac{E_{eff}}{T}\right)} + K_0 n_0 \exp\left(\frac{-E_0}{T}\right). \tag{5}$$

In our previous work [9], the assumption was made that recombination of surface adatoms dominated their sublimation (i.e. $A\exp(E_{eff}/T) \gg 1$), leading to:

$$J_{out} = Y_{ps}J_{in} + n_{ad}K_{ad}\exp\left(\frac{-E_{eff}}{T}\right) + K_0 n_0 \exp\left(\frac{-E_0}{T}\right). \tag{6}$$

However, Eq. (6) is not strictly correct and does not take into account the possibility of sublimating a large fraction, or possibly even all, the adatoms created. The difference between Eqs. (5) and (6) is exhibited in Fig. 1. In order to clearly see the distinctions between the two equations the thermodynamic sublimation term has been omitted from the plot of both equations. Both equations accurately depict the experimental data over its temperature range, but at higher temperature the rate of sublimation of adatoms is restricted to their creation rate in Eq. (5), whereas Eq. (6) allows the sublimation rate of adatoms to increase indefinitely.

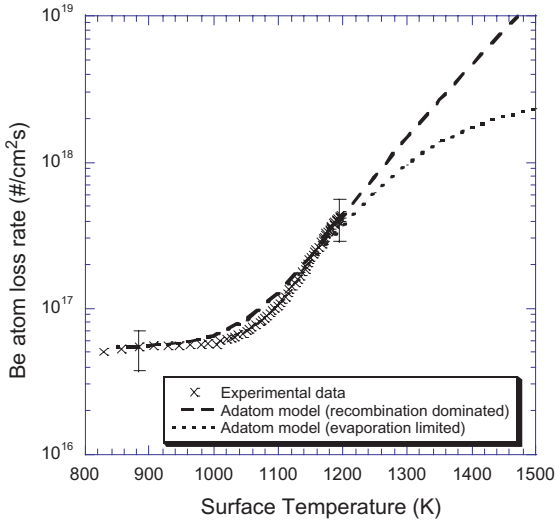

Fig. 1. Comparison between the adatom model dominated by surface recombination and this model which includes the limiting effect of substantial sublimation/evaporation of adatoms from the surface before recombination.

3. Comparison to experiments

Fig. 2 plots each of the three terms in Eq. (5) along with the experimental data obtained during deuterium plasma bombardment of a Be sample biased to −50 V. The ion flux measured during these experiments was 4×10^{18} ions/cm^2 s. The unknowns present in the adatom term of Eq. (5) were allowed to vary to obtain a best fit of the data. The values of those quantities used here are; $E_{\text{eff}} \sim 2$ eV, $A = 2 \times 10^{-7}$ and $Y_{\text{ad}}/Y_{\text{ps}} = 50$. It is encouraging to note that the best fit to the data still occurs at an adatom binding energy that agrees with the measured value and with MD simulations of a Be surface [9]. The numerical values of these terms are quite similar to those used previously in [9]; $E_{\text{eff}} \sim 2$ eV, $A = 1 \times 10^{-7}$ and $Y_{\text{ad}}/Y_{\text{ps}} = 20$. The primary difference being the adatom creation yield. However, a value of 50 for $Y_{\text{ad}}/Y_{\text{ps}}$ at 50 eV bombarding energy is in better agreement with MD calculations [10]. As is evident in Fig. 2 the inclusion of the adatom term produces an increase in the erosion rate over a finite temperature range. At sufficiently high temperature, the erosion rate asymptotically approaches the thermodynamic sublimation rate from the material.

The observation of enhanced erosion is observed over a variety of experimental conditions. Fig. 3 shows the erosion rate from Be samples exposed to a series of incident ion energies. Also shown in the figure are the associated fits to the data using Eq. (5). Only at the extremely low energy of 25 eV does the model appear to deviate from the data. However, at these low energies the threshold value for adatom creation is being approached and the adatom yield may change significantly. No attempt was made to obtain a better fit to the data with different values of E_{eff}, A and $Y_{\text{ad}}/Y_{\text{ps}}$.

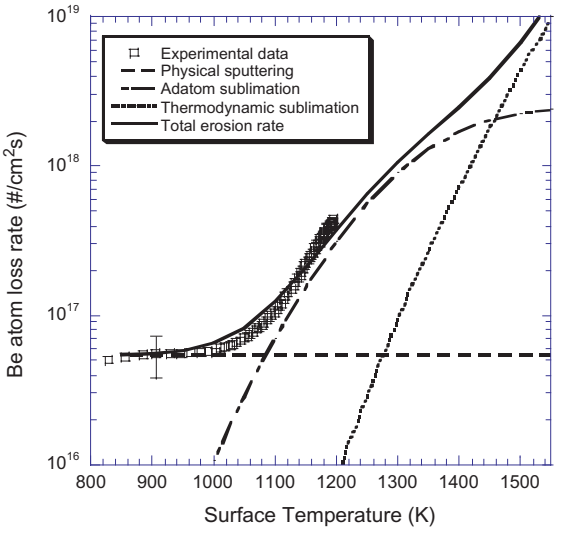

Fig. 2. Each of the three terms in Eq. (5) are plotted separately, along with the comparison of their sum to the experimental data collected during a −50 V bias run.

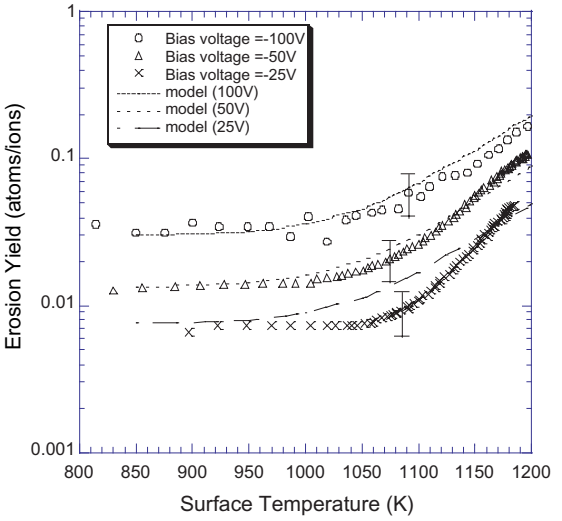

Fig. 3. Comparison of three different bias voltages during deuterium plasma bombardment of Be samples with the model predictions for each case.

It should be noted that in the discussion above all three terms, E_{eff}, A and $Y_{\text{ad}}/Y_{\text{ps}}$, are treated as variables to justify the model. In reality, however, the effective binding energy can be calculated from an MD simulation [9] and the ratio of the adatom to sputtering yield (which depends strongly on the incident ion energy) is predicted from MD simulations [10]. The variable A is the only unknown that is evaluated by fitting to the experimental data at a single surface temperature.

The rate of release of adatoms, and hence the significance of this term in Eq. (5), is also dependent on the incident energetic particle flux, J_{in}. The model can be exercised by keeping all parameters fixed and varying only the incident particle flux. Fig. 4 shows the magnitude of the adatom erosion effect over several orders of incident flux. The adatom erosion effect becomes more pronounced at higher levels of incident particle flux. Also indicated in the figure are typical values of particle flux in the PISCES-B device and in ion beam facilities. Because of the low flux values achievable in ion beam facilities, thermodynamic sublimation will tend to dominate the erosion rate before the adatom terms becomes significant.

Another drawback to observing adatom erosion in ion beam devices is the fact that they tend to operate at higher incident energies. Fig. 5 shows the same flux scan using Eq. (5), but assuming an incident ion energy of 3 keV D^+. The adatom yield at higher energies is smaller due to a decrease in the interaction of the incident particles with the surface of the material. Fig. 5 assumes $Y_{\text{ad}} = 4$ at 3 keV [10]. The combination of these

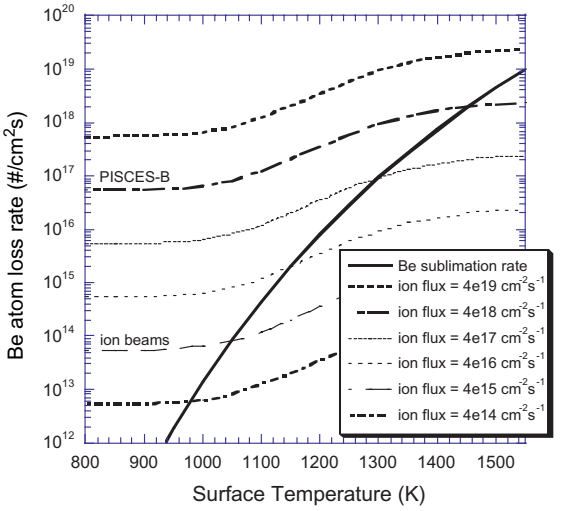

Fig. 4. Model predictions of 50 eV deuterium ion bombardment of Be at different incident ion flux. Typical flux values of PISCES-B and ion beam devices are indicated.

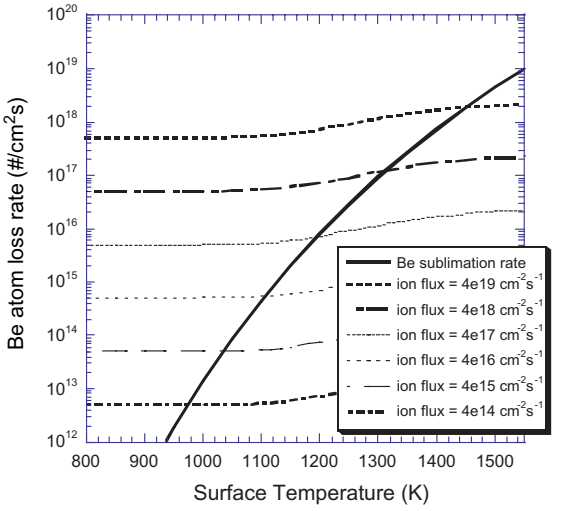

Fig. 5. Model predictions of 3 keV deuterium ion bombardment of Be at different incident ion flux. The smaller adatom yield at higher energy leads to smaller magnitude effects.

effects may make this phenomenon challenging to observe during ion beam measurements [11].

This is not to say, however, that adatom erosion cannot be observed in ion beam facilities. Since no temperature dependent erosion measurements of Be are available from ion beam facilities, we can exercise our model on enhanced erosion of lithium data from ion beams available in the literature [12]. Fig. 6 shows this comparison. Values of $E_{\mathrm{eff}} = 1$ eV, $A = 1 \times 10^{-7}$ are used in the model calculation, which compare well to values obtained previously from PISCES-B ($E_{\mathrm{eff}} = 1.1$ eV and

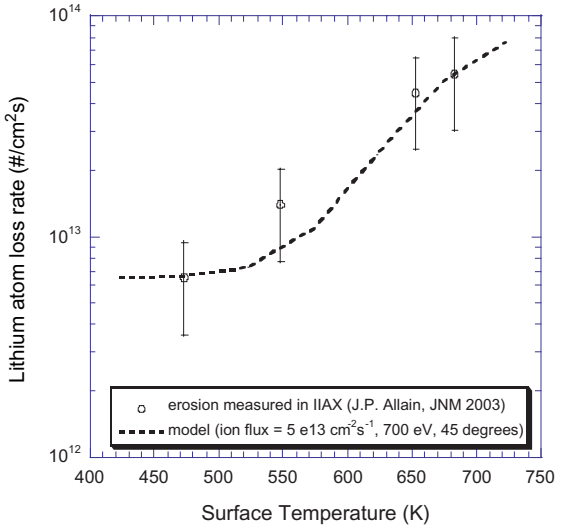

Fig. 6. Comparison of model to ion beam erosion measurements of lithium [12].

$A = 1 \times 10^{-6}$ [9]), especially considering the four orders of magnitude difference in ion flux between the two facilities. An increase in the ratio $Y_{\mathrm{ad}}/Y_{\mathrm{ps}}$ (by a factor of 3 above the value associated with 700 eV bombardment at normal incidence [10]) has also been included in the model to take account the 45° incidence angle in IIAX. In order to observe enhanced adatom erosion in ion beam devices, it is necessary to maximize the ion flux in the beam, and at the same time maximize the ratio of $Y_{\mathrm{ad}}/Y_{\mathrm{ps}}$ (i.e. operate using low energies). In addition, we speculate that by moving toward grazing incidence and thereby increasing the interaction with the surface layers, it may be possible to maximize the creation rate of adatoms even at larger incident particle energies.

4. Summary

The model for adatom evaporation has been modified to account for the condition where adatom sublimation dominates over adatom recombination into the surface. As the number of adatoms sublimating approaches the number created, the enhanced erosion saturates. This saturation phenomenon, if observed experimentally, may prove the validity of the model. The dependence of the adatom model on variations of experimental parameters has been described.

Acknowledgments

These experiments are only possible because of the professionalism and dedication of the PISCES Be workers. We gratefully appreciate their effort. We also value

the many valuable discussions we have had with members of both the US and European PFC communities. This work was performed as part of the US-EU Bilateral Collaboration on PMI Experiments on C/Be/W Mixed-Materials for ITER and supported under USDOE contract DE-FG03-95ER-54301.

References

[1] D.G. Whyte, G. Tynan, R.P. Doerner, J.N. Brooks, Nucl. Fusion 41 (2001) 47.
[2] G. Federici, P. Andrew, P. Barabaschi, et al., J. Nucl. Mater. 313–316 (2003) 11.
[3] D.M. Meade, Fus. Eng. Des. 63 (2002) 531.
[4] R.P. Doerner, M.J. Baldwin, S.I. Krasheninnikov, D.G. Whyte, J. Nucl. Mater. 313–316 (2003) 383.
[5] R.W. Conn, R.P. Doerner, F.C. Sze, et al., Nucl. Fusion 42 (2002) 1060.
[6] E.P. Vaulin, N.E. Georgieva, T.P. Martynenko, L.V. Feoktistov, Sov. J. Plasma Phys. 2 (1981) 437.
[7] J. Roth, W. Moller, Nucl. Instrum. and Meth. B 7&8 (1985) 788.
[8] V. Philipps, E. Vietzke, H. Trinkaus, J. Nucl. Mater. 179–181 (1991) 25.
[9] R.P. Doerner, S.I. Krasheninnikov, K. Schmid, J. Appl. Phys. 95 (2004) 4471.
[10] H. Gades, H.M. Urbassek, Phys. Rev. B 50 (1994) 11167.
[11] V. Philipps, these Proceedings.
[12] J.P. Allain, M.D. Coventry, D.N. Ruzic, J. Nucl. Mater. 313–316 (2003) 641.