Low Energy Data on Radiation Enhanced Sublimation of Graphite

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ABSTRACT

Erosion of POCO graphite by helium in PISCES-A was measured by carbon spectroscopy for a temperature range from 900º-2000ºC, ion energies of 30-300 eV, ion fluxes of 1-6 x10^{22} m^{-2} s^{-1} and electron temperatures of 4-22 eV. Yields at low energies were higher than predicted in current models. The role of redeposition is discussed.

I. Introduction

Radiation-enhanced sublimation (RES) is unique to carbon-based materials and high erosion rates occur when, at elevated temperatures (>1200ºC), these materials are exposed to ion bombardment from plasmas. Data and models describing RES[1-11] were recently reviewed by Philipps.[12] Both the impurity generation and the loss of material due to erosion are of concern in fusion applications. For example, RES is involved in the onset of carbon catastrophes in the Joint European Torus (JET) and of carbon blooms in the Tokamak Fusion Test Reactor (TFTR). RES also severely limits the operating temperature for carbon-based plasma facing materials in the Compact Ignition Tokamak (CIT) and the International Thermonuclear Experimental Reactor (ITER).

Our previous paper[13] presented weight loss data and confirmed that the roughly linear dependence of RES yield upon flux persists at high fluxes. These data extended the flux range for RES data to high fluxes (~10^{23} ions/m^2) relevant for CIT and ITER. This paper provides more detailed information on the energy and temperature dependences of RES obtained from spectroscopic measurements of carbon erosion from POCO graphite in helium plasmas.

II. Experimental Technique

Our earlier paper[13] gives information on experimental conditions and references to other PISCES work. The experimental conditions include sample temperatures from 800º to 2000ºC, helium (He) bombardment energies from 20 to 250 eV, ion fluxes of 10^3-10^4 A/m^2, electron temperatures (T_e) from 4 to 22 eV and plasma densities from 10^{17}-10^{18} m^{-3}. He plasmas rather than hydrogen were used to avoid effects from hydrocarbon formation. The vacuum prior to gas fill was typically 1-2 x 10^{-6} torr.

Erosion rates were monitored spectroscopically with a 1.3 meter Czerny-Turner monochromator using the C-I line at 9095Å. Data were adjusted for changes in photon efficiency with T_e. Values of efficiencies in the range of 5-10 eV were extrapolated from data above 10 eV. An experiment performed in conjunction with this one gives more detail on spectroscopic techniques.[14]

Spectroscopic data were obtained at low energies where a strong effect from a threshold energy would be expected. The bombarding energies were established by electrically biasing the sample with respect to the plasma chamber. The ion bombardment energy, given by

the difference between the bias and the space potential of the plasma, was typically less than 10 eV in these experiments. Unless noted, reported values of ion bombardment energy are corrected for the space potential. Ion bombardment in PISCES-A occurs at perpendicular incidence because the potential drop due to the bias greatly exceeds the ion temperature (range of 1-3 eV).

We define "spectroscopic yield" as the spectroscopic signal corrected for the photon efficiency (ε) and divided by flux (Γ in A/cm^2).

$$\text{spectroscopic yield} = \text{signal} \times [4\pi\, \varepsilon / \Gamma] \qquad (1)$$

The form of the expression is standard but in these tests most neutrals emitted from the sample do not become ionized so that an "escape" branch exists. Since the "spectroscopic yield" is a suitable parameter for the discussion regarding the dependence of RES on energy and temperature, no calibration to absolute values of yield was done.

III. Results and Discussion

Spectroscopic yield versus sample temperature and energy are shown respectively in Figures 1 and 2. These data will be compared with predictions by the model in current use. Possible effects from redeposition in interpreting these data will also be discussed.

Let us begin with a brief review of the current model.[10] The expression below for the yield from physical sputtering and RES has been applied to ITER.[11] Values of 41 and 42 eV for the threshold for He and C, respectively, have been used in the model.

$$Y = Y_{phys.} + Y_{RES} = F1\{E_{th}\}\ F2\{E_{TF}\}\ Q \qquad (2)$$

$$Q = Q_{phys.} + Q_{RES} = Q_{phys.} + 54\ m^{1.2} \exp(-0.78/T) \quad \text{for T in eV} \qquad (3)$$

F_1 and F_2 depend respectively on the threshold energy, E_{th}, and the Thomas Fermi energy, E_{TF}. Q has a temperature independent term for physical sputtering and a temperature dependent term for RES. The value -0.78 eV in the exponent corresponds to an activation energy for RES and will be compared to the value extracted from our data.

3. Temperature Dependence of RES Yield

The temperature dependence in Figure 1 is consistent with the data in our previous paper.[13] The more numerous spectroscopic data here permit a conclusion regarding temperature dependence. From Equation 3 we would expect the slopes in Figure 1 to be proportional to $0.78/T^2$ and that ln(yield) versus 1/T would have a slope of -0.78.

Figure 3 shows spectroscopic yield versus 1/T with data at low temperature excluded. The slopes (-0.41 to -0.50 for $\log_{10}$) correspond to -0.94 to -1.15 eV for an activation energy. The values are 20% - 50% greater than the activation energy used in the model (-0.78 eV).

The amount of redeposition anticipated in these experiments is believed to be small but was not rigorously quantified. Redeposition of some carbon is likely and this would steepen the slope in Figure 3. The equations below describe carbon erosion in a 1-D geometry for a He plasma where RES is the dominant erosion mechanism and other erosion channels are neglected.

$$\Gamma_{Cg} = \frac{\Gamma_{He}\, Y_{HeRES}}{1\text{-}Y_{CRES}\, R_C} \qquad \text{gross erosion} \quad (4)$$

$$\Gamma_{Cn} = \frac{\Gamma_{He}\, Y_{HeRES}\, (1\text{-}R_C)}{1\text{-}Y_{CRES}\, R_C} \qquad \text{net erosion} \quad (5)$$

The Γ's are fluxes; Y_{HeRES} and Y_{CRES} are the RES yields of graphite bombarded by He and C respectively; and R_C is the fraction of carbon leaving the surface that is redeposited on the sample. The effect that redeposition would have on data showing the temperature dependence of RES can be seen from the expression below, derived from Equations 2- 4. (Note: both Y_{HeRES} and Y_{CRES} depend on temperature.)

$$\Gamma_{Cg}/\Gamma_{He} = \frac{Y_{HeRES}}{1-Y_{CRES}R_C} = \frac{K_{He}\exp(-0.78/T)}{1-K_C\exp(-0.78/T)R_C} \quad (6)$$

$$\frac{\partial[\ln(\Gamma_{Cg}/\Gamma_{He})]}{\partial[1/T]} = \frac{-0.78}{1-Y_{CRES}R_C} \quad (7)$$

Redeposition produces a higher apparent slope. Its effect is significant only when the product of Y_{CRES} and R_C is significant. From 1200º to 2000ºC, the RES self sputtering of carbon at 100-300 eV is near unity[9] (range of 0.8 to 1.1). Redeposition of 25%, if it occurred, would increase the apparent activation energy by about 30%. The amount of redeposition in this experiment is presumed to be much less than 25%. Extracting an estimate of redeposition by comparing the measured slopes to the model is unsatisfactory because these low energy data are not consistent with the energy dependence of RES yield in the model.

4. Energy Dependence of RES Yield

Figure 2 shows raw data for spectroscopic yield versus energy. At low energies, the implied curves decline less rapidly than one would expect for a threshold energy for RES of about 40 eV, a value within the generally accepted range of 25-40 eV for the displacement damage energy in carbon.[15] The 1500ºC data in particular suggest that some mechanism produces significant yields at energies as low as 30 eV.

Implicit in this conclusion are two concerns regarding interpretation of the data. First, a constant background signal would add proportionately more to lower yield values and thereby produce a curve of apparent yield with a gentler slope. Second, does redeposition have some effect?

In Figure 4, the 1625°C, 1500°C and 1400°C data from Figure 3 are replotted with a value of 1.8×10^5 (equal to the 1400°C point at 17 eV) taken as the background level (for the data taken on 10-19) and subtracted from the other data for 1400°C and 1600°C. With the corrections, the 1400°C and 1625°C data are more consistent with the 1500°C data. Figure 4 also shows a curve for the RES yield at 1500°C calculated using the model and normalized to our data at 300 eV.

Before discussing the effect of redeposition, more comments about background signals are appropriate. To observe the background signal, some runs were made with the sample retracted downstream from the spectroscopically monitored plasma volume to a distance much longer than λ_{mfp} for RES. In these tests, the signal decreased with distance but did not fall to zero and still increased with bias. (The distance was still much shorter than λ_{mfp} for physical sputtering, so some signal from physical sputtered carbon would be expected.)

Carbon from deposited films on the chamber wall, hypothetically a secondary source of carbon, could be released through self sputtering when physically sputtered carbon from the sample strike the walls, and one would expect this signal to increase with bias. The wall area is large; however, physical sputtering from sample is the only source of energetic carbon and the net yield of secondary carbon would be proportional to the square of this yield, which is about 0.1 for energies near 100 eV and much less at lower energies, so the effect is small.

Also, oxygen (impurity) in the plasma would produce significant sputtering. The yield for oxygen is near unity for a wide range of energies. Release and excitation of carbon as molecules break up in the plasma might contribute directly to the signal, or ionized carbon from this break up could return to the sample and cause RES self sputtering. Mechanisms that introduce carbon which then self sputters the sample have an effect similar to redeposition.

Redeposition of carbon does not reduce the apparent threshold energy. The spectroscopic signal (and the data in Figures 3 and 4) is proportional to the gross RES yield (Γ_{Cg}/Γ_{He}). Although Equation 6 has no explicit energy dependence, we can deduce that the curve of spectroscopic yield versus energy will have a steeper slope when redeposition occurs because Y_{CRES} increases with energy, and thus the denominator will decrease with increasing energy.

Of the mechanisms mentioned above, only the direct contribution of excited carbon from sputtering by oxygen would be a concern here. In this regard, the correction made in deriving the data in Figure 4 would seem admissible, since the result is then more consistent with the data at 1500°C. However, other data in Figure 3 with signal intensities below the value taken above as "background" indicate this value may overestimate any background signal. The fundamental point is that even with the corrections for background, the measured yields at low energies are significantly higher than values given by the model.

6. Conclusions and Recommendations

The principal conclusion of this work is that the current model of radiation enhanced sublimation appears to underpredict the yields at low ion energies. One possible hypothesis consistent with these new data is that less well bound carbon atoms are being preferentially dislodged. The near surface of samples exposed to high flux ion bombardment rapidly accumulate radiation damage. The current basic premise for RES, that dislodged carbon interstitials migrate to the surface and are released, would require little modification to include a subpopulation of less well bound carbon atoms, e.g., on platelet edges or adjacent to damage sites, that contribute exclusively to RES at energies below the energy for creation of a Frenkel pair in undamaged graphite.

The initial tests in PISCES show several effects that warrant further study, specifically the RES yield at low energies and the effects of redepostion. The effects of redeposition should be studied with redeposition fractions approaching unity. Such studies can be done at the PISCES facility but require a different experimental arrangement than the experiments reported here.

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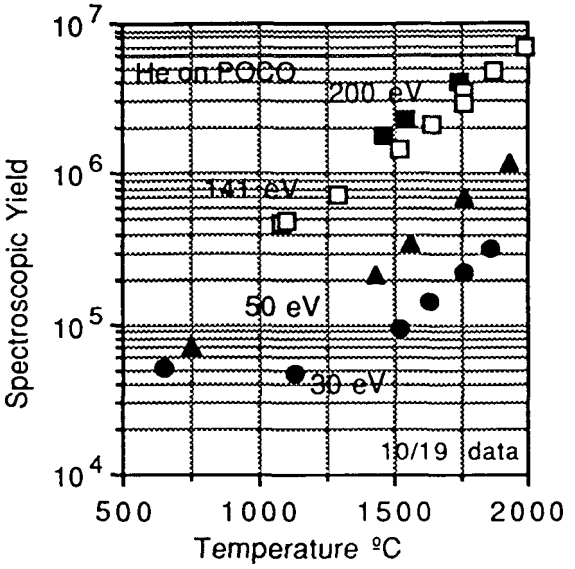


Figure 1: Spectroscopic yield versus sample temperature for He on POCO

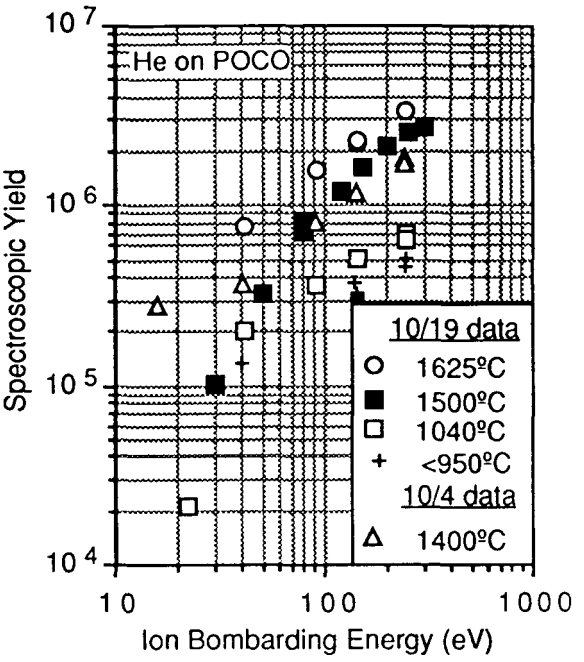
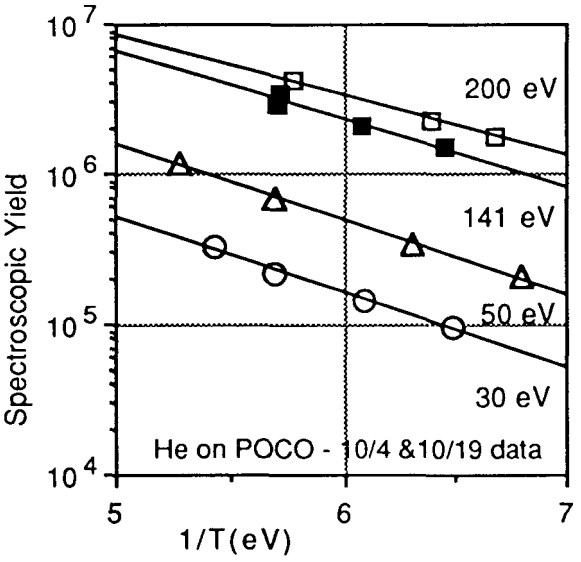


Figure 2: Spectroscopic yield versus ion energy for helium on POCO graphite



least squares fit for slopes

y200 = 9.2E8 x 10^(-0.41x)
y141 = 11.8E9 x 10^(-0.45x)
y50 = 4.5E8 x 10^(-0.49x)
y30 = 1.5E8 x 10^(-0.50x)

Figure 3: Spectroscopic yield versus inverse sample temperature (in eV) with slopes for least square fit

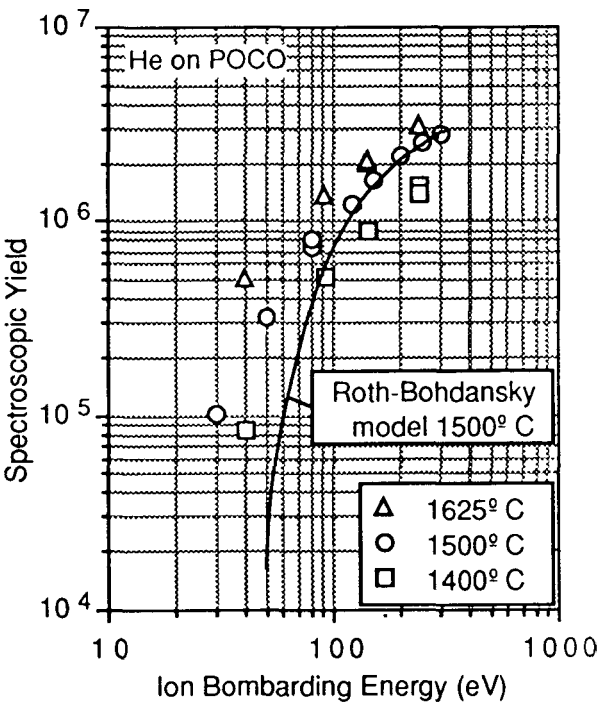


Figure 4: Spectroscopic yield corrected for background versus ion energy