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Helium-ion-induced release of hydrogen from graphite*

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R. A. Langley

Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831

The ion-induced release of hydrogen from AXF-5Q graphite was studied for 350-eV helium ions. The hydrogen was implanted into the graphite with a low energy (~ 200 eV) and to a high fluence. This achieved a thin (~ 10 -nm), saturated near-surface region. The release of hydrogen was measured as a function of helium fluence. A model that includes ion-induced detrapping, retrapping, and surface recombination was used to analyze the experimental data. A value of $(1.65 \pm 0.2) \times 10^{-16} \text{ cm}^2$ was obtained for the detrapping cross section, and a value of $(0.5-4) \times 10^{-14} \text{ cm}^2/\text{s}$ was obtained for the recombination coefficient.

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I. INTRODUCTION

This experiment was undertaken to measure the release of hydrogen implanted into POCO AXF-5Q graphite for low-energy (~ 350 -eV) helium ions. Previous experiments in fusion plasma devices¹ have used helium discharges to "condition" the wall and internal components by obtaining wall pumping, which subsequently led to well-controlled tokamak discharges. A major fraction of the surface area of the wall and internal components consisted of graphite; specifically, POCO AXF-5Q graphite was used in the Tokamak Fusion Test Reactor (TFTR). In a previous investigation,² Wampler and Myers studied the ion-induced release of deuterium from carbon by hydrogen, helium, and carbon ions at energies from 3 to 60 keV. Their model for the release included retrapping of the detrapped deuterium, in contrast to previous work³ in which the release was parameterized using two detrapping cross sections for the release. In modeling the present results, there is no need to invoke either retrapping or the two detrapping cross section approach. This model uses only detrapping and recombination to describe the release. In addition, it was found that very little (if any) hydrogen diffused from the bulk to the depleted surface layer at room temperature.

The experimental procedure and results are described in Sec. II. The model and the results obtained with it are summarized in Sec. III.

II. EXPERIMENTAL PROCEDURE AND RESULTS

The experimental arrangement used for this measurement was the same as that used to study the retention of hydrogen in graphite.⁴ A schematic diagram of the experiment is shown in Fig. 1. The system is pumped by a turbopump-forepump combination; the base pressure after a mild bakeout at 100°C is less than 5×10^{-7} Torr. A residual gas analyzer (RGA) is installed in the pumpout arm and connected to the discharge chamber with a relatively large valve and a variable leak. For this measurement, only the variable leak

was used. The gas flow through the variable leak was adjusted so that the RGA signal for mass 4 was easily read and the anticipated signal for mass 2 could be observed. The flow rate through the variable leak was measured before and after each hydrogen release run; it had an insignificant influence on the total pressure measurements.

The graphite used for this experiment was POCO AXF-5Q, as in the hydrogen retention experiments.⁴ The surface region was saturated with hydrogen to a concentration of about 0.5 H/C (Ref. 5) and to a depth of ≈ 10 nm (Ref. 6) before the release run. For the experiment, the glass bell jar was filled with helium to 172.6 mTorr and a glow discharge was initiated with an applied voltage between the anode and the cathode of 400 V and with a current of 115 mA (i.e., 5×10^{14} He/cm²·s). The energy of the impinging helium ions is estimated to be 350 ± 25 eV (Ref. 7). The total pressure and the mass 2, 3, and 4 signals from the RGA were measured as functions of elapsed time after discharge initiation. The result for the mass 2 release is shown in Fig. 2 as open circles (0 h). The mass 4 signal remained constant within a measurement uncertainty of 2%. Previous results⁵ have shown only very small retention of implanted helium, consistent with the present results. After 600 s the discharge was terminated and the bell jar was pumped out. The procedure was repeated 1 h after the first run was completed (triangles in Fig. 2) and 23 h after the second run was completed (squares in Fig. 2). The RGA mass 2 signal shown in Fig. 2 was calibrated using the total pressure rise measured with the capacitance manometer. A second procedure based on ionization and RGA transmission efficiency was used to calibrate the RGA for hydrogen; the results were found to be in agreement with the total pressure measurement calibration to within 5%. The total pressure measurement time response of 0.5 s was less than the RGA signal time response of ~ 1 s. After this series of runs with helium gas, the bell jar was evacuated and filled with H₂ to a pressure of 155 mTorr. A glow discharge was initiated and the pressure was measured as a function of elapsed time from discharge initiation; the results are shown in Fig. 3.

The data shown in Fig. 2 are interpreted in the following manner. For time $t = 0$ h, the initial rise is a result of helium-ion-induced release from the hydrogen-rich surface region and reaches a maximum of 13.2 mTorr with a ^{*n e-folding*} time constant of ~ 8 s. The decrease beginning at about 90 s results from the reimplantation of a portion of the released hydrogen, determined by equilibrium conditions in the glow discharge and surface region; it is 0.9 mTorr and has ^{*an e-folding*} time constant of ~ 70 s. For $t = 1$ h, the mass 2 increase is again the helium-ion-induced release of hydrogen from the hydrogen-depleted, but reimplanted, surface region. This increase is about 1.2 mTorr, in reasonable agreement with the amount of reimplanted hydrogen. The long-term (about 200 s) decrease is about 0.3 mTorr. For $t = 24$ h, a H_2 release of ≈ 0.6 mTorr is observed, with a subsequent decrease due to reimplantation. The difference between the amount of hydrogen reimplanted during the $t = 1$ h run, 0.3 mTorr, and the amount of hydrogen released during the $t = 24$ h run, 0.6 mTorr, could result from the migration of hydrogen from the hydrogen-rich region just below the depleted portion of the surface region or from experimental uncertainty. The interpretation of the data in Fig. 3 is that initial filling of the previously depleted surface region is followed by migration of implanted hydrogen into the graphite bulk along grain surfaces.⁴ The time constant for reimplantation of the released hydrogen was estimated to determine what effect, if any, this might have on the release results. Information obtained from mixed-gas (H_2 and He) glow discharge results⁸ and an unpublished result for low-^{*cross section*} energy detrapping of hydrogen by deuterium of 10^{-18} cm² (Ref. 9) were used to estimate a time constant, ~ 50 s. This is much longer than the release time constant, 8 s, and close to the reimplantation time constant of 70 s. Therefore, it was concluded that the reimplantation of hydrogen via its release into the glow discharge did not significantly affect the results for helium-ion-induced release.

III. DISCUSSION, RESULTS, AND MODEL

The experimental parameters that must be included in any model are an incident helium flux of 4.9×10^{14} He/cm²·s and a total of 6.76×10^{16} hydrogen atoms available for release, as determined from the maximum in the pressure rise for the $t = 0$ h curve in Fig. 2. The helium flux is determined from the glow discharge current, and the charge of the helium ion is assumed to be +1. This amount of hydrogen represents a depletion depth of 0.8 nm, assuming a H/C ratio of 0.5 and a carbon density of 2 g/cm³. Retrapping of detrapped hydrogen can easily be included in the model, but in that case an assumption must be made as to a characteristic length for retrapping, L . Wampler and Myers² have used the McNabb-Foster formalism¹⁰ and determined that a reasonable value for L is 1.5 nm. Since the depletion depth is only 0.8 nm, considerably less than the characteristic retrapping length, retrapping is expected to play only a minor role.

The equation governing the release of H₂ is

$$\frac{dN_m}{dt} = \sigma\phi N_t - \alpha D N_m (N_0 - N_t) - 2k_r N_m^2, \quad (1)$$

where σ is the detrapping cross section; ϕ is the incident ion flux; N_t is the number density of trapped hydrogen ions within the depth where hydrogen can be detrapped, R ; N_m is the number density of mobile hydrogen atoms in R ; N_0 is the total trap concentration in R ; α is the trapping rate coefficient; D is the diffusion coefficient for untrapped hydrogen; and k_r is the recombination coefficient. The first two terms of Eq. (1) are taken from Ref. 2; the third, from Ref. 11. Equation (1) was solved numerically for the initial condition $N_t = N_0$ at $t = 0$. As stated earlier, retrapping was thought to be small, since the depth of the detrapping was considerably less than the characteristic length for retrapping, $R = 0.8$ nm $< L = 1.5$ nm. Checks carried out with values obtained by Wampler and Myers² for the retrapping proved that this assumption was correct. A parametric study was carried out by varying both σ and k_r . The "best fit" to the experimental data was obtained for $\sigma = (1.65 \pm 0.2) \times 10^{-16}$ cm² and $k_r = (0.5-4) \times 10^{-14}$ cm⁴/s. This is shown in Fig. 4. The modeling results are more sensitive to changes in the detrapping cross section than to

corresponding changes in the recombination coefficient, as reflected in the uncertainties of these two parameters.

This experimental technique does present one potential problem. As alluded to in a previous paper,⁴ carbon deposits can gradually build up on the anode and its insulating glass shield. The hydrogen content of these deposits and their influence on the results presented here are not known, but they are thought to play a minor role. A rough extrapolation of the detrapping data of Wampler and Myers² at higher energies (i.e., 3 and 30 keV) predicts a detrapping cross section of $(1-2) \times 10^{-16} \text{ cm}^2$; this brackets the present result of $1.65 \times 10^{-16} \text{ cm}^2$.

In future experiments, we plan to measure the helium-ion-induced desorption at both higher and lower energies and the deuterium-ion-induced desorption of hydrogen.

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Figure Captions

- Fig. 1. Apparatus for measurement of helium-ion-induced release of hydrogen. Explanation of symbols and abbreviations: $\otimes$, valve; $\otimes^*$, variable gas leak; IG, ionization gauge; RGA, residual gas analyzer; SS, stainless steel; HV PS, high-voltage power supply; BAR, sensitive pressure manometer.
- Fig. 2. Time evolution of H_2 signal during helium glow discharge. 0h: result for initial discharge; 1 h: result for second discharge, an hour after initial discharge; 24 h: result for third discharge, 24 hours after initial discharge.
- Fig. 3. Time evolution of H_2 pressure during hydrogen glow discharge following experiments whose results are shown in Fig. 2. See text for explanation of ΔP and dashed line.
- Fig. 4. Fit of model to experimental data shown in Fig. 1 (0h). σ is the detrapping cross section of hydrogen in graphite by helium (in cm^2), k_r is the recombination rate coefficient (in $cm^4/atm \cdot s$).

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RELEASE OF IMPLANTED HYDROGEN FROM GRAPHITE BY 350 eV HELIUM

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