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Hydrogen recycling in graphite at higher fluxes

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Abstract

Understanding hydrogen recycling is essential for particle control in fusion devices with a graphite wall. At Extrap T2 three different models have been used. A zero-dimensional (0D) recycling model reproduces the density behavior in plasma discharges as well as in helium glow discharge. A more sophisticated one-dimensional (1D) model is used along with a simple mixing model to explain the results in isotopic exchange experiments. Due to high fluxes some changes in the models were needed. In the paper, the three models are discussed and the results are compared with experimental data.

Keywords: EXTRAP T2; Flux dependence; Hydrogen trapping in graphite; Isotope exchange; Local molecular recombination; Recycling; RFP

1. Introduction

The particle balance in a fusion experiment is strongly influenced by the uptake and release of hydrogen in the surrounding material. To describe hydrogen recycling in graphite different models have been developed. The models are based mostly on the results of ion beam experiments at low flux densities. Annealing experiments show that particles can be trapped in graphite, the activation energy for trapped particles being $\sim$3 eV and the concentration about 42% of the graphite concentration [1]. Particles can also be solute in graphite with the activation energy $\sim$0.06 eV [2], the concentration of solute sites is not known. Hydrogen can be released from the wall as atoms but faces then a potential barrier of $\sim$1 eV. At temperatures $T \leqslant 500$ K and moderate flux densities the hydrogen release is known to be dominated by recombination into molecules which have a higher diffusion coefficient and can leave the surface through a reduced potential barrier.

A zero-dimensional (0D) model based on these ideas has been applied to the hydrogen recycling in Extrap T2, with flux densities in the range 1–10$\times 10^{22}$ m^{-2} s^{-1} [3]. The 0D model could reproduce the density evolution in RFP-discharges at low densities and to some extent the effect of hydrogen trapped in the wall. However, it could not completely describe the shot to shot increase in density. To estimate the size of the wall reservoir that is interacting with the plasma, isotopic exchange experiments have been used. The 0D model did not reproduce the isotopic exchange behavior. In the present report we compare the 0D model with a simpler isotopic mixing model and with a more sophisticated one-dimensional (1D) model.

2. Experiment

The first wall in the Extrap T2 reversed field pinch (RFP) is completely covered with high density POCO graphite and is symmetric with respect to the plasma. The experiment is operated with the former OHTE vacuum vessel, of dimensions $R = 1.24$ m and $a = 0.18$ m [4]. The operating parameters are; plasma current $I_p \in [100, 270]$ kA, loop voltage $V_{loop} \in [80, 150]$ V, central electron temperature $T_e(0) \in [100, 200]$ eV, line average electron density $n_e \in [0.7, 8] \times 10^{19}$ m^{-3}, pulse length $\Delta t \in [5, 15]$ ms, edge temperature $T(0.18) \approx 15$ eV and edge density $n_e(0.18) \approx 4 \times 10^{17}$ m^{-3}. The experiment is operated at room temperature but is routinely baked up to $\sim$130°C to remove impurities, mainly water and carbon oxides. Glow discharge in helium is used to

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remove hydrogen from the wall [5]. RFP discharges in Extrap T2 extend over about 50 particle confinement times and the average particle flux to the wall during flat top is $0.5 - 4 \times 10^{22}$ m^{-2} s^{-1}. The ion flux is uniform whereas the electron, and hence the heat flux can be localized due to wall locking of internally resonant tearing modes, and to protruding edges.

Extrap T2 is typically operated with a filling pressure of 2–3 mTorr, no extra hydrogen is added during the RFP-discharge and the characteristic pumping time is 400 times longer than the pulse length. Thus, the hydrogen density depends entirely on recycling. The density behavior in an RFP-discharge can be seen in Fig. 1(a) where the peak around 5.5 ms is due to local heating. The line average density increases linearly in a series of discharges following a helium glow discharge [5]. Extrap T2 can be operated at densities down to 0.7×10^{19} m^{-3} by extending the glow discharge time from 40 to 120 min. To avoid early termination of the discharge due to wall pumping, the fill pressure has to be increased. The result from a high fill pressure (8 mTorr) experiment can be seen in Fig. 1(b). High fill pressure in turn entails a rapid buildup of the wall hydrogen reservoir. This effect can be mitigated by working with methane fueling rather than hydrogen.

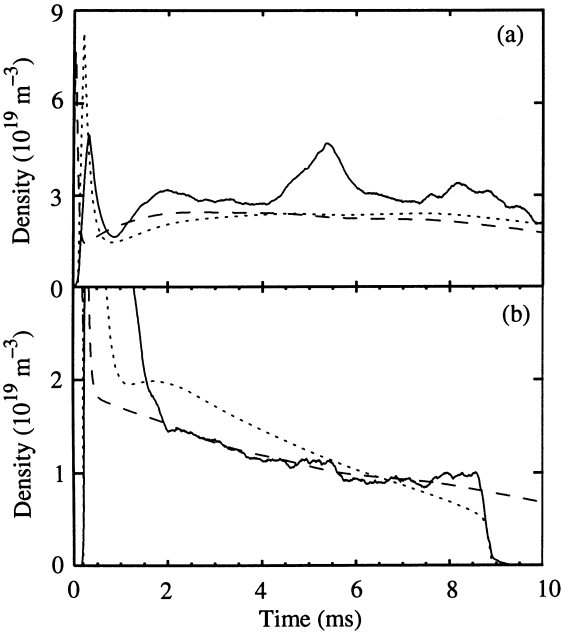

Fig. 1. Line average electron density in RFP discharges, (a) shows a standard discharge with local out-gassing due to local heating at 5.5 ms and (b) results from high fill pressure experiments (8 mTorr). The solid line is experimental data, the dotted line shows results from the 0D model and the dashed line from the 1D model.

In an isotopic exchange experiment deuterium was introduced in the chamber following operations with hydrogen. The D:H ratio was monitored by comparing the D_α and H_α signals measured with a high spectral resolution 1 m Czerny–Turner spectrometer equipped with an optical multichannel analyzer (OMA). By reading only a part of the detector array a time-resolution of 2 ms was obtained. The spectral line profiles were fitted with a Voigt function. The main source of error was the effect of afterglow in the detector, where approximately 10% of the signal in a spectrum is transferred to the consecutive one. A fitted spectrum can be seen in Fig. 2 and the time evolution of the D:H ratio during two discharges can be seen in Fig. 3. The first data point is taken at 1.5 ms, which corresponds to approximately 10 particle confinement times. The evo-

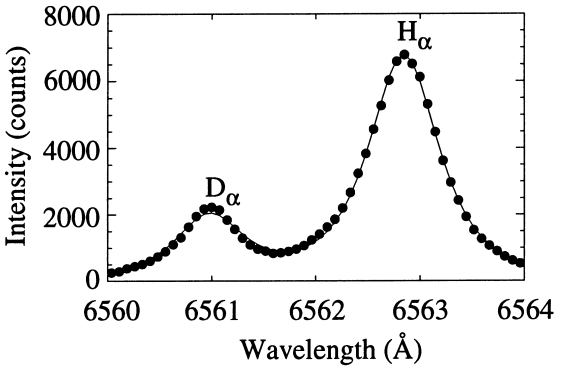

Fig. 2. A fitted spectrum showing D_α at 6561.0 Å and H_α at 6562.8 Å. The data was recorded between 5.4 and 5.6 ms in the eleventh discharge in the isotopic exchange experiment.

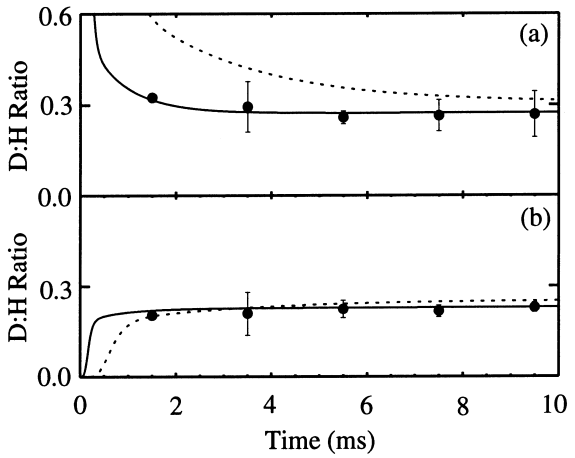

Fig. 3. The isotopic exchange behavior in two discharges, one where the wall is being loaded with deuterium (a) and one when going back to hydrogen (b). The solid line originates from the 1D model and the dotted one from the 0D model.

lution of the D:H ratio in a series of discharges shows a linear increase (Fig. 4(a)). A linear fit to the data gives the line equation $r_{DH}(i) = 0.127 + 0.0215i$ where i is the discharge number. The D:H ratio evolution when changing back to hydrogen fill gas is showed in Fig. 4(b). Even though H_α is a local measurement the ratios obtained show very little scatter. This indicates that the measured ratios represent global conditions.

3. Models

In the first analysis of the isotopic exchange experiment, a simple mixing model (SMM) [5], similar to the one used by McCracken [6] was used. A number of deuterium atoms, N_D are admitted as filling gas in every discharge. The fill gas is assumed to mix perfectly with the particles in a saturated implantation layer, the wall reservoir. The total number of hydrogen in the wall reservoir is $N_W = N_{WD} + N_{WH}$ where N_{WD} and N_{WH} denote the deuterium and hydrogen reservoirs, respectively. Thus, the observed D:H ratio (r_{DH}) at the end of the ith discharge would be

$$r_{DH}(i) = \left(\frac{N_D}{N_W} + 1\right)^i - 1. \quad (1)$$

Using experimental data for r_{DH} and i and Eq. (1) an estimated value of the total wall reservoir can be calculated.

A 1D model, based on the bulk recombination model by Möller and Scherzer [7] with an additional recombination term introduced by Morita and Hasabe [8] has been developed. The model is mass conservative and considers hydrogen in the wall either to be trapped or solute, the particles in the plasma are divided into neutrals and ions. This model is visualized in Fig. 5 and the constants used have been published [3].

Haasz et al. [2] have presented a microscopic treatment of hydrogen recycling in graphite developed from the model by Möller and Scherzer. This model considers graphite composed of graphitic crystallites and diffusion. The hydrogen gets implanted in the grains, where it can be trapped or it diffuses to an internal surface. Haasz et al. calculated the latter transport as a time constant, assuming a size of the crystallites (1–5 nm) and a diffusion constant. The atoms at the internal surfaces have a higher diffusion coefficient and are allowed to recombine into molecules, which are assumed to leave the wall. The 1D model is visualized in Fig. 6.

Calculation made at higher fluxes show that the concentration of atoms solute in the grains increases to an unphysical level. Two different solutions to this problem are suggested. The first one allows the solute

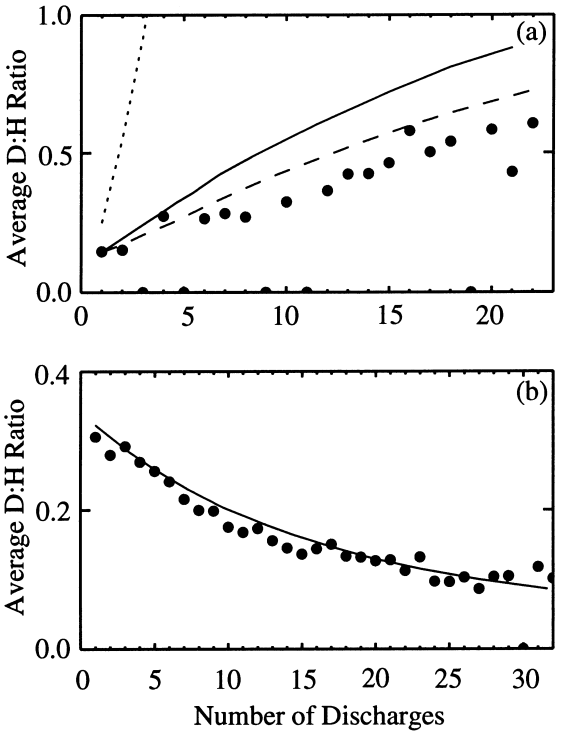

Fig. 4. Evolution of the D:H ratio when loading the wall with deuterium (a) and when going back to hydrogen (b). The dotted line shows the results from the 0D model. The solid and the dashed line originate from the 1D model, the dashed line being corrected for diffusion to the bulk by calculations using Eq. (2).

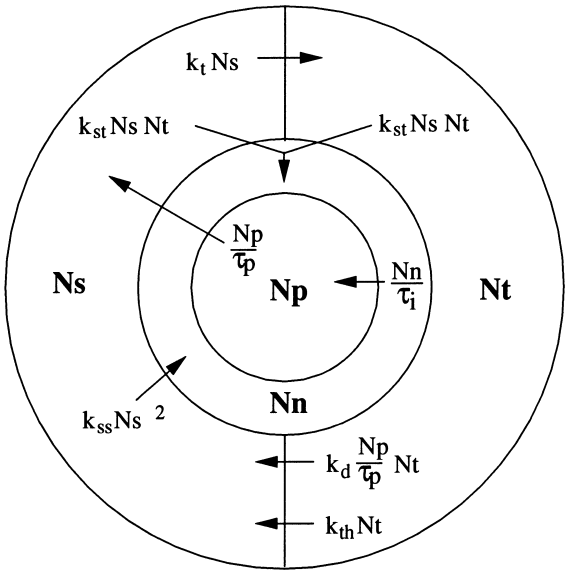

Fig. 5. The 0D model consisting of three different reservoirs. N_p are ions in the plasma, N_n neutrals in the plasma vessel, N_t trapped atoms in the wall and N_s solute atoms in the wall. k_t, k_{st}, k_{ss}, k_d and k_{th} are rate coefficients.

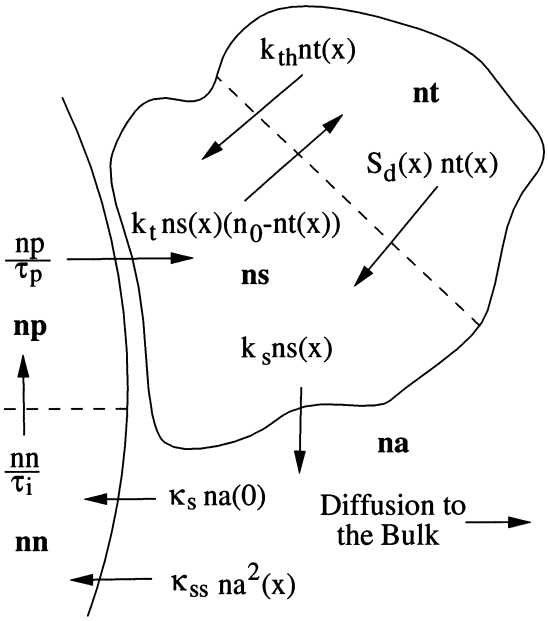

Fig. 6. The 1D model with the concentration of n_t trapped atoms in the grains, n_s solute atoms in the grain, n_a atoms at a internal surface, n_n neutrals in the plasma volume and n_p plasma ions. The rate coefficients are given in Table 1.

atoms in the grains to recombine to molecules. Another way of dealing with the problem is to assume that there is a limited number of solution sites. In the case of high flux the concentration of solute atoms within the grains can exceed the concentration of sites. The resulting decreased activation energy would produce a faster diffusion, the same way as an increased temperature does. We prefer the latter solution as being more physical and in better agreement with experimental data. This possible effect has been included in the model by increasing the diffusion constant by a factor 20 when the concentration of solute atoms gets larger than 10% of the carbon concentration, these values are empirically chosen. The other constants were taken from Haasz [2] and are listed in Table 1. Since the experiment is operated at room temperature, thermal release is negligible.

The original model neglects diffusion of the solute atoms in the direction perpendicular to the surface (the x-direction). This diffusion is slow in comparison with the other processes but might have an effect on the shot to shot evolution. For calculating the input profiles in a discharge the value $[n_0 - n_t(x)]$ was assumed constant and the equation

$$\frac{\partial n_s}{\partial t} = \frac{\partial^2 n_s(x)}{\partial x^2} - [k_s + k_t(n_0 - n_t)]n_s \tag{2}$$

could be solved analytically using a boundary value Fourier method. The same diffusion constant with the same high concentration correction as when calculating the transport out of the grains was used, that is $D(300\ \mathrm{K}) = 2\text{–}40 \times 10^{-18}\ \mathrm{m^2\ s^{-1}}$.

The implantation and ion induced detrapping profiles are obtained by computer simulations using the Monte Carlo code TRIM [9]. The results are fitted to triangular shaped profiles with maximum peak at 60% and 20% of the total implantation depth, respectively (Fig. 7). Since the TRIM program is not validated for energies below 1 keV the profiles have been extrapolated down to the expected 100 eV energy and a maximum implantation depth of 4.0 nm. The plasma parameters going into the 0D and 1D models, the ionization time τ_i and particle confinement time τ_p are calculated using the plasma current I_p. When the plasma current is below 30 kA, τ_i is set to 1000 s, if its above 30 kA, $\tau_i = 25$ μs. The particle confinement time is calculated using the empirical formula $\tau_p = 50 + I_p$ μs where I_p is in kA, consistent with spectroscopic and passive edge probe measurements. The roughness of these assumptions does not make the simulation able to explain transient phenomena at the beginning and end of the discharges but is valid for flat tops.

Table 1
Rate coefficients for the 1D model

Parameter	Prefactor	E_{act}
Bulk transport (k_s)	2.04×10^2 1/s	0.06
Bulk trapping (k_{st})	8.73×10^4 1/s	0.00
Surface diffusion (Δ_s)	6.1×10^{-9} m^2/s	0.06
Recombination (κ_{ss})	3.31×10^{13} 1/s	0.06
Atomic release (κ_s)	1.0×10^{13} 1/s	0.94

4. Results and discussion

Both the 0D and the 1D models reproduce the density within standard RFP-discharges with low fill

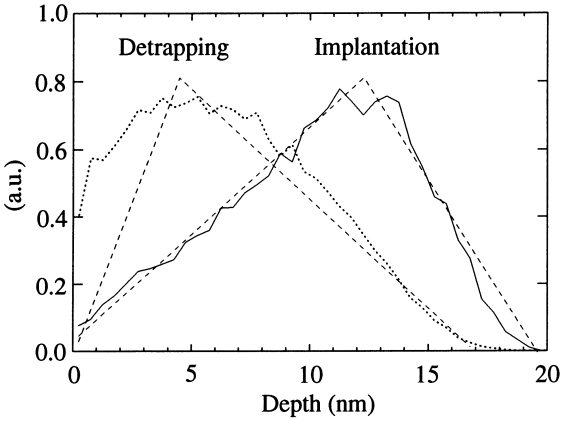

Fig. 7. The profiles calculated with TRIM for 1 keV. The fitted triangles shown as dashed lines are extrapolated down to an implantation depth of 4.0 nm.

pressure (Fig. 1(a)). The parameter being adjusted is the concentration of hydrogen in the wall prior to the shot. The models predict a net increase of trapped hydrogen during a discharge resulting in a higher density in the following discharges. The 0D and 1D models can reproduce the shot to shot density evolution observed in the experiment for the first seven and ten shots, respectively. The models do not predict any accumulation of trapped particles at higher densities and therefore no increasing density in consecutive shots. Selecting the number of trapped particles prior to the shot, constant densities up to 2.5×10^{19} m^{-3} can be explained by the 0D model. For the 1D model this value is 4.5×10^{19} m^{-3}.

For the high fill pressure experiment the 0D model fails whereas the 1D model, apart from the cold, high density phase at the beginning of the discharge, is in fair agreement with the experimental results. To obtain the data for the 0D simulation shown in Fig. 1(b) the pre-shot concentration had to be decreased from the normal range 50–65% of the saturation density down to less than 20%. The 1D simulation was made with a more reasonable pre-shot concentration of 50%, the typical range in these simulations being 55–80%. The difference can be explained by a local loading at a certain depth in the 1D case, an effect the 0D model cannot predict.

The SMM model has been used to analyze the results from the isotopic exchange experiment. The fill pressure in the discharges were 2.9 mTorr corresponding to $N_D = 1.9 \times 10^{20}$ atoms. The D:H ratio in the first discharge $r_{DH} = 0.14$ and Eq. (1) indicates a total wall reservoir $N_W(1) = 1.3 \times 10^{21}$. This reservoir corresponds to the wall being saturated in a 3.5 nm deep surface layer. For the following discharges the second order expansion of N_W, calculated using the line fitted to the experimental data follows the equation

$$N_W(i) = \frac{N_D i}{0.119 + 0.0188i - 2.31 \times 10^{-3} i^2} \quad (3)$$

Thus, the reservoir mixing with the implanted deuterium increases. This can be explained by diffusion to the bulk.

The 1D model reproduced the experimental observation of constant D:H ratio in a RFP discharge (Fig. 3). Using the profiles of particles in the wall at the end of a discharge as input data results in an overestimated ratio in the following discharge, Fig. 4. Calculating starting parameters using Eq. (2) shows that the increase of trapped deuterium within the implantation range should be decreased by approximately 30%. The result obtained with these calculations can be seen as the dashed line in Fig. 4(a). Thus, the experimental data cannot be explained by normal diffusion, there must be other processes not included in the model which account for the mixing. The conclusion is that the high concentration of solute particles due to high flux density results in an increased mixing of particles between the implantation layer and the bulk. When changing back to hydrogen fill gas no additional diffusion has to be used to reproduce the experimental results (Fig. 4(b)). The input parameters used in the calculations were taken from the previous discharges. For the first discharge, profiles are taken from calculations resulting in the experimentally observed ratio for the last discharge in the deuterium series.

5. Conclusions

A 0D model reproduces the density behavior in Extrap T2 discharges at normal fill pressures, the density increase in the first seven shots and hydrogen removal by helium glow discharge. It fails to explain discharges with high fill pressure, densities above 2.5×10^{19} m^{-3} and results from isotopic exchange experiments. The 1D model reproduces hydrogen densities at different fill pressures, shot to shot evolution for ten discharges and to some degree the isotopic exchange. Two possible modifications to explain recycling at higher fluxes have been introduced. A concentration dependence of diffusion in the grains and diffusion of solute particles in the x-direction.

The localized heat load results in a higher temperature at a part of the wall. At higher temperature, thermal detrapping, atomic release, diffusion and recombination rates increase. Calculations assuming that a fraction of the wall is heated to a higher temperature have been made. The models will also be compared with results from other fusion experiments.

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