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Hydrogen-Deuterium Changeover Experiments in a
Plasma-Wall Interaction Simulator***MASTER**

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ABSTRACT

Hydrogen recycling from walls and limiters of tokamaks constitutes the major source of plasma particles after the initial few milliseconds of a plasma pulse. The physical processes involved in recycling are thus very important to the control of plasma density and composition. Isotope changeover experiments in the laboratory show that thermally activated processes such as recombination and diffusion play significant roles in recycling... These processes are important not only during the plasma pulse but also continue after the plasma pulse ends, thus the length of time between pulses, the wall temperature and the vacuum pumping rate all influence changeover dynamics. This paper shows that for type 304 stainless steel walls changeover can range from > 95% to < 50% complete in one 200 ms pulse depending upon these parameters. The initial part of changeover during a plasma pulse is very rapid and involves equilibrating the isotopic compositions of the plasma reservoir and near-surface wall hydrogen reservoir involved in recycling. The rate limiting step then seems to become vacuum pumpout rates. Because one of the steps in gas evolution during and after the pulse is the recombination process, which is dependent on the square of the surface hydrogenic concentration, changeover can be more rapid if the hydrogenic concentration at the surface is kept high by repeated plasma pulses in the new isotope. The old isotope is evolved as HD.

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1. Introduction

Hydrogen recycled from walls and limiters of tokamaks, stellarators and EBTs constitutes the major source of particles to the plasma,¹ hence control of plasma density and composition depend on recycling. The inventory and permeation of tritium in the walls of future machines is also very much controlled by the details of hydrogen recycle. (Some of the plasma particles do not return to the plasma.)^{2,3} The presence of large amounts of hydrogen in the walls of future machines could also alter the mechanical properties of the walls and their resistance to radiation damage and thermal cycling. These are all important reasons to understand the recycling mechanisms.

Isotope changeover experiments have been used in several machines⁴⁻⁶ in attempts to learn about recycling in actual fusion experimental devices. However, the details of the recycling processes are very complex and not easily studied in tokamaks or other fusion experiments. The basic elements of recycling processes can be specified⁷ and models have been developed to explain the results of machine behavior and to predict the behavior of future devices.^{1,3,8} Laboratory experiments however are required to provide data for these models and to test the predictions in relatively easily analyzed situations. Laboratory studies using the isotopes of hydrogen provide a way to attack some of the questions of system dynamics and inventory.

2. Purpose and General Considerations

The experiments described here were initiated to show the effects of some of the controllable variables which can influence hydrogen recycle, and more specifically isotope changeover times in tokamaks.

It has been shown that for 300 series stainless steels near room temperature, diffusion and recombination processes are very important in recycling.^{9,10} Recombination of dissolved atomic hydrogen into molecules which are desorbed back into the plasma may often play a dominant role. This step is described by

$$\phi = K_r c_o^2$$

where ϕ is the rate of production of molecules, K_r is the recombination coefficient and c_o is the surface concentration of hydrogen. When hydrogen isotopes H and D are considered, molecules of H_2 , HD and D_2 will be evolved and

$$c_o^2 = [C_H^2 + 2C_H C_D + C_D^2] \quad .$$

Since recombination and diffusion are both thermally activated processes and continue during the interval between plasma pulses, the length of the interval, the temperature and the vacuum pumping characteristics of the system should influence recycling and isotope changeover times. (Surface composition, flux density and temperature have previously been shown to be important parameters.⁹) In isotope changeover experiments the gas input to the plasma chamber is switched from one isotope to the other and the rate at which the plasma changes to the new isotope is measured or a minority isotope is rapidly puffed into an established plasma of the other isotope and the resulting isotopic composition changes measured. Both experiments are useful in studying hydrogen recycling.

Two experiments are described below. In the first, the simulator was operated with short pulses of plasma (200 ms), simulating ISX-B tokamak discharges, first in one isotope, hydrogen, and then between shots (without interrupting the timing of pulses) the fill gas was switched to the new isotope, deuterium.

The discharge in the new isotope was then monitored to determine how much of the old isotope was present as a function of time and other variables. Sets of changeover data were made by changing one variable for each data set. In the second experiment hydrogen was puffed into a deuterium steady-state discharge to determine how rapidly the plasma and wall compositions mix and change. Data from this experiment can be used to estimate the inventory of hydrogen isotopes in the plasma and the near-surface region of the wall and to observe the dynamics of isotope changeover for long pulse devices.

3. Experimental Apparatus and Choice of Operating Conditions

A glow discharge plasma device, developed to simulate the most important aspects of the hydrogen recycle processes, has been previously described.¹¹ An extended choice of the parameters affecting recycle is available including especially wall temperature, particle fluxes and energy, plasma pulse duration and intervals between pulses. In addition wall materials can be changed and surface composition monitored with the aid of surface diagnostics.*

In the simulator a glow discharge is used to supply ions for simulating the flux of energetic plasma particles to the walls of a fusion device. Ions are accelerated through the sheath between the glow and the wall by an electric potential from an external power supply so that walls are uniformly bombarded with ions of known but selectable energy (100 eV for this study). The ion species include H^+ , H_2^+ and H_3^+ and similar species

*The surface composition plays a key role in recycling and its control and modification must be included in any serious attempt to study the details of the recycling processes. For example, the effect of small amounts of oxygen contamination on wall hydrogen content and recycling dynamics is dramatic.¹¹

containing part or all deuterium. At the relatively high pressures used, 3.3 Pa (25.0 mtorr) the ratios are approximately $H^+:H_2^+:H_3^+:1:3:10$. These ratios are a function of pressure and change as the simulator pressure changes, but the ion species can be measured with a quadrupole mass analyzer and the ion energy distribution determined with retarding grids.

The monitoring and control of glow discharge and wall current parameters together with vacuum pumping, gas input and test chamber pressure has been the primary means of determining wall influx and outflux rates and thus following particles during the recycle process.^{9,10} An optical spectroscopy system has now been added to monitor the H_β and D_β optical emission lines from excited neutral hydrogen isotopes and thus continuously measure the isotopic composition of the plasma. The time constants for the optical measurements are about 5 ms. Results of all measurements are recorded by a digital data acquisition system at 1000 Hz.

For this study we used a surface with about 15% surface oxygen level,* a hydrogen flux density of $\approx 6 \times 10^{16} \text{ H/cm}^2 \text{ s}^{-1}$ and an average ion energy of 100 eV/molecule. These conditions approximate conditions expected at the wall near the limiter or gas puff inlet in the ISX-B tokamak. Pulse lengths were 200 ms except where noted and times between pulses were 1 or 10 min except when DC plasmas were used to study changeover during gas puffing. The normal vacuum system time constant (volume $\times$ pumping speed⁻¹) was 1 s. A 40 s vacuum system time constant was used to show how reduced pumping slows changeover.

*Based on the ratio of Auger electron line intensities and sensitivities without correction for depth distribution using the method described in ref. [12].

The time constant, τ_p , for hydrogen in the simulator to hit the wall during a plasma pulse, i.e.,

$$\tau_p = \frac{n_H \text{ in the plasma phase}}{(n_H \text{ hitting the wall per second as ions})}$$

is about 10 ms. In ISX-B the plasma particle containment time is ≤ 20 ms in the early part of a discharge. Thus changeover in the simulator should be on a time scale similar to that in ISX-B for otherwise comparable conditions. (But of course wall fluxes and other conditions are not uniform and often not measurable in ISX-B.)

4. Results and Discussion

4.1 Change of Fill Gas

Figure 1 shows the D_β and H_β signals, the pressure and the current to the wall during the first 200 ms discharge in deuterium following previous discharges at 1 min intervals in hydrogen. The ratio of H_β and D_β line intensities give the ratio of H and D concentrations in the plasma. Individual isotope concentrations are available from RGA data or by multiplying the fractional composition, from H_β and D_β signals, by the total pressure. Figure 2 shows the hydrogen isotope concentration during a 0.8 s plasma pulse identical to Fig. 1 conditions except for pulse length. Note the very rapid increase in hydrogen isotope concentration as hydrogen is evolved from the simulator wall during the plasma pulse and the relatively slow decrease. The time constants for the rise (due to equilibration of the isotopic ratios of the plasma and wall inventory involved in recycle) and fall (due to removal of gas by the pump) are of the order of 60 ms and several seconds, respectively.

Figure 3 shows the isotopic composition of the gas in the discharge at the end of each shot for several series of shots during which the fill gas was changed. The reference case shows the changeover for 1 min pulse intervals; the vacuum system time constant, $\tau_v = 1$ s; the temperature $T = 25^\circ\text{C}$. Changeover is about 65% complete, i.e., 35% hydrogen at the end of the first pulse and 92% complete (8% hydrogen) at the end of the third shot for the conditions listed above and in Section 3. In contrast the first pulse was still 62% hydrogen at the end of the first shot when the $\tau_v = 40$ s and ~38% after the seventh pulse. If the time between pulses is changed to 10 min from the standard condition, the first discharge ends with only 17% hydrogen and the second with only 5% hydrogen.

A still more complete changeover is obtained at 300°C where the first pulse in deuterium at 10 min after 100% hydrogen pulse produced about 2% deuterium (which is near the limit of detection in these particular measurements). These data serve to illustrate the significance of temperature and time between plasma pulses and the effect of vacuum system pumping on changeover. The values given above would show somewhat more complete changeover if the pressure in the discharge chamber were kept constant by adding the new isotope during the discharge, but this would not alter the conclusions.

If ion induced processes were the only important processes in producing changeover, there would be little effect of changing the pulse interval, wall temperature or vacuum system time constants through the range of the values used here. Loading and release from the wall would occur only during the relatively short plasma pulse (independent of temperature) and the pumpout between pulses would remove most of the released gas. (Even the 40 s vacuum time constant would allow the removal of 78% of the mixture during the interval between pulses at 1 min intervals.)

Thermally activated processes (diffusion and recombination) however continue both during and between plasma pulses and thus time between pulses, temperature and pumpout rates play a significant role as the wall inventory decreases and is pumped away between pulses.

The minority species is present almost entirely as HD during and after the plasma pulse as one expects if surface recombination is occurring rapidly, as it is here.

It should be noted that per unit time (e.g., 10 min) changeover is faster in every case when plasma pulses are more frequent. This may be, in part, due to ion bombardment effects but is also to be expected simply as the result of keeping the surface hydrogenic concentration higher and thus increasing the recombination of atoms to HD molecules and allowing them to be desorbed and pumped away during and between pulses.

It should also be noted that not all of the data sets show faster changeover at 300°C. The cause of this behavior is not completely obvious but will be investigated using a modeling technique.¹

4.2 Puffing Hydrogen into a Deuterium Discharge

Figure 4 shows the hydrogen isotope concentration in the plasma (based on H_β measurements) resulting from a fast puff of hydrogen into a deuterium discharge which has been running for several minutes with simulator walls at ~25°C. The recycle coefficient for deuterium is very near unity at the time of the puff. Note that the initial hydrogen concentration decays very rapidly until enough of the hydrogen from the plasma volume is loaded into the wall to provide nearly 100% recycling. This means that the isotopic composition of the recycling hydrogenic species is nearly the same as that of the plasma and to a first

approximation the near surface isotopic composition has come into equilibrium with the plasma composition. This results in about a factor of 2 drop in hydrogen isotope concentration in the plasma. We conclude that the wall reservoir involved in rapid recycling is at least as large as that in the plasma. The initial hydrogen loading is rapid followed by a slow change as the mixture is exhausted to the pumps and new deuterium fill gas enters. The time constants here are less than 50 ms for the loading and several seconds for the exhaust. These values are consistent with data from Fig. 2 for the exhaust phase and shorter for the loading phase. This is to be expected since the evolution of hydrogen in the last case is from a surface with a high hydrogenic concentration, already recycling 100% of the hydrogenic flux before the puff. On the other hand a few tens of milliseconds are required for the surface to reach hydrogenic concentrations high enough to provide recycling approaching 100% in the case shown in Fig. 2.

5. Conclusions

1. During the plasma pulse two separate and distinct changeover rates are apparent: (a) a short time for the plasma inventory to approach isotopic equilibrium (or "mix") with the near-surface wall inventory and (b) a longer time for the vacuum system to remove the resulting gas mixture.
2. The difference in times to achieve "mixing" for short pulses and for gas puff experiments is attributed to the time required to build up the near-surface hydrogenic concentration in the case of the short pulse.
3. Continuing evolution and removal of both isotopes after each pulse is an important part of the changeover process for today's machines.

4. The rate of removal of the minority isotopic species during a changeover sequence of plasma pulses appears to be enhanced by the presence of high concentrations of the majority species. This continues during and between plasma pulses producing mostly HD molecules which are quite evident in the effluent gases. This suggests that plasma pulses in deuterium before and after tritium pulses would help reduce the inventory of tritium in experimental fusion devices and that injected tritium would recycle from the wall to the plasma much more rapidly if the walls are preloaded with deuterium. (However much more deuterium will also be evolved into the plasma from the wall.) Glow discharges might be used to speed the removal of tritium from the walls of experimental devices and thus reduce the tritium inventory.

It should be cautioned that in a tokamak, wall fluxes, compositions, and temperatures vary considerably from one part of the machine to another and that the net changeover will be a convolution of many regions and conditions.

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FIGURE CAPTIONS

Fig. 1. Typical data for the first 200 ms plasma pulse using deuterium fill gas following previous pulses in hydrogen. This data was obtained as part of a pulse sequence with 1 min between pulses, $\tau_v = 1$ s, temperature = 25°C. Initial deuterium pressure 3.3 Pa (25 m torr) and the ion current to the wall ≈ 1.5 A.

Fig. 2. Hydrogen isotope concentration ($\frac{H}{D} \times \text{total pressure}$) in the discharge chamber as a function of time during a 0.8 s plasma pulse. Other conditions are identical to those of Fig. 1.

Fig. 3. Percent hydrogen in the plasma at the end of each pulse during changeover sequences with parameters changed as indicated. The curve labeled 1 M-7S-300 was obtained with 1 min intervals between pulses, a vacuum system pumping time constant $\tau_v = 1$ s and wall temperature of 300°C. Other curves are labeled in a similar manner.

Fig. 4. Total pressure and hydrogen isotope concentration in a continuing deuterium discharge as a function of time. Hydrogen was puffed into the discharge at 0.1 s.

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Fig. 3. Percent hydrogen in the plasma at the end of each pulse during changeover sequences with parameters changed as indicated. The curve labeled 1 M-1S-300 was obtained with 1 min intervals between pulses, a vacuum system pumping time constant $\tau_v = 1$ s and wall temperature of 300°C. Other curves are labeled in a similar manner.

Fig. 4. Total pressure and hydrogen isotope concentration in a continuing deuterium discharge as a function of time. Hydrogen was puffed into the discharge at 0.1 s.