Hydrogenic Fuel Recovery and Retention with Metallic Plasma-Facing Walls in the Alcator C-Mod Tokamak.

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Abstract. The retention and recovery of hydrogen (H) and deuterium (D) fuel in the Alcator C-Mod tokamak are studied. C-Mod solely employs high-Z molybdenum (Mo) as its plasma-facing armor material, with intermittent application of thin boron (B) films. The C-Mod wall materials are found to retain large fractions, ~20-40%, of the D$_2$ gas fuelled per shot, regardless if the Mo surfaces are bare or partially covered by B films. The retention appears to be linearly proportional to ion flux to the wall, with ~0.5% D ions incident to the wall retained, and with no indication of the retention rate saturating over 30 s of plasma exposure. The retention is inconsistent with D in Mo retention from laboratory ion beam studies, which would predict rapid saturation of the fuel inventory in the wall a few discharges. Neither is the retention consistent with co-deposition of the B since it occurs with cleaned Mo walls. Overall these results raise concerns for use of high-Z materials in long-pulse D-T devices like ITER. Planned disruptions are exploited to recover H and D from the wall in C-Mod. The disruptions produce rapid heating of the wall surfaces, releasing the H/D from the wall for recovery by the pumps; a technique proposed for tritium recovery in ITER. It is shown that disruptions are effective for removing hydrogen absorbed into the wall during water exposure during vents. Overall control of the H/D wall inventory is demonstrated using frequent, planned disruption, with the result that the C-Mod wall can be net depleted of fuel over a run day.

1. Introduction

Controlling the hydrogenic fuel inventory in plasma facing materials will be necessary for the successful operation of burning plasmas that use tritium (T) as a fuel. In ITER, the on-site limit of tritium is 350 g for safety reasons, while ~100 g is fuelled into the vessel for a full power discharge [1], indicating the requirement for low global retention rates. The amount of carbon plasma-facing components (PFCs) in the present ITER design has been minimized at least in part due to the capability of plasma-deposited C films to store large amounts of T (T/D ~0.1). Tungsten is also proposed for use as PFCs in ITER and reactors due to its expected lower T retention and advantageous nuclear properties. However, tokamak divertor experience with regards to hydrogenic fuel retention in high-Z materials is limited. Alcator C-Mod has only high-Z molybdenum as the PFC material with intermittent thin boron (B) films applied (boronization). A study documented the negative response of plasma performance to the removal of the B film from the Mo PFC [2]. The boron removal further provided a valuable opportunity to study H/D retention in Mo, which is reported in Section 3.

In addition to understanding retention, it is desirable to develop in-situ methods of recovering hydrogenic fuel from PFC materials. Methods will be needed to remove T accumulating in devices like ITER, in order to minimize delays in plasma operations. An in-situ tritium recovery method has been proposed for ITER that uses planned radiative terminations [3]. The resulting rapid radiative heating of plasma-viewing surfaces causes the D/T to be released as molecules to be recovered by the vacuum pumps, while the underlying substrate material is undamaged. We describe successful recovery of H/D from the C-Mod wall using planned disruptions in Section 4, demonstrating global control of the fuel inventory in the wall.

2. Experiments
The particle balance of Alcator C-Mod is accurately determined by closing all valves to vacuum pumps for the duration of the shot. Gas is injected in known quantities through calibrated valves (these calibrations have been cross-checked by injection into an empty vessel). The pump valves remain closed for 5-10 minutes after the discharge in order to measure the equilibrium vessel pressure and hence recovered fuel particle inventory, which is then evacuated by opening the pump valves. The particle balance inventory is very accurate (+/- 3x10^19 D) because for these experiments C-Mod has no other external fuel sources (e.g. neutral beams) nor any other active particle sinks (e.g. cryopumps) and discharge cleaning (e.g. electron cyclotron discharge cleaning, ECDC) is not used between discharges. Therefore, any deficiency in recovered D as compared to the injected D must reside in the PFC surfaces, while an excess of D recovered is a result of net outgassing or depletion of the wall. In-vessel gas composition is measured with a quadrupole mass spectrometer. All discharges used here are diverted and fuelled with D_2 gas injection only. Auxiliary heating utilized H-minority ion cyclotron radio frequency (ICRF) waves, although many discharges use ohmic heating only. Typical plasma parameters used are: major/minor radius R/a=0.67/0.22 m, plasma current I_p=0.8-1 MA, toroidal field B=5.3 T, shot duration 1-2 s. C-Mod has intermittently applied boronization films since ca. 1996. For these present experiments, the boron films were removed from all interior vessel PFC surfaces using wipes or ultrasonic cleaning [2]. Surface analysis of the cleaned tiles showed ~80-90% pure Mo surface with the remainder being made up of boron. This is in stark contrast to the situation before cleaning, where the surface Mo coverage was < 1 %, and the B film thickness 6-10 microns thick over most of the wall, i.e. boron was by far the dominant element facing the plasma. A noticeable exception to the boron coverage was the outer divertor, which was found to have substantial Mo surface exposure (~50%), caused by net erosion of the boron films at that location during plasma operations.

3. Deuterium fuel retention

Experiments were carried out to compare the D retention of cleaned Mo walls (“pre-boronization”) to the retention of the wall after the application of boron films (“post-boronization”). The D retained per shot shows several interesting features (Fig.1). First the level of retention is relatively large as compared to the typical C-Mod fuelling level ~ 2x10^{20} D/shot, indicating a high global retention rate. Secondly, and most surprising, there is no significant difference in the D retention between the boronized and cleaned walls. This indicates that the low-Z boron films are not directly causing the D retention, despite the ability of boronization films to retain large atomic fractions of H/B~0.4 through co-deposition (similar to carbon.
films) and that B is the dominant plasma facing element. Thirdly, there is a no strong dependence of retention with confinement mode or ICRF heating. Lastly, the retention is dependent on plasma line-averaged density, exhibiting a rather sharp threshold at $n \geq 10^{20} \text{ m}^{-3}$ towards higher retention rates. This suggests a link between retention and ion/recycling flux to the wall which depends strongly on density $-n^{2-3}$. We now examine more carefully the nature of the D retention with bare/cleaned Mo surfaces.

Fig. 2 shows the dependence of plasma duration on the plasma flux to the wall and the D retention. The divertor and main-wall ion fluxes are of comparable magnitude (D+/shot) and both scale approximately linearly with total plasma duration. The D retention per shot also increases monotonically with plasma duration indicating that total D$^+$ fluence to the wall is important for retention. This first indicates that transient effects, for example during plasma startup or rampdown, do not dominate the D retention, although may be playing a role in setting the linear offset seen in Fig. 2b. Secondly, both the main-wall and divertor can be considered candidates for the location of the D retention since they are similar magnitude sinks for the D ions and recycling. We point out that the data of Fig. 2 were taken in sequential shots. The monotonic increase in D retention suggests little or no saturation of the retention capability of the PFC materials.

The repeatability of retention rates throughout a run day and the effect of the strikepoint position is shown in Fig. 3. This run was carried out post-boronization and in addition, there was measurable titanium dust in the vessel resulting from previous plasma interactions with the lower-hybrid launcher [2]. Special attention was paid to avoiding disruptions, and the associated transient heating of PFC surfaces, in the current rampdown phase of these discharges. In fact other particle...
balance experiments, with no special avoidance of rampdown disruptions, showed approximately zero net D retention integrated over a run day due to the D recovery induced by the naturally-occurring disruptions (see Section 4 for more details). Additionally, based on Fig. 1, the line–averaged density was kept constant from shot-to-shot in these L-mode discharges. The data was acquired after 15 discharges had been carried out for another experiment. In the standard LSN configuration, i.e. with the outer strikepoint on the divertor vertical target, one immediately finds on shot 16 the D fuel retention (∼7×10²⁰ D/shot) expected from the other experiments at nₑ ∼ 1.6×10²⁰ m⁻³ (Fig. 1). This verifies that the boron, and in this case the extra titanium dust in the vessel, are not playing a dominant role in the retention. One also finds that the retention is a large fraction, ∼50%, of the fuelled D (15×10²⁰ D/shot). Subsequent repeat discharges find the same level of retention with no change of wall saturation, although the fuelling requirements evolve somewhat. There is an immediate drop in the D retention per shot, as well as the required D fuelling, when the outer strikepoint is switched to the horizontal floor of the divertor. Indeed the wall is nearly a zero net sink/source of D for the first two discharges. However the D retention reaches a positive and constant value, ∼25% of fuelled D, after three shots. Subsequent shots with strikepoint at the vertical target plate, and upper divertor discharges, show repetitive high D retention per shot. This suggests a complex interplay between the plasma transport, wall fuelling requirements and the history of the particle fluence and retention in the wall. These results further suggest that the outer divertor plays a significant role in the D retention since plasma conditions to other locations did not vary greatly with the LSN strikepoint position change. This also implies that Mo surfaces are important in the D retention, since the outer divertor tends to be a location where the boron films are removed by plasma net erosion. This is at least consistent with the unchanging D retention with boronizations and titanium dust. However while the retention rate can evolve if plasma conditions suddenly vary, the D retention quickly equilibrates (in a few seconds of discharge) to a steady value that shows no signs of diminishing with time, i.e. the wall inventory of D does not appear to saturate on these timescales.

The linearity of the D retention for a cleaned Mo wall (after the removal of titanium dust) was further examined in a

Fig. 4 Series of repeated shots with nₑ =1.6×10²⁰ m⁻³, plasma duration 2 s, cleaned Mo walls. (a) Total D ion flux to the wall and D fuelled per shot (b) D retention per incident D ion to the wall and per fuelled D (c) D retained per shot: C-Mod experiments compared to model calculations based on D retention in Mo from ion beam data [4].
series of nine repeat discharges (Fig. 4) in which the line-averaged density and plasma shape were kept constant, again avoiding disruptions in the current rampdown phase. The plasma current was decreased from 1 MA to 0.8 MA at shot #7. Fig. 4a shows that the fuelling requirements and total ion flux to the wall (Fig. 2) remained constant over these discharges. It is important to note that the D ion flux to the wall is approximately 50 times larger than the fuelling rate, a natural consequence of strong particle recycling in the plasma edge. Fig. 4b shows the D wall retention normalized to both the D ion flux and D fuelling. While the retention per shot evolves over the course of the first four shots (~8 s plasma exposure), the retention equilibrates to a constant value of ~28% per fuelled D atom and ~0.6% per incident D ion to the wall. Indeed Fig. 5 shows that over the course of 16 repeated discharges (~30 s plasma exposure) that avoid disruptions, the retained D fluence remains linear with incident D ion to the wall at an average rate of 0.75%.

Fig 4c compares the measured retention D retention per shot with those calculated from a model based on ion beam measurement of 1 keV D ion retention in room temperature Mo [4]. The ion beam data showed that the retained fluence ($\Phi_{\text{Retained}}$ in D/m$^2$) did not saturate, but the retention increased weaker than linearly with incident D ion fluence ($\Phi_{\text{D+}}$ in D/m$^2$). Specifically, the retained fluence was found to be given by $\Phi_{\text{Retained}} = 3 \times 10^{20} \left( \Phi_{\text{D+}}/10^{24} \right)^{0.345}$. We have assumed reasonable values of the wetted areas for the divertor (~0.5 m$^2$) and main-wall (5 m$^2$) in order to calculate incident average D ion fluence based total D ion flux (Fig. 2), and hence predict total retention based on the ion beam results. We have maximized the possible D retention values by assuming that the Mo is “empty” of D at the beginning of the shot sequence. It is clear from Fig. 4c that this model does not replicate the C-Mod data. Based on the ion beam data the ability of the Mo to retain D should quickly approach effective saturation within at most 3-4 shots, after which the maximum retention per shot would fall below our detection limit of $3 \times 10^{19}$. In contrast, recent laboratory in-situ D retention experiments, using D plasma exposure on Mo at 400K, have shown linear retention rate with D ion fluence comparable to those of C-Mod [5]. This suggests that some aspect of the plasma exposure (high flux density, transient heating, etc.) different than the ion beam exposures is the cause of the higher-than-expected D retention in C-Mod. This remains a subject of investigation for the laboratory D retention experiments such as DIONISOS [5].

4. Hydrogenic fuel recovery using planned disruptions

It has been proposed to use planned radiative terminations, initiated by massive impurity injection, as an in-situ technique to recover retained tritium from the plasma-facing surface of ITER [3]. This recovery technique relies on rapid (~1 ms) transient heating of all plasma-viewing surfaces, caused by radiative dissipation of the plasma thermal energy. The elevated
material temperatures promote de-trapping, recombination into molecules, and diffusion of the D/T out of the material. If properly controlled the heating can lead to substantial D/T release in molecular form from the surfaces without damage to the substrate materials, with the released gases removed by the intrinsic pumping system, therefore not requiring any special vacuum system intervention. Calculations based on laboratory data show that the routine radiative termination of ITER discharges in the rampdown phase (when I<7 MA) could lead to substantially lower or controlled tritium inventories [3]. While it is desirable to test this technique in present tokamaks, however radiative termination cannot typically deliver sufficient energy density to the wall in present devices like C-Mod to promote H/D release. This is due to the much smaller energy density (thermal energy normalized to surface area, Wth/Awall) of present devices as compared to ITER. Therefore we have utilized planned disruptions (i.e. without impurity injection) in order to provide localized surface heating in an attempt to promote H/D release from the C-Mod walls. This technique expands on disruptive discharge cleaning used in TFTR for limiter conditioning [6].

There are two objectives to the planned disruption tests carried out on C-Mod: 1) Reduction of H/D isotope ratio in the plasma and 2) Recovery of the trapped D from the wall caused by the high linear retention rate (Section 3). With regards to 1), hydrogen is present in the wall surfaces after vents due to exposure to water-laden air. The post-vent hydrogen isotope ratio, H/(H+D), must be reduced to ~ 0.05 in order to maximize ICRH H-minority heating efficiency. This typically requires several days of ohmic D plasma operation. Surface analysis of post-vent Mo tiles measured high H atomic fractions (~0.2 –0.3) distributed over a depth ~ 0.5x10^-6 m at the surface. This implies a starting near-surface H inventory ~ 5-10 x 10^{22} in the entire C-Mod wall, whose removal could be expedited using planned disruptions. Simultaneously one would expect recovery of retained D from the wall since the temperature-induced desorption applies equally to all hydrogenic isotopes.

A series of planned disruptions were applied on C-Mod, carried out soon after a vent, and with cleaned Mo walls. Two kinds of planned disruption were used: 1) Vertical Displacement Event (VDE) disruptions where the plasma is intentionally made unstable to vertical movement, resulting in the core plasma limiting into portions of the wall and 2) q=2 disruptions produced by ramping down the toroidal field at constant plasma current. Both disruption types tend to produce localized heating at the wall, rather than the uniform distribution of plasma energy through radiation. As in the D retention measurements, pumping valves were closed for these discharges in order to obtain accurate gas balance, and residual gas analysis was used to distinguish between recovery of H₂, D₂ or HD.

![Fig. 6 Dependence of H recovery on target plasma conditions before the plasma disruption: (left) Central plasma temperature (right) stored thermal energy. Disruption types include VDE and q=2.](image-url)
The physical picture of the disruption-induced H/D recovery described above was generally validated by the experiments. Fig. 6 shows the level of recovered H as a function of the core plasma parameters immediately preceding the disruption. There exists a clear threshold in central T and stored energy after which H recovery becomes very efficient per disruption. Note that only D is fuelled during plasma operations and the typical D retention values are < 10^{21}. Such an energy threshold is expected from model calculations [3], due to the exponential dependence of hydrogenic recombination and diffusion with surface temperature. In other words, a critical surface temperature must be obtained to promote H diffusion and release on the ~ ms disruption timescale, and this requires a minimum amount of plasma energy. Interestingly, the H recovery threshold is better organized to core T than stored energy, indicating that plasma heat conductivity (∝ T^{3/2}) is important in the surface heating. Also, it was found that the H/D ratio in the recovered gas closely tracked the H/D ratio in the target plasma, indicating that the disruption heating was recovering fuel from the same surface inventory available for plasma fuelling from the wall.

The practical H recovery rate was found to be enhanced using the planned disruptions. Fig. 7 shows the H removal efficiency compared between overnight ECDC cleaning rate, regular D fuelled ohmic discharges and planned disruptions. The H removal efficiency is defined as the number of H atoms recovered from the wall per 15 minutes of “wall-clock” time, i.e. the H recover in a C-Mod shot-cycle. The planned disruptions were 5-10x more efficient for the H global recovery rate than the standard techniques. Note that this improvement could be made even larger given higher stored energy target plasmas (Fig. 6).

Fig. 7 H recovery efficiency of different methods as measured by H atoms recovered per shot-cycle. One shot-cycle is taken as 15 minutes of ‘wall-clock’ time for C-Mod.

![Fig. 7](image)

Fig. 8 Cumulative effect of planned disruption H/D recovery over a C-Mod run day. H and D recovered, and the D fuelled/injected are compared. Planned disruptions produce the stepwise increases in H/D recovery.

![Fig. 8](image)
Fig. 8 displays the time-integrated effect of using planned disruptions over the course of a C-Mod run day. The amount of H removed during the day is ~ 1/3 of the estimated starting H inventory (~5-10 x10^{22}) in the near-surface (< micron) of the Mo tiles H/(H+D) ratio in the plasmas was reduced from 0.5 to 0.3 during the day, also consistent with recovering ~ 1/3 of the H wall inventory, indicating that the planned disruptions can be a useful in-situ tool for controlling H isotope fractions in the wall. A substantial fraction of the H was recovered in HD molecules, indicating that isotope exchange is playing an important role.

Besides the recovery of just H, Fig. 8 also demonstrates that planned disruptions removed substantial amounts of D from the wall. Indeed, the planned disruptions resulted in net global fuel depletion of the wall, i.e. the opposite of the strong D retention increasing linearly with time for quiescent discharges shown in Section 3. This is an important demonstration of active fuel control in a confinement experiment, without the requirement for intervention into the vessel.

5. Discussion and Conclusions.

The strong global retention of D in Mo PFC surfaces seen on C-Mod clearly raises concerns for long-pulse devices like ITER. It should be noted that C-Mod closely matches ITER for edge plasma parameters of heat and particle flux densities. If the measured fuel retention rate (Figs. 4,5) were directly extrapolated to ITER, it would result in ~25 g of tritium trapped per full D-T ITER discharge (25% of T fuelled per shot), requiring shutdown and active T recovery after only 12 shots. Most worrying and puzzling is the apparent inability to obtain saturation of D in the wall. Given the absence of codeposition with a Mo-dominated wall, this suggests a combination of D permeation and dynamic trap production as the cause of the retention. Further laboratory studies are being carried out to better understand this process [5], so that one may reliably extrapolate the C-Mod experience over a cumulative ~30 s pulse to the case of ITER with a 500 s pulse.

On the positive side, planned disruptions appear to be a useful tool for controlling fuel inventory in a confinement device and the underlying physical principles of the technique have been confirmed. In addition, “naturally” occurring disruptions, for example in the current rampdown, tend to keep the long-term D wall inventory low on C-Mod. An outstanding question from the C-Mod experiments is if the location from which the D is recovered is coincident with the location of D retention. If not, this would eventually limit the effectiveness of the disruption H/D removal. Fortunately, the technique scales favorably to large devices like ITER, which have much large stored energy density, and would therefore allow H/D/T removal from even larger portions of the wall area.

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References.
[1] ITER Physics Basis