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# Hydrogenic retention with high-Z plasma facing surfaces in Alcator C-Mod

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Abstract. The retention of deuterium (D) fuel in the Alcator C-Mod tokamak is studied using a new 'static' gas balance method. C-Mod solely employs high-Z molybdenum (Mo) and tungsten (W) for its plasma-facing materials, with intermittent application of thin boron (B) films. The primarily Mo surfaces are found to retain large fractions,  $\sim$ 20-50%, of the D<sub>2</sub> gas fuelled per quiescent discharge, regardless if the Mo surfaces are cleaned of, or partially covered by, B films. Several experiments and calculations show that it is improbable that B is retaining significant fractions of the fuel. Rather, the retention is occurring in Mo and W surfaces through ion bombardment, implantation and diffusion to trap sites. Roughly 1% D of incident ion fluence,  $\Phi_D$ , to surfaces is retained, and with no indication of the retention rate decreasing after 25 s of integrated plasma exposure. The magnitude of retention is significantly larger than extrapolated from the results of laboratory studies for either Mo or W. The high levels of D/Mo in the near surface, measured directly post-campaign (~ .01) in tiles and inferred from gas balance, are consistent with trapping sites for fuel retention in the Mo being created, or expanded, by high D atom densities in the near surface which arise as a result of high incident ion fluxes. Differences between C-Mod and laboratory retention results may be due to such factors as the multiply-ionized B ions incident on the surface directly creating traps, the condition of the Mo (impurities, annealing), and the high flux densities in the C-Mod divertor which are similar to ITER, but 10-100x those used in laboratory studies. Disruptions produce rapid heating of the surfaces, releasing the trapped hydrogenic species into the vessel for recovery. The measurements of the large amount of gas released in disruptions are consistent with the analysis of tiles removed from the vessel post-campaign - the campaign-integrated retention is very low, of order 1000x less than observed in a single, non-disruptive discharge.

#### **1. Introduction**

Controlling the hydrogenic fuel inventory in plasma facing components (PFCs) will be necessary for the successful operation of burning plasmas that use tritium (T) as a fuel. In ITER, the invessel limit of tritium is 350-700 g for safety reasons, while ~50 g is fuelled into the vessel for a full power discharge [1]. If the desire is to reach 1000 discharges before the T limit is reached then only 0.7 - 1.4% of the injected T (on average) could be retained in each discharge. Larger retention rates would mean more frequent stoppages to effect reduction of the in-vessel T through some method. A recent analysis of the database of retention measurements from current carbon PFC machines [2], including T retention experiments in TFTR and JET [3-6], led to the conclusion that ITER retention rate will be higher than the requirement introduced above but with quite large uncertainties. The prediction of T retention is thus a central focus of edge plasma research as it has important implications for the operation of ITER and reactors. We note that the current ITER design calls for a mixture of materials with PFCs in the high-heat flux regions of

the divertor manufactured from carbon fiber composites, the remainder of the divertor surfaces (baffles and dome) from tungsten, and PFC surfaces beyond the divertor of Be.

Most present tokamaks operate with carbon (low-Z) PFCs. Gas particle balance is a widely used tool to study fuel retention in these devices and show that ~25-50% of fueled gas is retained [7-9, 2, 10-12], ~2-3 times higher than post-campaign (ex-situ) accounting of D found in surface films integrated over a run campaign [13-16, 12]. Such research has led to the widely held conclusion that retention of the fueled gas in C devices is dominated by co-deposition of deuterium in plasma-deposited films. Therefore the rate of fuel (D/T) retention is set by C erosion and transport, together with the D concentration (D/C) in growing films. Recovery of the fuel is obtained by heating or removing those films [17-19, 2].

Unlike carbon, there is relatively little tokamak experience of fuel retention measurements with refractory metal PFCs such as W or Mo. Both metals are used due to their ability to withstand high heat fluxes and their low hydrogenic sputter yields. Tungsten is presently planned for use in ITER as a PFC, and may be desired over carbon in reactors due to its lower erosion rate[20-25]. In general the retention of hydrogenic isotopes in tungsten has been studied primarily in laboratories as opposed to tokamaks. Like carbon, co-deposition can occur in W, but it will be reduced relative to C due to its higher erosion resistance (physical sputtering is ~10x lower than chemical and physical sputtering for C) and low T/W in films (another factor of 5-10[26]). Unlike carbon, retention in tungsten PFC surfaces is likely dominated by ion implantation into the tungsten surface, followed by diffusion and trapping within the 'bulk' of the material, where here we define bulk as past the implantion zone < 10 nm. Reviews of the many laboratory studies of tungsten and molybdenum can be found elsewhere (e.g. [22, 27]). Molybdenum, like tungsten, is a body-centered cubic (BCC) metal having the same lattice structure and general characteristics – both are from the same column, group 6, of the periodic table. These refractory metals have high H diffusivities, even at room temperature, ~ 11 orders of magnitude higher than the diffusivity of H in carbon, such that in relatively short times the H can access the bulk by diffusion from a surface source such as a plasma. Hydrogen also has an extremely low solubility in molybdenum and tungsten in comparson to carbon. Hydrogen is found to diffuse in solution through the lattice and reside, long-term, in potential energy wells, or 'traps' [22, 27] which can be due to lattice vacancies, dislocations, interstitial atoms (an extra atom in the lattice) or larger

defects (e.g. voids, clusters of vacancies or dislocations). We note that the effective solute density at tokamak-like gas pressures should be many orders of magnitude less than that of inherent traps ( $C_{trap} \sim 10^{-5} \text{ x n}_{Mo}$ ) and the potential well depths for solute sites are much smaller (< 0.4 eV) than that for traps (1-2 eV).

Alcator C-Mod operates with PFC components fabricated exclusively of solid molybdenum and tungsten. All PFCs are Mo with the exception of one toroidal row of the tiles in the outer divertor high heat flux region. That row was replaced with solid tungsten PFCs employing a lamella geometry during one of the run campaigns from which data is included in this paper. C-Mod, like all other major tokamaks, intermittently coats PFC surfaces with thin, ~100-200nm, boron (B) films (boronization). Amorphous boron (Z=5) films are similar in their capability to store hydrogenic fuel as carbon (H/B, H/C ~ 0.4[28, 29]). Comparison of C-Mod operation with and without the B coatings was recently made [30]. In general the plasma radiation was found to be dominated by molybdenum with  $n_{Mo}/n_e$  reaching  $10^{-3}$  during ICRF (Ion Cyclotron Radio Frequency) heating when all the B films had been removed prior to the start of the campaign. When the B coatings were re-applied the level of Mo in the plasma dropped as much as a factor of 100, but generally returned to pre-boronization levels within 10s of discharges. The rate of Mo radiation increase after application of a new boron layer was primarily dependent on the cumulative amount of injected ICRF power which gradually wears off the B layers to re-expose Mo underneath.

In parallel with the above boronization studies fuel retention data was acquired both with and without boron layers and is the subject of the current paper. The experimental evidence strongly points toward fuel retention occurring in molybdenum PFCs through ion implantation. The amount of retention, when normalized to incident ion fluence, is  $\sim 1\%$ .

In addition to understanding retention, it is desirable to understand what processes might remove D from the surface. As we will show, disruptions lead to rapid radiative heating of plasmaviewing surfaces. This causes a significant fraction of the D retained in the first few hundred microns from the impacted surface to be released as molecules and recovered by the vacuum pumps.

In section 2 we describe the methods we have used to measure retention. Section 3 describes D retention during quiescent discharges with surfaces both cleaned of boron and with boronization

layers on PFCs. The evidence strongly, and somewhat surprisingly, indicates that the observed retention is due to ion implantation into Mo divertor surfaces as opposed to co-deposition in boron surface films. Section 4 covers the long-term retention which, due to disruptions, is much lower than that for a single, quiescent discharge. Lastly, in the discussion section, we explore how such tokamak data compares to laboratory studies.

#### 2. D retention measurements

The particle inventory remaining inside Alcator C-Mod after a single discharge is determined two ways[30]: 1) 'Integral gas balance' - Integrating the gas removed from the vessel by torus pumps during and after a discharge; and 2) 'Static gas balance' - Keeping all torus pumps closed off from the vessel for a period of 5-10 minutes, starting before the tokamak discharge until long afterwards, during which the pressure in the torus comes to equilibrium.

Central to the integral gas balance method of measuring retention is calibration of the pumping speed of the torus pumps as a function of pressure. While pump manufacturers publish general guides for pumping rates it is best to calibrate pumping speeds in situ to take into account the various conductances empirically. The pumping rate calibration is done by filling the vessel to a known pressure of D<sub>2</sub> followed by opening the torus pump valves and recording the torus pressure vs time. Calibrated capacitance manometers are used over a range in pressures from 13.3 Pa down to ~  $1 \times 10^{-3}$  Pa. Such gauges have the added benefit of being independent of gas type (although the calibrations were done with D<sub>2</sub>). Ionization gauges are used in the range where they overlap with the manometers ( $1 \times 10^{-1}$  Pa to  $1 \times 10^{-3}$  Pa) as well as below that. The vessel volume along with the derivative of vessel pressures determines the pumping rate as a function of torus pressure. Over repeated derivations of the pumping rate in this manner the spread in the derived pumping rates can be very large (> 20% at low pressures). The uncertainty is lowest at the highest pressures (13 to  $1 \times 10^{-2}$  Pa), more in the range of 10%.

Over the last 5 years we have developed a new static gas balance method which is a more accurate measure of gas retention occurring during a plasma discharge [30, 31]. While the valves to the pumps remain closed (starting before the discharge) the amount of gas remaining in the vessel following a discharge is thus a measure of the difference between that injected and that retained by vessel PFCs. The C-Mod discharge gas injection system consists of calibrated plena

volumes with attached capacitance manometer gauges. The amount injected through a specific valve into the C-Mod vessel is determined by the drop in the corresponding plenum pressure.

The same calibrated injection system is used to determine the C-Mod vessel volume; a known amount of gas (pressure drop in injection plenum x plenum volume) is injected into an empty vessel with all torus pump valves closed. The resulting pressure in the vessel is deduced from that of the injection plenum by the ratio of volumes. One limiting accuracy of this measurement is associated only with the accuracy of the gauges (~ 1%) and plena volume measurement (< 1%). Examining a series of 'fizzle' discharges provides another measure of the overall uncertainty: Fizzles are discharges where the initial formation of the plasma (a few 10s of kA for 10 ms) is accomplished but the plasma current rampup fails due to loss of plasma position control during the plasma breakdown. There is very little plasma wall interaction compared to a successful tokamak discharge, and essentially none in the divertor regions since the plasma is limited. Based on the variation from fizzle to fizzle the particle accounting with gate valves closed is within ~  $0.06 \text{ Pa-m}^3$  (~  $3.5e^{19}$  D atoms). This serves as the limiting uncertainty below levels of retention of ~  $3.5x10^{21}$  D atoms given the gauge uncertainties cited above. Because of the better accuracy of the static gas balance method, we have relied primarily on it and employ it implicitly in this paper unless otherwise noted.

An example of the measurement of gas retention/release using the static gas balance (method 2) is shown in Fig. 1. The pressure in the vessel reaches an equilibrium value before the gate valves



Figure 1: Pressure in the vacuum vessel vs time for a non-disruptive discharge (Time=0 corresponds to plasma discharge initiation). There is no pumping until just before 300s when the gate valves to the pumps are opened. The gas injected is shown for comparison.

to the pumps are opened 300 s after the discharge. Any slow increase in the pressure just before the valves are re-opened, if it exists, is extrapolated in time until the next discharge; an exponential is fit to the pressure vs time data over the last 100-150s. Note that the period of gate valve closure, 5-10 minutes, is a large fraction of the typical time between discharges,  $\sim$  15 minutes. It is our opinion that any temporary, or dynamic retention, which we define to be solute D implanted in the near

surface that is not residing in material traps, will diffuse out of the surface in that time period:

- a) It is likely that the implantation zone (< 5 nm) is super-saturated having D density over and above the natural solute level. Such D will quickly diffuse the short distance to the surface and recombine immediately (< millisecond).</p>
- b) The D implanted during the plasma discharge can diffuse into the material and is either trapped or remains mobile as solute D. Assuming a 200K difference in material temperature between the discharge and afterwards the solute D diffusivity will vary by a factor of ~100 (10<sup>-10</sup>m<sup>2</sup>/s to 10<sup>-12</sup>m<sup>2</sup>/s [32]). This means that the ~ 10 micron depth filled in by diffusion during the ~ 1 second discharge will need 100 seconds to allow the D to diffuse back to the surface and be released. The shape of the time trace suggests the effect of changing surface temperature as the D comes out most quickly soon after the discharge when the surfaces are warmest consistent with thermally-activated release of solute D. In other words, over short time scales when the surface is hottest the pressure rise is quickest (e-folding time ~ seconds, characteristic of a high diffusivity). Over long time scales the pressure rise is much slower (e-folding time ~ 100 seconds). In between the pressure evolution, and characteristic e-folding time, changes continuously. Of course some significant fraction of the D in the surface can diffuse to available solute and trap sites sites further into the material as well. But clearly such deeply-diffused D cannot be considered a temporary reservoir for solute D.
- c) During the period between discharges the absolute pressure in the vessel is in the range  $10^{-5} 0.1$  Pa and cannot impede any further release of solute D. Therefore when the pressure stops rising the reservoir of mobile deuterium in solution, which is presumably responsible for dynamic retention, has simply been depleted. Any D remaining in the material is effectively trapped.

Thus the static gas balance mesaurement measures the long-term (trapped) D. The use of the term 'static' retention has also been applied to long-term retention in carbon[33, 11].

The residual gas analysis (RGA) spectrum is utilized to occasionally analyze the gas species remaining in the vessel after a discharge. When the torus pumping is closed off from the

tokamak a low-conductance bypass is typically open allowing for the mass spectrum to be monitored. Deuterium is by far the dominant constituent in the gas remaining in the vessel after a discharge, once the H/D ratio of plasmas is < 5% as is typical for the majorty of run campaign. For example, the data from a set of discharges constituting a plasma density scan, typical of what we obtained at other times, show that the contribution to the overall pressure by  $C_xH_y$ ,  $C_xD_y$ , CO, H<sub>2</sub>O, D<sub>2</sub>O and other non-pure hydrogenic gases is in the range 0.1 – 1.5%. We do not account for such gases in the retention measurments of this paper. This leads to a slight underestimate of the amount of retention.

One concern with such pressure measurements is the variation in gas temperature between the injection plena volume and in the the tokamak. During operation the vessel surfaces, on average (spatially), are kept at ~ 0°C. There are regions that are colder due to the proximity of liquid nitrogen cooled coils and imperfect coverage of external vessel surfaces with heaters. Any differences between vessel and plena temperatures is folded into the vessel volume calibration since it is done under standard operating conditions (magnets cold), but without plasma. The surfaces that are typically heated by the tokamak discharge (100-200 °C) are relatively small (0.25 - 0.5 m<sup>2</sup>) compared to 60-70m<sup>2</sup> of vessel and internal hardware surface area. During a disruption even smaller areas (~10<sup>-3</sup> m<sup>2</sup>) are heated to high tempertures. As surfaces are heated by the plasma the external heater power, required to compensate for cooling from nearby coils is reduced to keep the vessel at the set temperature. Because the plasma-heated areas are so small the average gas temperature after a discharge should be unaffected by disruptions or typical discharges. Even if surfaces are heated, the gate valves are closed for a long enough period that most surface temperatures to drop back to the normal setting.

We note the accuracy of C-Mod retention measurements is not complicated by some other potential issues. There are no additional external fueling sources (e.g. heating neutral beams or pellets) nor any other active particle sinks (e.g. cryopumps, neutral beam lines) which greatly ease the ability to make accurate gas balance. Discharge cleaning (e.g. glow, or electron cyclotron discharge cleaning, ECDC) is generally *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 D in the wall. The recent installation of an internal cryopump in Alcator C-Mod indicates that its use is compatible with this method of determining fuel retention. One simply needs to regenerate the pump between

discharges, while the valves to external pumps are closed, in order to determine the gas pumped during the discharge. While the data shown in this paper are taken without the cryopump active (pump surfaces kept at room temperature), the small set of experiments utilizing the cryopump to date show that its use does not affect the absolute amount of retained D. However, the use of the cryopump forces the use of much larger amounts of injected gase for the same plasma density. In addition, the amount pumped and puffed varies as the pumping geometry is varied even as the retention remains constant. Such dependencies makes the comparison of retention to injected gas difficult in a quantitative sense.

All discharges from which data for this study are obtained are diverted and fuelled with  $D_2$  gas injection only, unless otherwise specified. Both ohmically-heated and auxiliary-heated discharges are studied. Auxiliary heating utilizes H-minority ion cyclotron radio frequency (ICRF) waves. Typical plasma parameters used are: major/minor radius R/a=0.67/0.22 m, plasma current I<sub>p</sub>=0.8-1 MA, toroidal field B=5.3 T, shot duration ~2s, with ~ 1-1.5s spent with flattop (constant) I<sub>p</sub>. The ion flux to divertor surfaces is determined utilizing Langmuir probes embedded in those surfaces [34]. The ion flux to the main chamber surfaces is inferred from neutral pressure and  $D_{\alpha}$  brightness measurements assuming toroidal symmetry and that the local neutrals are the result of local recycling as opposed to divertor leakage [35]. A description of the boronization technique, which is intermittently applied during each experimental campaign can be found elsewhere [30].



Figure 2: Profile of B/Mo on tile cleaned in an uiltrasonic bath showing the B diffusing into the Mo surface.

In preparation for each run campaign included in this study all PFC surfaces were 'cleaned' of the fairly thick B coating (> 1 micron) accumulated during the multiple boronizations (each depositing ~ 150 nm thickness B averaged over PFC surfaces) of the previous run campaign. The cleaning consisted of either an in-situ wipe of PFC surfaces with cloths wetted with distilled water or, if tiles were removed, ultrasonically cleaned in a heated distilled-water bath. RBS ion beam surface analysis of cleaned tiles revealed B/Mo fractions in the range of 15-30% (~  $2x10^{22}$  B/m<sup>2</sup>). It appears that a substantial amount of B diffuses into the Mo to depths of order a micron (Figure 2) forming boron-impregnated Mo, thus not a film on the front surface. The concentration of C and O in, and on, the tiles is typically below 1%.

For the purpose of this study we break the run campaigns up into three periods: Period 1 is after startup with tiles 'cleaned' of B. This can last 1-3 months and range from ~ 300 - 600 discharges. The longest period (2005) allowed us to thoroughly examine operation with all Mo surfaces [30]. Period 1 is used to condition the ICRF antennas to deliver high power as well as to lower the intrinsic H levels (due primarily to H<sub>2</sub>O from the vacuum break) in the discharge as required for efficient H-minority heating (H/(H+D) ~ 5%). The latter constraint is also important for D retention studies as we know the contribution of surface outgassing of H will be low and not contributing significantly to the gas balance measurements. Period 2 is the time immediately following (30-50 discharges) each boronization when the newly-deposited B surfaces release D under plasma bombardment and dominate fueling of discharges. Lastly, period 3 occurs sufficiently past each boronization levels) and the fueling requirements for the discharge have returned to pre-boronization levels.

It is important to note that in the boronization process the B and D are in fact co-deposited (D/B  $\sim 0.4$ )[28, 29]. Thus, in contrast to bulk carbon surfaces, erosion of boronized films (and liberation of D) followed by subsequent redeposition cannot possibly increase global fuel retention; it just changes the spatial location where the D is held by the boron films We also know from ion beam analysis of tiles removed from C-Mod that the boron layers build up over most of the PFC surfaces during the run period. The few regions where the B is most eroded are the active outer divertor surfaces (lower and upper divertors when both are used) as well as the outer, large major radius, poloidal limiters (at 2 toroidal locations).

All retention measurements shown are for non-disruptive discharges except for those described in section 4 where we are explicitly examining the effect of disruptions. Discharges where disruptions occur during current rampdown at low enough levels (below 200-300 kA) have been found not to affect significantly the measurement of retention and are included in the following single-discharge retention studies.

## 3. Deuterium fuel retention

### 3.1 Period 1, pre-boronization

We first examine the fuel retention in discharges immediately following a vacuum break when the surfaces have been cleaned of boron. Retention data 0.2is obtained only after we have determined that the H outgassing (due to H residing on and in surfaces as a result of water uptake during the vacuum break) approaches the low equilibrium levels required for ICRF heating (< 5% H/[H+D] fraction); this typically requires < 200 discharges after startup.

The amount of D retained during a C-Mod plasma discharge in period 1 of run campaigns exhibits several interesting features (Fig. 3a). First the level of retention can be relatively large in comparison to the typical C-Mod fuelling level (Fig. 3b), indicating a high global retention of ~ 30% of the injected gas averaged density for varying levels of boron (Fig. 3c). Secondly, there is a no strong dependence of retention on confinement mode or ICRF heating. Finally, while there is an initially strong increase in



Fig. 3 Deuterium wall retention versus flattop line coverage & confinement mode. All discharges are from period 1 in the 2005 run campaign:  $I_p=1$ MA, B=5.4 T and magnetic equilibrium held constant.

retention with line-averaged density near  $10^{20}$ m<sup>-3</sup>, further increases in density do not change the retention significantly (Fig 3a).

We note that the level of retention normalized to injected gas is similar to that widely quoted for carbon PFC tokamaks (see Introduction). It is higher than recently reported for ASDEX-Upgrade [36]. However, those ASDEX results are with strong cryopumping; C-Mod retention, when normalized to the amount of fueled gas, indeed drops by a factor of  $\sim 10$  to a level similar to that found in ASDEX-Upgrade when the cryopumps are used. Yet the total number of D retained per discharge remains constant. In any case, as we have discussed already in Section 2, comparison

of retention normalized to fueling across machines is problematic. It also does not address the underlying plasma implantation, diffusion and trapping processes.

The data in Figure 3 are strong evidence that co-deposition of D with B is not leading to significant retention in period 1 discharges. Using the B remaining on PFC surfaces after cleaning, ~  $2x10^{22}/m^2$ , we can estimate an upper limit for the total amount of retention due to codeposition with B: The typical plasma wetted region of PFCs is  $< 0.5 \text{ m}^2$  but for simplicity we will assume  $1 \text{ m}^2$ . We further assume that the maximum D/B in co-deposited layers to be 0.4. Then, the maximum amount the B can retain would be  $0.4x2x10^{22} = 8x10^{21}$  deuterons. However, this requires that a) all boron films were net eroded and re-deposited elsewhere, and b) the initial B layer, before erosion, holds absolutely no H or D. This upper bound estimate corresponds to the observed retention in of order 10 C-Mod discharges before the B co-deposition would saturate. However, the discharges shown in figure 3 were from a particularly long campaign to study plasma performance without boronization after a vacuum break; they occurred ~ 550 discharges into the campaign, and there was no evidence that the retention decreased during that period of the campaign. We note that the assumptions given above are certainly overestimating the effect of B co-deposition on retention. With respect to assumption a) the remaining B is actually not in a film, but distributed throughout the first ~micron of the Mo surface (Fig. 2), so an effective net erosion of the accompanying Mo at the same rate as the B would be required, which is difficult to achieve. Also we know that assumption b) underestimates the lack of D in the as-deposited B layer: From post-campaign tile analysis D/B in the boron layers is typically around 0.1 (not 0), roughly 1/4 of that expected for deposition during boronization and likely consistent with expected D/B levels when embedded in Mo rather than existing as an amorphous film. Thus the little B left in the chamber after cleaning is far too small in magnitude to provide the observed retention unless, somehow, the D that is co-deposited in the B layer were both completely desorbed of all hydrogen and deuterium, net eroded and removed from the vacuum chamber every 10 discharges. This seems unlikely. As will be discussed later (Section 5.1), even in period 3, when B films cover most of the PFCs and chamber, the co-deposition of D with B is also too small to explain the observed retention.

Retention processes that are active during a discharge without boron co-deposition must be related to ions implanting into surfaces. Adsorption of gas on surfaces is not important given that



Figure 4: Ion fluence to the lower divertor surfaces (measured by Langmuir probes) as well as to the surfaces outside the divertor (inferred from pressure measurements assuming all neutrals are due to recycling outside the divertor proper).

changing incident plasma fluxes.

that there is no observed retention when gas is injected into an empty chamber. Figures 4b-d display the ion fluence to the two lower divertor surfaces as well as that inferred to be impacting main chamber surfaces for the same discharges as Figure 3. The ion fluence to all surfaces are of similar magnitude and all increase with density. The outer divertor ion fluence correlates most closely with the D retention, both in the strong increase with density and the flattening at densities above  $1.5 \times 10^{20}$  m<sup>-3</sup>. If we normalize the retention to the divertor fluence (Figure 4d) we find that the retention fraction is in the range 1-3% of the incident ion fluence and varies with density. One concern about this dataset is that the rise and leveling of the outer divertor ion fluence (4b) occurs at higher densities than for retention (4a). We suspect that this shift may be due to the fact that we performed the density scan too quickly; we took only one discharge at each density which perhaps did not allow for the surface to fully equilibrate with To more directly address this question of surface equilibration, the density scan of Figures 3 and 4 was conducted again (period 1 data only) with the requirement that discharges be repeated with constant conditions (e.g. density, plasma current) until it appeared that the retention reached an equilibrium level, i.e. not changing from shot to shot. These new discharges were run at lower current (0.8MA) than for the discharges of Figures 3-4 (1 MA) to lower the detachment threshold. Separatrix strike point positions on the divertor plates were also adjusted in order to optimize the measurement of ion fluxes with divertor Langmuir probes. Based on the data shown in Figure 5, it is clear that some equilibration period is indeed needed that is dependent on

operating density or local fluxes. The first 5 discharges of the study ( $n_e \sim 8x10^{19} \text{ m}^{-3}$ ) evidence this effect most clearly. The amount of retention (Figure 5a) is quite high initially, ~ 5% of the divertor fluence (Fig. 5d), and drops over the next 4 discharges. The next 4 discharges were at a higher density ( $n_e \sim 1.15x10^{20} \text{ m}^{-3}$ ) where the decrease in retention from the 1<sup>st</sup> to last of the set was more rapid. These were followed by 2 discharges at the second lowest density ( $n_e \sim 1x10^{20} \text{ m}^{-3}$ ) where the retention equilibration was immediate. The next three sets of single density data were all run in order of increasing density ( $n_e \sim 1.3, 1.4, 1.55x10^{20} \text{ m}^{-3}$ ).

We again plot the retention normalized to divertor ion fluence (Fig. 5d). In all cases the fraction of divertor ion fluence retained reaches an equilibrium level between 1-2%. This supports ion implantation as the primary mechanism for retention. Including the inferred wall fluence as well lowers the retained



Figure 5: Density scan for a series of 0.8 MA Lmode plasmas with low amounts of RF power (~ 1MW). a) D retained over a discharge, b) ion fluence to both inner and outer divertor surfaces; c) ion fluence to main chamber surfaces inferred from pressure measurements; and d) the retention as a fraction of the divertor fluence.

fraction slightly further. We do not include neutral fluxes in this normalization since we do not have such a measurement. However, our estimate is that such fluxes should be of similar magnitude as the divertor ion fluxes. To put the C-Mod retention fractions in perspective we return to the example of ITER, but in the case where all divertor PFCs are tungsten. The total fluence to the ITER divertor plates (ions +



Figure 6: Retention characteristics for a sequence of constant, period 1, discharges.

neutrals, D+T) is ~  $1 \times 10^{25}$ /s [37]. If, as in the introduction, we ask what the maximum retention fraction would be that corresponds to reaching the 350-700g limit at 1000 discharges the answer is ~ .01%, 2 orders of magnitude lower than the singledischarge retention fractions of Figure 5. In other words a simplistic extrapolation of C-Mod results to ITER would lead to reaching the in-vessel tritium limit in of order 10 discharges. While we do not have any confidence in such an extrapolation it points out the necessity of understanding the underlying physical processes controlling the retention.

Up to some level ( $n_e \sim 1.4 \times 10^{20} \text{ m}^{-3}$ ) increasing density requires waiting for the plasma to 'equilibrate' with the surfaces. The amount of fluence required for equilibration is similar for the set of discharges at  $n_e = 0.8$  and  $1.1 \times 10^{20} / m^3$ 

(divertor fluence ~  $2x10^{21}$ ). Assuming this was implanted over 0.5 m<sup>2</sup> and using the diffusivity of D in Mo,  $D_{Mo}$ , of ~10<sup>-11</sup> m<sup>2</sup>/s [32] those atoms retained during the equilibration period (5 discharge periods) could reach a depth of ~ 7 microns implying a concentration of D/Mo ~ 0.01, a high value. However, the near surface D density should change immediately (< ms) so it is not clear what sets the equilibration period. These observations point to a process occurring deeper in the material (microns) that is perhaps related to varying thermal excursions of the surfaces. Note also that if the low level of residual B during Period 1 were playing a role through codeposition one would expect that the retention to only change when the fluxes, and therefore B deposition rates, change. This behavior is clearly not observed in Figure 5.

For the highest density case in Figure 5 the outer divertor is detached (the inner divertor is detached for all data points except for partial attachment in the lowest density case). As

detachment occurs and parallel pressure loss moves up the divertor plate the flux profile broadens. Thus, while the ion flux in the region of the strike point region has dramatically decreased, the overall fluence merely stops rising. Of course the profile broadens past the extent of the outer divertor probes and the total fluence to the divertor may be continuing to increase.

#### **3.2 Retention saturation in Mo and W and scaling with fluence**

An important characteristic of retention, useful in understanding the process, is how the retention scales with fluence. For laboratory studies of hydrogenic retention using beam implantation of ions there is a range of scalings. At room temperature, retention in W has been observed to saturate [38]. However, in general, laboratory retention experiments at higher material temperatures do not find saturation (Mo [38, 32], W [39-41]). Those same studies (usually performed at constant flux, as opposed to varying flux in a tokamak) show that retention scales with fluence,  $\Phi$ , of the form  $\Phi^{\alpha}$ , where  $\alpha$  varies from 0.345 – 1.0.  $\alpha$  of 0.5 would indicate diffusion-limited transport of the D deeper into the material. Diffusion for D in Mo or W consists of the D atom moving from one potential well to another in the lattice, the thermal energy of the D (~ 1/40 eV) being smaller than the potential well depth; Those potential wells can either be due to the periodic nature of the metal lattice ('solute', with well depths of ~ 0.4 eV) or due to defects in the material ('traps' corresponding to well depths in the range ~ 1-2 eV). Values of  $\alpha$  $\geq 0.5$  could be explained by various processes: a) the temporary enhancement of diffusion[27] (i.e. well depths or activation energy were decreased somehow due to ion implantation), b) the formation of new traps (potential wells) for the D to reside over and above that found to exist in 'virgin' material, not exposed to plasmas; and c) expansion of existing traps due to ion implantation such that a single trap site can hold more H atoms or even from molecules[42, 41, 43].

To test the scaling of retention with fluence a series of similar discharges were run (period 1), 6 discharges (1 MA) followed by 3 discharges at lower current (0.8 MA) with all other characteristics held constant. The retention/discharge (Fig. 6b) drops slightly over the first 3-4 discharges and then is constant, consistent with typical observations of equilibration times (section above). The ion fluence per discharge to the various PFC surfaces is roughly constant, increasing slightly on the transition from 1.0 to 0.8 MA; the 0.8MA plasma was in a higher recycling regime. The fraction of divertor ion fluence retained (Fig. 6c) also initially drops and then remains in the 1% range. These fairly similar discharges were followed by 6 more of varying pulselength (shown in Figure 7). The retention/discharge continued without rolling over, and essentially linear in fluence, over a total plasma time of ~ 20-25 seconds of diverted plasma (30 s including current ramp up and down) corresponding to a divertor fluence of  $\sim 4x10^{23}$  (wall fluence of  $1.7x10^{23}$ ). Thus the retention rate in C-Mod does not saturate or follow a  $\alpha \sim 0.5$ 



Figure 7: Retention (a) and ion fluence (b) for a series of different length discharges in period 1. c) Retention from a) normalized by each fluence of b).

diffusion power law. At first glance the constant retention fraction of 1% would seem to indicate  $\alpha$  ~1. This parameter will be discussed later in Section 5.2.

Another way of addressing the dependence of retention on ion fluence is to vary the plasma duration. Figure 7 shows that the retention (a), as well as divertor and main-wall ion fluxes (b), all increase approximately linearly with discharge duration. Three sequential discharges were run for each condition. Extrapolation of the linear fit (Fig. 7a) to zero pulselength could indicate that plasma startup (mostly in limiter configuration) and/or rampdown (mixture of limited and diverted) plays a role in D retention. This is not surprising or inconsistent with divertor retention since the plasma is diverted early in the discharge (~ 0.3 s) in current rampup and is diverted through most of rampdown.

The role of the current rise and fall phases is of decreasing importance as discharge duration increases. An alternative explanation for non-zero retention at zero pulselength is related to the fact that the divertor surface temperature is increasing through the discharge; if the D retention rate was higher at low temperatures, at the beginning of the plasma-divertor interaction, then the retention rate would decrease through the discharge as observed. Again, like the results of Figure 6, the results suggest little or no saturation of the retention capability of the PFC materials.

#### **3.3 Effect of boron layers**

Following period 1, which typically corresponds to  $\sim 300$  discharges, but in one case  $\sim 600$  discharges, the PFC surfaces of C-Mod are boronized. We find that the boron layers (100-200 nm) provide additional clues as to where the retention is occurring and the role of surface layers.

Operation of the tokamak immediately following a boronization (period 2) dramatically changes the retention characteristics of plasma discharges. Past studies of the effect of boronization on core plasma performance[30, 31] showed that there was little control of the plasma density following a boronization due to outgassing of PFC surfaces and fueling the plasma. This is not surprising in that when boronization films are deposited they are typically saturated with H or D (H/B ~ 0.4 [28, 29]) depending on the type of diborane used (B<sub>2</sub>D<sub>6</sub> in C-Mod), and thus represent a significant additional fuel source at the wall. In fact, integrating the gas release/discharge from Fig. 8, and assuming a surface area of 10 m<sup>2</sup>, the gas release after boronization during discharges

is roughly half of that coedposited during the boronization. Such partial depletion of the D in the B films is roughly consistent with the D/B~0.1 measured in B layers in post-campaign analysis of tiles. Note that the dominant PFC fueling of the plasma just after a boronization contraindicates B codeposition as an important fuel-retention process, since the codeposition rate will be highest when the plasma shots net erode the newly-deposited B films, and based on IBA patterns, redeposit at inner divertor.

Based on Figure 8 we see that there is a very gradual



Figure 8: D retained per discharge as a function of discharge number following a boronization (period 2). All data inferred through method 1 except the 2 points with squares (method 2).

changeover from wall-fueling to wall-retention, taking ~ 40-50 discharges past a full boronization. Past measurements show that molybdenum source rate at the outer divertor first drops precipitously at a boronization, and then returns to pre-boronization levels [44] as the boron is eroded. *The B erosion occurs on a time scale similar to the changeover from wall-fueling to wall-retention shown in Figure 8, which suggests that the retention requires a bare Mo surface in the outer divertor.* The period past the end of Figure 8, where the gas balance stabilizes to net retention, is designated as period 3 as described earlier.

The erosion of B and re-appearance of neutral molybdenum line emission from in front of divertor surfaces is consistent with post-campaign measurements. Analysis of a poloidal array of tiles removed from the vessel after the run period showed that the B layers build up on most other surfaces (are not eroded) over the multiple boronizations that occur. The areas of minimal remaining B correspond to where the most power goes - the lower outer divertor and the 2 low-field side poloidal limiters where B is eroded by RF effects or possibly startup. More recently operation of high power discharges has been with the upper divertor as well and we infer that the B layers there are also eroded on a similar time scale to the lower outer divertor. To summarize, B layers are quickly eroded from divertor surfaces that experience high power loadings from plasma contact. Such surfaces have some mixture of B and Mo due to the constant B flow to, and erosion from, the surface. The B layers elsewhere are generally not eroded during the entire

campaign and generally increase in thickness.

Once period 3 operation is achieved we are able to compare, from one discharge to the next, operation with plasma-wetted surfaces being mostly Mo to those with thick B films. The effect of changing the divertor strike-point position on retention is shown in Fig. 9 for a set of discharges where again the line–averaged density was kept constant (L-mode). The first 15 discharges of the runday were utilized for another



Figure 9: D injected and retained as the plasma-wetted surface was changed from the lower divertor vertical plate (a) to a 'slot' divertor (b), back to the lower divertor (a), and finally to an upper-single null equilibrium (c). d)  $D^0$  injected and retained.

experiment. In the standard LSN configuration, i.e. with the outer strikepoint on the lower outer divertor vertical target (Fig. 9a), the usual D fuel retention ( $\sim 7x10^{20}$  D/discharge) is found as expected from period 1 experiments at  $n_e \sim 1.6 \times 10^{20}$  m<sup>-3</sup> (e.g. Figures 3, 4 & 6 for 1 MA plasmas). Subsequent repeat discharges 17-21 result in the same level of retention with no evidence of retention saturation, although the fuelling requirements evolve somewhat. There is an immediate drop in the D retention per discharge, as well as the required D fuelling, when the outer strikepoint is switched to the horizontal floor of the divertor (see Figure 9b), a location normally avoided for plasma operation, and which has thick B films post-campaign. The D retention reaches a small but measurable value after three discharges, about 25% of what was measured for the first 6 discharges of this sequence. The last three discharges of the sequence shown in Fig. 9 are with the strike points on the upper divertor, upper single-null plasmas. The retention returns to the typical level associated with the lower divertor. Note we do not have ion fluence data for discharges 22-26 as there were no divertor probes in those surfaces. We want to emphasize that the retention during period 3 discharges is indistinguishable from period 1 as long as the strike points are in their usual positions. We note also that when the slot-divertor discharge (Fig. 9b) was repeated during period 1 section of a following campaign, net retention occurred at the usual levels consistent with normal outer divertor strike point location.

All the empirical observations listed in the above three sections are consistent with ionimplantation in Mo as the retention process and the outer divertor (LSN) or upper divertor (USN) as the primary location. There may be an alternative explanation of the retention as occurring in the B layers but so far we have not been able to construct one that matches the phenomenology or magnitude of overall fuel retention in C-Mod. In particular, since the boronization process in C-Mod produces boron layers saturated with D, then any erosion of B (and D) and subsequent co-deposition should not lead to net retention; the D is merely being shifted around. In fact, in such a situation one would expect a fuel surplus and that is exactly what is observed. This should be contrasted with carbon PFC tokamaks where the source of unsaturated low-Z impurities is intrinsic and essentially infinite. Since carbon surfaces are always being eroded there are always 'virgin' areas of C (without any D) being uncovered and net-eroded; there is no such source in C-Mod. An added argument against the importance of co-deposition of D with B is the fact that we find approximately the same level of retention in period 1 and 3. It is improbable that D/B codeposition, if it occurred in period 3, would always lead to the same level of retention as ionimplantation (period 1). Certainly the amount of boron in the machine is orders of magnitude different in the 2 periods.

#### 3.4 A process for enhanced retention in molybdenum

If ion-implantation is the primary mechanism for fuel retention in C-Mod, then what is the physics that yields the observed retention rate and non-saturated behavior? The concentration of inherent 'traps' in W,  $C_W$ , is less than  $10^{-4}$  for un-annealed material, as much as a factor of 20 higher than when the material is annealed at 1673K for 1 hour [45] (Mo, also being a group 6 BCC metal should have similar levels of traps). But measurements of D concentration in the near surface region of laboratory samples range from a few % (D/Mo [32], D/W [41] to 5% (for D/W[40]). The preliminary estimate of D/Mo from C-Mod of ~ 1% given earlier in the discussion of Figure 5 is consistent with the laboratory measurements. The high level of D/Mo

C-Mod data, indicates that either the existing traps have expanded to accommodate more D (e.g. voids grow), or new traps are being created.

relative to intrinsic  $C_{Mo}$ , together with  $\alpha > 0.5$  from

The energy of the impinging  $D^+$  is not high enough to displace Mo atoms in the lattice directly and create new traps. In the case of Mo the atom displacement threshold energy is  $E_{disp} = 35 \text{ eV}$  [46]. Since the maximum energy transfer to Mo from incident ions with energy E is

$$E_{disp} = 4M_1M_2E/(M_1 + M_2)^2$$
(1)

the minimum incident  $D^+$  energy to displace Mo atoms is ~ 440 eV.

One possible mechanism for creation of new, or expansion of old, traps is for the pressure of atoms in the lattice to be high enough to distort the lattice [47, 40, 41, 32]. The density of D within the lattice



Figure 10: The gas injected and retained (a), divertor and wall fluence (b), and retention fraction for a density scan using He fuel gas.

is determined by a combination of two mechanisms: 1) the limiting rate of D atoms diffusing back to the surface from their implantation range (a few nm); and 2) the rate of D atoms recombining into D<sub>2</sub> and being released at the surface. Recombination, which has been found to be the limiting rate in a laboratory study of Mo [32], would enhance the D density beyond that of diffusion alone. In that case one can write down an equilibrium relationship, equating the incident ion flux,  $\Gamma_{D+,in}$ , with the release rate of D<sub>2</sub> from the surface,  $\Gamma_{D2,out}$  (neglecting the diffusive flux further into the solid which, as we know from the retention fraction, is of order 1% of the incident flux):

$$\Gamma_{D+,in} = \Gamma_{D2,out} = \mathbf{R} \cdot \mathbf{n_{D,s}}^2 \tag{2}$$

where R is the recombination rate  $(m^4/s)$  and  $n_{D,s}$  is the local D atom density at the surface. Unfortunately we cannot directly calculate  $n_{D,s}$  as the recombination rate in Mo and W is very uncertain, ranging from  $1x10^{-5}$  m<sup>4</sup>/s [48] (W) to  $1x10^{-26}$  m<sup>4</sup>/s [45] (W) to  $1x10^{-30}$  m<sup>4</sup>/s[32] (Mo).

To test the importance of pressure-induced trap formation/expansion model in C-Mod we have performed a density scan in 1MA L-mode (some RF heating) He plasmas during period 3. The release of He from the Mo surfaces is not limited by surface recombination. Furthermore, the diffusivity of He in Mo is 1600x higher than for D in Mo[49]. Thus, even without recombination playing a role, we expect  $n_{He}$  at the implantation range to be 1600x lower than  $n_D$  for equivalent incident He<sup>+</sup> and D<sup>+</sup> ion flux conditions (and the same surface temperature). Taking recombination into account would obviously increase that difference. Prior to the run day the vessel surfaces were conditioned utilizing low-temperature He plasmas (generated by electron cyclotron RF waves). Figure 10a displays both the He injected (solid squares) and that retained as a function of line-averaged density for that set of lower-single-null diverted discharges. At the lowest ion fluence/discharge (plasma density), there is some retention at a low level, 0.3%, less than found for D-fueled discharges. At higher plasma densities or fluences > 1.5x10<sup>22</sup> there is no retention within error bars (and possibly some net outgassing), consistent with the idea that no additional sites, or expansion of sites, for He retention has occurred.

It is possible that even after He discharge pre-conditioning of the PFC surfaces that  $D^+$  implantation was still playing a significant role (albeit, at low levels). We monitored the brightness of the  $D_{\beta}$  and HeI (502 nm) emission lines for a periscope view integrating emission across the inner and outer divertor plasmas. The ratio of those lines gives a gross measure of the relative recycling of He and D in the divertor. That ratio (Figure 11) was found to drop dramatically from the 2nd to the 4<sup>th</sup> discharge of the run day (the line brightnesses were not monitored for the first discharge) and then stayed constant at a low value for the density scan data included in Figure 10 (a criterion for inclusion in that dataset). The drop in D recycling light at the divertor correlated with a drop in retention at the same operating density (same Figure 11). The main inference to draw from the data of Figures 10-11 is that the amount of retention correlates with the amount of D in the discharge and flux to PFCs. The results are consistent with a model of trap formation or expansion driven by high D pressures in the near surface. The high D density increases the recombination-limited release of D<sub>2</sub> from the surface (Eq. 2). An alternative explanation is that the retention during D-fueled discharges is driven by co-deposition



Figure 11: The ratio of Balmer  $D_{\beta}$  to HeI in the divertor at the beginning of the run day.

with B and conversion to He discharges removes that retention mechanism. However, as will be discussed later, we cannot justify the D/B co-deposition process as important for retention in C-Mod in period 3 or 1.

Note that one might expect He plasmas to lead to higher retention if lattice distortion due to the high pressures were not playing an important role. Assuming both He and D are implanted into Mo at the same rate He is more likely to nucleate, forming a gas and distorting the lattice[50, 16]. In order to ascertain whether the He discharges with no retention had reached an equilibrium, 5 discharges were run sequentially with approximately the same characteristics. The data are shown in Figure 10 for line-averaged density ~  $1.4 \times 10^{20}$ . Those same 5 He discharges are shown in Fig. 12 followed by 3 discharges with D<sub>2</sub> as the fueling gas (same plasma characteristics as for the preceding He discharges). The lack of retention is consistent across the He discharges. Following the changeover to D<sub>2</sub> we find that both the atoms injected and retained dramatically increase. By the third discharge of the D<sub>2</sub> sequence the retention has dropped to a 'normal' value (~ 30% of injected gas is retained, ~1% of divertor ion fluence). The strong retention following the changeover from He- to D<sub>2</sub>-fueled discharges would be consistent with the model of D<sup>+</sup> impact leading to trap formation; n<sub>D,s</sub> in the near surface must be much higher than the n<sub>He</sub> in that region to increase the surface loss of D into

 $D_2 - Rxn_{D,s}^{2}$  (Eq. 2).

Figure 12 is also illustrative of the effect of boron layers as described in section 3.3. Following the changeover to  $D_2$ , a sequence of discharges were also run where the plasma geometry was changed from diverted to limited on the inner wall tiles for the entire discharge (2.5 s) as opposed to only the first 0.25 seconds before the plasma topology switches to diverted. The retention then drops back to essentially zero. Using the



Figure 12: A sequence of discharges where the gas and plasma-surface interaction surface was varied. The atoms injected and removed/retained are given.

profile of  $D_{\alpha}$ -derived recycling at the inner wall we have estimated the ion fluence to the limiter surfaces integrated over the discharge to be ~  $1.5 \times 10^{22}$ , similar in magnitude to the divertor fluences earlier in the day (Figure 10b), so we would have expected similar levels of retention. We believe thick (100s of nm) boron coatings on the inner wall are the reason for this lack of retention. Note that since the diffusivity of D in boron is 11 orders of magnitude lower than D in Mo near room temperature, the boron layer quickly saturates under ion bombardment and acts as a barrier to D retention in the underlying Mo.

#### 4. Long-term fuel retention

In the previous sections we have explored the D retention for single discharges as well as consecutive non-disruptive discharges (< 1 runday). Certainly such well-controlled discharges have high levels of retention that we have tried to explain in terms of ion-implantation into Mo with creation or expansion of trap sites within the material. We now turn our attention to the net



Figure 13: D concentration in an outer divertor tile removed from Alcator C-Mod.

retention over a long period of time. As mentioned earlier a poloidal array of tiles were removed during a vessel vent. Boroncoatings were thick over most of the tiles other than the outer divertor as the tiles were removed after many campaigns without any cleaning off of B from surfaces. As expected there was significant D in the B layers, roughly at an average level of D/B ~ 10%. This is similar to that previously measured [51]. Since that older study and this current tile analysis show most D retention to be in

the B layers outside the divertor, we initially assumed that co-deposition was the dominant D retention process. As a result the depth profile of retention in the molybdenum substrate was only measured in one tile from the outer divertor where the B was mostly eroded but ingrained in the Mo as shown in Figure 2. Looking at the results of this analysis, shown in Figure 13, one can integrate the D within the Mo to a depth of 2.5 microns. The resulting retention is roughly  $2x10^{21}$  D/m<sup>2</sup>. TDS analysis of the same tile (4 years later) gives the D retention a factor of ~ 3 lower. In either case the amount retained is small, of order 1 discharge worth assuming 0.5 m<sup>2</sup> of wetted area in the divertor. The effective retention rate averaged over the entire campaign would be of order 1000x less than inferred for the single discharges discussed earlier. The question then arises - what happened to the strong retention inferred for a single discharge? Either the analysis of the single-discharge retention in the Mo is incorrect or some process is removing the D over the run period, essentially as fast as it is being implanted. We think that the probability of the first is very low. We next offer evidence for the latter.

When we move away from controlled, non-disruptive discharges to what can and will occur during normal C-Mod operations we find that disruptions are removing D from within tiles and



Figure 14: Example of a set of discharges during a run campaign. Boronizations are marked with 'B'. The dicharges are a mixture of period 2 and 3. Gas balance measurements were made with method 1.

are playing a significant role in gas balance. Figure 14 displays the amount of D retained or removed/discharge for a sample set of discharges including periods after boronizations. There is a mixture of discharges that both lead to net retention and to net gas removal. The latter are sometimes driven by disruptions, although post-boronization discharges also lead to net gas removal as shown in Figure 8. The largest disruptions that occur during the sample period shown lead to liberation of an amount of gas equivalent to approximately one day's worth of controlled, non-disruptive, discharges of the type shown in Section 3.1-3.2. Integration of the removal/retention over the entire period shown leads to net removal. However, that does not take into account the D co-deposited during boronizations and how much is then removed during conditioning before plasma discharges. Given those effects as well as long-term evolution of gas from the surface we cannot make an accurate accounting of the net gas removal/retention over long periods. However, it is clear from Figure 14 that it can be much smaller than the singledischarge, non-disruptive discharges examined in previous sections. In general we find that there can be net retention or net removal in a given day (again, including naturally-occurring disruptions as in Fig. 14) depending on whether the tokamak is being operated at the edges of operational space (so more disruptive) and whether the stored energy in the plasma is high.

Certainly the idea of using disruptions to heat surfaces and release gas has been suggested as a method of T removal for ITER[18]. As for other tokamaks, the thermal quench time in Alcator

C-Mod is of order 100-200 microseconds. However, the current quench time, where typically the poloidal energy, ~ 5x the thermal quench energy, is dissipated in the plasma in a period ~ 10xlonger than the thermal quench time. This is shorter than other tokamaks due to C-Mod's size and thus, together with C-Mod's high energy densities, provides a unique opportunity to strongly heat surfaces. Since the temperature rise scales like the square root of time (and linear in power) the thermal and current quench in C-Mod lead to similar increases in surface temperature, one following the other, assuming the heat loads occur roughly on the same surfaces. The current quench would lead to deeper heating. This discussion also demonstrates why higher stored energy and higher currents can lead to larger temperature excursions in PFC surfaces during a disruption. Since the diffusivity and de-trapping rates for D in Mo and W increase exponentially with material temperature the amount of gas liberated in a disruption should be a strong function of thermal quench energy. Examination of a controlled set of disruptions supports the above reasoning[52]. Preliminary numerical modelling of the effect of disruptions [53] has been performed taking into account the range in plasma and magnetic energy, wetted area, timescale and the thermal diffusivity for Mo ( $D_{Mo,T} \sim 5 \times 10^{-5} \text{ m}^2/\text{s}$ ). The depth to which significant heating (1000-2000 <sup>0</sup>C) can occur on the disruption heating timescale,  $\tau_d$ , is  $\delta \sim (D_{Mo,T}\tau_d)^{0.5}$ , in the range of 150-300 microns, which is 10-100x deeper than the inferred depth of retention during a C-Mod discharge. Such high temperatures lead to enhancements of D diffusivity in Mo by factors of  $\sim 10^6$  with the result that D in the heated region can easily reach the surface and be released on a timescale of order a second. We note that such high temperatures are also higher than found for the main peaks in the thermal desorption spectra (TDS) of D from molybdenum tiles. We suspect that a single disruption does not anneal traps due to the short timescales. The details and sensitivity of this numerical modeling will be examined in a future article.

Given the above discussion we have examined all disruptions occurring during one run campaign to determine what fraction of discharges they occur in and when they occur. The majority of



Figure 15: Histogram of the disruptions occurring over an entire run period. For disruptions occurring during current rise  $I_{P,Max}$  is defined as the programmed value of the maximum current.

these plasmas had maximum currents in the range 0.8 - 1.0 MA although ~ 10% have higher currents, up to 1.5 MA. The results, displayed in Figure 15, show that disruptions occurring during the current flattop when the stored energy is highest, constitute 15% of all discharges. Disruptions during current rise and fall account for another 15% of all discharges. Unfortunately, we do not have the local measurements (temperature, D concentration within the Mo) to determine whether disruptions remove D from the same locations as where retention occurs during a discharge. However, in an overall sense there is a consistent picture connecting disruptions to removal of gas retained during

tokamak discharges: 1) Liberated gas is mostly the retained fuel,  $D_2$ ; 2) C-Mod disruptions terminate in vertical displacement events (VDEs), globally depositing energy on the divertor where, based on the results presented in previous sections, the majority of fuel retention occurs. Stronger evidence for the importance of disruptions in removing retained fuel comes from an analysis of the 15% of current flattop discharges from the survey above. *We find that those disruptions, on average, remove* ~ 6-7*x that which is retained during an average, non-disruptive, discharge*. With the caveat that this is a gross analysis of all such disruptions the net result over a run period would be near zero retention. Further work is required to look at specific trends in controlled experiments.

For completeness we also review the outgassing of surfaces over weekends between run weeks as this may be playing a role in the long-term loss of gas from surfaces. Over weekends the vessel temperature is increased from ~ 0 to 55  $^{\circ}$ C. As shown in Figure 16a the pressure in the vessel first rises due to the vessel temperature rise followed by a decrease over the following

several days. We have verified that during this period  $D_2$  dominated the RGA spectrum. In Figure 16b we show the integral of the gas pumped during each of 9 weekends studied. The total gas released in each weekend is equivalent to somewhat less than 10 retention-dominated discharges. It is apparent from the figure that the amount removed from in-vessel surfaces is independent of the number of rundays preceding the weekend and thus the number (or type) of discharges. We also point out that the amount of gas removed is independent of whether the weekend bake is



pre-boronization (period 1) or later in the run period. We do not expect such a bake to liberate D that is trapped. Thermal desorption spectra (TDS) analysis of tiles shows that the lowest temperature peak in the TDS spectrum of W and Mo tiles is typically above 600K[38, 40, 54]. But perhaps the gas evolved comes from D weakly attached to B layers.

In summary the weekend vessel bakes appear not to release gas corresponding to that retained during tokamak plasma discharges.

#### 5. Discussion

#### 5.1 Role of boron in retention

It is natural to ask whether boron is playing an important role in C-Mod fuel retention. Like carbon it can co-deposit with D. During most of the experimental campaign the walls are primarily covered with B layers thicker than the penetration range of the impacting ions.

The pre-boronization period of our run campaign (period 1) gives rise to the strongest evidence against D/B being an important retention mechanism. As discussed in section 3.1 the amount of B remaining invessel following a vacuum break cannot provide enough retention due to co-

deposition to account for the continual retention observed. An additional piece of information is that the level of fully stripped B in the plasma discharge is ~ 5x lower (period 1) than later in the campaign after boronizations (period 3). Given that the same level of retention occurs in both periods it is either due to the same retention process (ion implantation into Mo), or the same level of retention is just a coincidence with D/B co-deposition becoming dominant in period 3 and some other process compensating for the change in the boron density. The scenario of D/B co-deposition leading to retention is even more unlikely since it is necessary that the D co-deposited with B during the boronization process is somehow removed before retention measurements so that B erosion does not just lead to transfer of D to another location.

We can make an additional estimate of the amount of D retention that could be due to codeposition *IF the D co-deposited with B in the boronization process has been completely removed prior to period 3.* Comparison of areas of inner divertor tiles that are shielded from plasma and not (both are exposed to boronization) indicate that B layers are forming at a rate of 1-2 nm/s during tokamak plasmas, a number similar to C PFC tokamaks[14] and roughly consistent with the inner divertor ion fluence of ~  $2x10^{22}$ /s if there were 1% B ions in the D<sup>+</sup> flow. Assuming 0.4 D/B (higher than that measured post-campaign, D/B~0.1) over the entire inner divertor (~0.5 m<sup>2</sup>), 2nm/s co-deposition would lead to ~  $4x10^{19}$  D/s retained/discharge, a factor of ~ 25 less than observed (< $10^{21}$ /s) making it impossible that inner divertor co-deposition is significant. Approximately 12 m<sup>2</sup> of PFC area would have to be consistently the site of codeposition to achieve the retention we measure in a discharge. That is not possible since the total area of C-Mod PFCs is ~ 7 m<sup>2</sup>, which includes large regions having little or no contact with the plasma as well as erosion-dominated surfaces. In summary, we have not been able to construct the proper logical sequence that is consistent with D/B co-deposition dominating D retention in C-Mod during any period of operation.

Lastly, the characteristic drop in retention with increasing fluence in Figures 5 & 6 is not consistent with D/B co-deposition where the retention/discharge should be a constant ( $\alpha$ =1). This is emphasized further in the next section in the similarity between C-Mod and laboratory results for fluence dependence of retention.

#### **5.2** C-Mod retention relative to laboratory studies

As mentioned earlier the hydrogenic retention properties of both Mo and W have been studied under more controlled, non-tokamak conditions, herein termed 'laboratory' studies. Surfaces are typically prepared for testing by annealing at high temperatures (> 1000 °C) for short periods (~ 1 hour) to remove defects in the material due to manufacturing that could lead to hydrogenic retention (traps). In addition the surfaces are often cleaned or eroded to remove impurities (e.g. oxides) and the general vacuum levels are better than for a tokamak in order to minimize surface impurities (which would, for example, change surface recombination), thus wholly Mo or W. Such preparations serve to better compare results from different laboratories without worries about different manufacturers or contaminants. Another important characteristic of laboratory

supplied by either ion beams or plasmas, and the retention, are much better characterized than for a tokamak where it must be inferred globally as opposed to at a known surface. Tokamak experiments such as C-Mod, by definition, have more elements in play: impurities both on the surface and in the plasma, energy distributions of incident ions, disruptive heating of surfaces.... C-Mod results are relevant as all PFCs are high-Z (mostly Mo), the D retention properties of which are very similar to tungsten.

fuel retention studies is that both the ion fluence,

To give the reader some perspective on the C-Mod retention in relation to laboratory results we have utilized a few representative sets of laboratory retention data to predict the retention in C-Mod. This is done by fitting the laboratory data to the functional form: retention =  $A\Phi^{\alpha}$ , where  $\Phi$  is the incident ion fluence (#/m<sup>2</sup>). Based on the fluence data of Figure 6 and the assumed plasma-wetted areas in C-Mod (inner divertor -



Figure 17: Data from Figure 6 repeated along with predictions of D retention in C-Mod based on fits to laboratory retention data (open symbols in panel b). Laboratory experimental details given in Table 1.

 $0.25m^2$ , outer -  $0.35m^2$ , main chamber -  $1m^2$ ) the predictions for each dataset are shown in Figure 17 along with the C-Mod retention data. Note that the laboratory predictions should overestimate the C-Mod retention because we have implicitly assumed that there is no ion flux/fluence to the surface prior to the set of discharges of the calculation; any value of  $\alpha < 1$  implies that the retention/discharge should secularly decrease as fluence increases from 0.

There are differences and similarities between laboratory and C-Mod retention data. First, the C-Mod retention is considerably larger than most of the laboratory results. (As an aside we mention that it is typical in comparisons of D or H retention in Mo and W that Mo has slightly lower retention[38, 55]). Second, while we have emphasized the lack of saturation and constant

Experiment	Mat'l	Energy	Surface	Flux	$D^+$	Analysis	А	α
		(eV)	temp. (K)	$(10^{21}/m^2/s)$	source	tech.		
Wright [32]	Mo	100	400	1	plasma	IBA	.0013	1.0
Ogorodnikova [40]	W	200	290	.0204	beam	TDS	19815	.71
Ogorodnikova [40]	W	200	473	.0204	beam	TDS	217	.76
Venhaus [39]	W	100	623	2-13	beam	TDS	529.4	.71
Haasz [38]	Mo	1000	300	.01	Beam	TDS	$1.66 \times 10^{12}$	.35
C-Mod	Мо	~10- 100	300-450	1-1000	plasma	inferred		

Table 1: Characteristics of the various experiments corresponding to the data in Figure 17.

retention from one C-Mod discharge to the next, the C-Mod data dependence on fluence is strikingly similar to a number of the laboratory cases where  $\alpha \sim 0.7$ . That would imply similar processes occurring in both cases even though the absolute retention is higher in C-Mod Mo PFCs. The value of  $\alpha$  for C-Mod data cannot be ascertained with any accuracy given the low fluence of the Fig. 17 data compared to the laboratory studies. Nevertheless the important conclusion is that  $\alpha > 0.5$  implying some process occuring to enhace retention over that limited by diffusion into the bulk.

#### 5.3 Processes that might explain the disparity between C-Mod and laboratory data

It may be that the characteristics of C-Mod plasmas and surfaces are the source of the retention

differences between laboratory and C-Mod retention results evident in Figure 17. One obvious difference between C-Mod and laboratory experiments is surface impurities. C-Mod has boron on and in the surface (see Figure 2) as well as oxides on the surface. As an example 0.5 monolayer of oxide on Fe was enough to reduce the hydrogen release rate (recombination rate) by orders of magnitude[56]. Perhaps in the right range of impurity layer thickness, less than the range of the impacting ions, the recombination rate is raised such that the pressure buildup inside the surface is higher than in laboratory experiments and leads to more changes in the lattice (e.g. damage/displacements or expansion of exisiting traps)? It is hard to estimate the magnitude of such an effect and the ultimate retention engendered.

Impurities in the plasma can also lead to enhancements in retention. With sheath potentials reaching 10s of volts an impurity ion can, upon impact, directly create vacancies within the lattice. Figure 18 displays the vacancies per incident ion as a function of impact energy for boron calculated using SRIM[57]. Past studies, based on analysis of Mo sputtering [44], have inferred that there is a significant fraction of impacting B ions that are three-times ionized. For a sheath corresponding to  $T_e = 20 \text{ eV}$  the B<sup>+3</sup> energy approaches 200eV and a vacancy creation rate of 1 vacancy per incident B<sup>+3</sup> ion. In one modelling study [58] the number of hydrogen atoms that

can be accommodated in one vacancy is in the range of 2-3, while in another study, potentially more than 6 H atoms can be stored [59]. Based on C-Mod charge-exchange measurements of fully-ionized B,  $n_B/n_e \sim 1\%$  in the plasma. Assuming the same ratio of incident D<sup>+</sup> and B<sup>+3</sup> fluxes, together with a vacancy creation rate of 1, B<sup>+3</sup>-created vacancies could lead to a D<sup>+</sup> retention rate as high as 6% of the incident D<sup>+</sup> flux! A reduction of T<sub>e</sub> by a factor of 2 would lead to a factor of 10 reduction in the vacancies/incident ion. A concern with such an explanation is that



the boron levels in the plasma during the pre-boronization period 1 are lower by a factor of 5 than in period 3. Given the same levels of retention in the two periods one would have to argue

for a small change in sheath potentials to counteract the differences in  $B^{+3}$  fluxes. While this is possible we have no evidence to support such an effect.

A third effect of involving boron is its role after implantation into the lattice. It is known that irradiation of Mo with He, O, Ne and Bi before exposure to  $D^+$  fluxes leads to enhanced retention [60-62]. Such pre-irradiations are performed at high energies (many keV) so that direct vacancy production is occurring along with implantation of those impurities into the lattice. Nevertheless, the impurities in the lattice were observed to enhance retention of hydrogen through modification of the local potential structure. A more recent work involved simultaneous irradiation of W with C and D and found an enhancement of retention over irradiation with D ions only[43]. Thus in the case of C-Mod, B in the Mo lattice could serve to enhance trapping.

A subtle difference between laboratory and C-Mod experiments is the preparation of the surface before exposure. Typically all samples for ion beam exposure are annealed (e.g. heated to 1273K for 1 hour before exposure [45, 40]). One study [45] showed that there was a factor of ~ 6 lower trap concentration,  $C_W$ , in W annealed at 1273K ( $C_W \sim 1x10^{-5}$  at 700K sample implantation temperature) for one hour in comparison to unannealed W ( $C_W \sim 6x10^{-5}$ ). Annealing at 1673K for one hour led to an additional factor of 2-3 drop in trap concentration in comparison to W annealed at 1273K, or a factor of ~ 12-18 overall drop in trap concentration in comparison to unannealed W samples. A similar difference in retention between annealing temperatures of 1673 and 1173K was observed by Ogorodnikova [40]. C-Mod tiles are typically baked at 500 °C for ~ 24 hours to make sure all oils/impurities from machining and as much water as possible are removed. Thus if the high D/Mo is due to expansion of existing as opposed to creation of new traps the un-annealed Mo in C-Mod should have relatively higher retention than laboratory experiments which generally use annealed materials.

A large difference between a C-Mod tile exposure to ion fluxes and the laboratory experiments is the flux levels. As discussed earlier C-Mod fluxes are generally higher than can be found in the laboratory studies cited (Table 1). One might expect that higher fluxes should lead to higher D pressures within the lattice; for a fixed recombination rate  $n_D$  should scale as  $\Gamma^{0.5}$  (see Eq. 2). Depending on the relationship between D pressure in the lattice and trap expansion or formation, the enhancement with increasing pressure could be large or small. Certainly the surfaces in C-Mod experience a range of fluxes during the density scan data of Figures 2 & 4 without any obvious change in retention normalized to incident ion flux. Several possibilities exist to keep the retention normalized to incident fluence constant at 1%: The trap creation might be proportional to  $n_D^{0.5}$  forcing trap formation proportional to flux; Lower trap formation by D<sup>+</sup> at low fluxes could be compensated by higher trap formation rates by B<sup>+3</sup> because T<sub>e</sub>, and thus the sheath acceleration, would be higher; Or, as assumed by models of trap formation, the trap density formation rate would be exponentially limited at high fluxes[40, 63].

#### 5.4 Inferred location and magnitude of retention

It is likely that the active outer divertor surface is the primary location where D is retained. When the plasma-wetted area is one of the divertors (top or bottom of the chamber) we observe large retention. The high fluxes of plasma to the outer divertor surfaces, as well as some regions of poloidal limiters, leads to strong net erosion of B as is measured with post-campaign surface analysis. When we move the primary plasma-wetted area to thick boron film surfaces (e.g. the inner wall) the retention is low or negligible. Global retention is essentially identical when comparing period 1 diverted discharges (boron-implanted Mo for ALL PFCs) to period 3 discharges where only the divertor surfaces are free of B films. All of this phenomonelogy points to the high flux regions of the outer divertor surfaces as being the primary retention locations, areas that are largely swept clean of any surface B, indicating bulk retention in Mo.

The measurements shown in this paper also have implications for what is occurring within the surface. Post-campaign tile analysis of the first 3 microns of tile surfaces show that D is distributed volumetrically throughout that region at a level D/Mo ~ 1%, i.e. at a concentration much higher than expected for undamaged Mo. This D/Mo level indicates that exposure to the C-Mod plasma has in some manner produced or expanded traps. Since the plasma ions are implanted in the very top monolayers (~ 5 microns) of the Mo surfaces due to their low incident energy, the exposure conditions must also allow for either transport of newly-produced traps or diffusion of solute D deep within the surface and expansion of traps there. The idea of trap diffusion, with levels of trap diffusivity approaching, or exceeding, the D<sup>0</sup> diffusivity, have been observed and modelled elsewhere[64-66, 32, 67]. Deep 'bulk' trapping of D is also supported by the discussion of the D inventory in the outer divertor for the repeated shot sequence (~25 s) of Fig. 6. The retained D must be stored 10s of microns into the Mo to be consistent with a maximum D/Mo ~ 1% concentration.

Disruptive removal of retained D from the top few 100 microns makes interpretation of campaign-integrated D retention difficult. While this is unsatisfactory from the point of view of obtaining a good understanding of the retention mechanisms and locations, the low D retention found from post-campaign analysis of tiles (Fig. 13) is in fact consistent with long-term gas balance including disruptions.

#### 5.5 Long-term retention and implications

It is not clear how the observed large differences between short-term and long-term retention applies to a steady-state or long-pulse discharge (e.g. ITER). Even the sequential set of nondisruptive C-Mod discharges shown in this paper correspond to a fluence less than one ITER pulse at its high flux regions. Simply extrapolating the fraction of ion fluence retained in C-Mod single, non-disruptive discharges (~ 1%) to ITER would be very uncertain and likely incorrect. For example ITER peak D+T fluences after 1000 discharges are ~  $10^7$  higher than the 9 discharge sequence of Figure 17  $(3x10^{23}/m^2)$  [37]. If one scales the retention from C-Mod as  $\Phi^{0.5}$ the retention fraction at the ITER fluences would be 0.0003%. Using a scaling of  $\Phi^{0.75}$  projects to a retention fraction of 0.02%. Of course a linear fluence scaling leads to the 1% retention fraction. Another possibility is that the retention in C-Mod would saturate if we ran enough sequential, non-disruptive, discharges which would have different implications for ITER. The role of ITER's thermally-equilibrated divertor surfaces and intrinsic and extrinsic impurities ( Be, O, C, Ne, Ar) would also be important for hydrogenic retention in ITER. Lastly, we should not forget that disruptions in ITER, even at reduced current, will heat PFC surfaces more than observed in C-Mod due to ITER's higher energy density. That could lead to recovery of large quantites of fuel. The relative time scales of the ITER discharge (sets the depth of diffusion) vs disruptive surface heating (sets the depth of heating) will determine (along with the energy density) how much of the fuel stored in a given ITER discharge will be liberated during a disruption. In summary we need to improve our understanding of the processes involved much more before we can make extrapolations to machines such as ITER. Nevertheless, it is clear such experiments should include in their arsenal of T control the ability to heat the surfaces in steady state, and briefly using disruptions. In the meantime C-Mod disruptions, due to C-Mod's high energy density and small size, are a good vehicle for studying such effects.

# 7. Summary

The retention of deuterium (D) fuel in the Alcator C-Mod tokamak was studied under ITER-like high-flux conditions using a new 'static' gas balance method that we feel improves the accuracy of such studies. The C-Mod plasma facing components are found to retain large fractions, ~20-50%, of the  $D_2$  gas fuelled per quiescent discharge, regardless if the surfaces are cleaned of, or partially covered by, B films. Such high fractions of gas fuel retained are similar to that observed with carbon PFCs where the retention is due to co-deposition of D with C. However, based on C-Mod experiments, we infer that the retention is due to ion implantation into the high-Z surfaces and is primarily occurring in high-flux regions such as the divertor where the films are either nonexistent or thinner than the ion range, allowing the impacting D<sup>+</sup> to reach to the underlying Mo substrate. The B films themselves are deposited saturated with D during boronization and so cannot lead to net co-deposition during a tokamak discharge; If D is liberated through B erosion and then co-deposited elsewhere there is no net D pumping. Only in the case where the B layer is denuded of D by some process can its erosion lead to net co-deposition with D injected during the discharge.

The retention in Mo is roughly proportional to ion fluence to divertor surfaces, with ~1% of incident ions retained, and with no indication of the retention rate saturating over 25 s of plasma exposure. The magnitude of retention is significantly larger than the results of laboratory studies for either Mo or W. The scaling of C-Mod retention with fluence is consistent with the form retention  $\propto \Phi_{D_{+}}^{\alpha}$ , and  $\alpha \sim 0.7$  when compared to laboratory scalings. Values of  $\alpha >0.5$  are consistent with traps being created or expanded.

Normalization of retention to the amount of injected gas is not a good quantity to compare across machines for several reasons. First, for ion implantation leading to retention the proper normaization should be the ion fluence. Second, the wide range of pumping speeds across machines, and even in the same machine with varying pumping geometry (and thus conductance), leads to a varying normalization even when the retention (and ion fluence) stay constant.

Experiments comparing D and He plasmas are consistent with sites ('traps') for fuel retention in

the Mo being created, or expanded, by high D atom densities in the near surface which arise as a result of high incident ion fluxes and limits on the release of D from the surface. Recombination-limited release of D at the surface  $(D \rightarrow D_2)$  is absent for He implantation into Mo. The much higher diffusivity of He in Mo thus leads to much lower He densities in the lattice than for D ion implantation.

The larger retention in C-Mod PFCs than in laboratory studies may be due to a number of factors. C-Mod surfaces experience a flux of multiply-ionized B ions (~ 1% of D+ flux) which can directly create vacancies or traps in the material. Boron on the surface can strongly reduce the  $D_2$  recombination rate, thus elevating the D density in the near surface needed to release neutrals. Boron in the surface could be modifying D transport and providing sites for D nucleation. The use of un-annealed Mo provides additional inherent traps that can be expanded by local high D densities. Finally, the flux densities in the C-Mod divertor, similar to ITER, are 10-100x that in laboratory studies.

Analysis of tiles following a campaign reveals a level of overall retention starkly different than that inferred for a single quiescent discharge. The level of retention is of order 1000x lower. Examination of the effect of disruptions indicates that they are probably the reason for the removal of retained gas over a campaign. Disruptions heat surfaces which leads to enormous enhancements of D diffusivity and detrapping rates, thus freeing D from traps and transporting it to the surface, where it is released. Normally occurring disruptions often lead to no net retention over a single run day.

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