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► **To cite this version:**

F. Waelbroeck, I. Ali-Khan, K. Dietz, P. Wienhold. Surface problems in magnetic confinement systems. Journal de Physique Colloques, 1979, 40 (C7), pp.C7-313-C7-316. 10.1051/jphyscol:19797445 . jpa-00219449

HAL Id: jpa-00219449

<https://hal.science/jpa-00219449v1>

Submitted on 4 Feb 2008

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Surface problems in magnetic confinement systems

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1. **Introduction.** — In the laboratory, plasmas originate both from gas present or introduced into the apparatus and from surrounding surfaces. In cases (material analysis) only wall particles, in others (controlled fusion) only fueled particles are desired. The latter requirement is easily satisfied in fusion schemes based on inertial confinement : the plasma pulse is shorter than the communication time to the walls. It is not stringent in magnetic mirror systems : impurities are poorly confined. But in toroidal reactors, the power producing pulses should be as long as possible and impurities are well confined (according to some theories, better than D^+ and T^+). For many materials, the release of less than 1 % of a monolayer prevents D-T ignition. Thus, as experiments to achieve burning plasmas are being built and planned, plasma-wall phenomena are now studied as attentively as the closely related problems of improved heating and confinement.

The liberation of wall particles can be :

- a) thermal (evaporation, desorption);
- b) current-induced (unipolar arcs, bundles of relativistic electrons);
- c) due to energetic particles (sputtering, back-scattering, ion-induced desorption, blistering), photons (photo desorption) or
- d) to a large concentration c of hydrogen dissolved in surface-near layers.

The first three topics, in particular *b*) and *c*) have been reviewed recently [1]. We concentrate on the fourth, and point out the dominant role which c plays on the :

- impurity production and release by chemical reactions between dissolved hydrogen and surface-near layers of carbides and oxides on steels,
- hydrogen exchange (recycling) between plasma and wall, and associated isotopic effects,
- tritium inventory in walls of devices which shall contain D-T plasmas,
- surface embrittlement effects.

Gaseous H_2 dissolves endothermally as interstitial atoms into SS and inconels, whereas H^0 and H^+ dis-

solve exothermally ; at wall temperature $T_w \leq 600$ °C, evaporation as atoms is negligible.

2. **Concentrations c of thermalized and c_h of non-thermal « hot » H in the wall.** — In a toroidal device with a minor radius of 20 cm, a density of 10^{14} cm^{-3} and a particle confinement time of 10 ms, the flux density of atoms (including Frank-Condon from recycling) which escapes radially to the wall is $\phi_1 \approx 4 \times 10^{16}$ $cm^{-2} s^{-1}$. Less than 10 % of these ($\phi_h \lesssim 4 \times 10^{15}$ $cm^{-2} s^{-1}$) have energies $E > 100$ eV. A fraction α_1 of ϕ_1 penetrates into the lattice. At high [2] and low [3, 4] energies, $\alpha_1 \approx 1$; for 2 eV $< E < 200$ eV, $1 > \alpha_1 \gtrsim 0.3$ values are expected [5]. Each second, $v_1 = \alpha_1 \phi_1$ atoms penetrate; some diffuse deeper into the wall ($v_D = -D \partial c / \partial x$), others return to the surface, recombine and are released ($v_r = 2 \sigma k_r c^2$) as H_2 molecule; σ is the ratio of the surface on which recombination occurs to the geometric surface of the wall, k_r is a phenomenological rate constant. In the quasi-stationary state, when diffusion becomes negligible, $v_1 = v_r$, i.e. ⁽¹⁾ :

$$c = (\alpha_1 \phi_1 / 2 \sigma k_r)^{1/2}. \quad (1)$$

Using the value of $\alpha_1 \phi_1$ given above and for k_r that [7] deduced from a review of available data, we find that at 40 °C, c is 500 times larger than the equilibrium c in H_2 at 1 atm. High c values under atomic exposures have been reported by Kass [8] in SS and measured [4] in iron. Since the rates of many phenomena depend quadratically on c , these unexpectedly high values make it likely that, at first unsuspected effects play dominant roles in the plasma-surface interaction.

During uninterrupted plasma exposures, the thickness of the H-containing layer grows about proportionally to $t^{1/2}$. For intermittent exposures, or alternate discharges in D and H, a computer code (non-linear boundary condition) is used [9].

1 keV D^+ ions and D^0 atoms have an impact velocity of $\approx 3 \times 10^7$ $cm s^{-1}$ and a range of $\lesssim 10^{-5}$ cm. They lose their kinetic energy in a time

⁽¹⁾ If large amounts of H_2O , CH_4 , are produced, eq. (1) must be modified [6].

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$\tau_h < 1$ ps ⁽²⁾ and belong thereafter (unless back-scattered) to the sea of thermalized atoms. The number of *hot* atoms in the wall is $\varphi_h \tau_h < 4 \times 10^3$ cm⁻². Thus even if the phenomena associated with the initial energy and momentum are impressive, the importance of other effects related to c^2 should not be forgotten. Their study has formed the center of the Jülich activities in this field.

3. Chemical reactions. — Pressure measurements and gas analyses in hydrogen, using devices incorporating hot filaments are known to be delicate : some H₂ dissociates into atoms which stick to walls ; H₂O, hydrocarbons, even CO and CO₂ are released from surrounding surfaces. Similar effects occur in toroidal devices under wall bombardment by atomic hydrogen : unless special measures are taken [10, 11, 12], O and C are the dominant plasma impurities and large amounts of the above-listed volatile compounds appear in the exhaust gases [13, 14].

Steel and inconel surfaces are covered by an (usually ≈ 50 Å thick) oxygen and carbon-rich layer [15, 16]. The production rates of methane v_{16} and of water v_{18} are [17] proportional to the concentration of surface carbides and oxides respectively and to c^2 , i.e., in the stationary case, to φ_1 (eq. (1)). Both v_{16} and v_{18} are large during plasma pulses. The gases released are dissociated and ionised by the hot plasma, converted to C and O ions which, during and at the end of the pulse, are redeposited on the wall. Plasma contamination results but little C and O are eliminated during the pulse, the pumping speed of the plasma being much larger than that of the pumps.

As shown in § 4, c does not decrease all the way down to zero at the end of the discharge : some CH₄ and H₂O are also produced between pulses. The methane is evacuated, i.e. the carbide layer is constantly depleted. Some C reappears however on the surface by diffusion from the bulk. When walls are at room temperature the H₂O produced is trapped in surface layers, probably in the form of metal hydroxides. Part of the water formed and trapped between pulses is released during the discharge, increasing the contamination rate ; furthermore only a small fraction of the water produced between discharges is eliminated via the pumping ports, limiting the clean-down rate of the system. A more detailed discussion [17, 18] shows that one method to limit the O-contamination is to decrease the concentration of surface-near metal oxides by careful prehandling of the wall : glow discharges in hydrogen [19] or dense cool tokamak discharges (to maximize φ_1 and minimize dissociation and ionisation of H₂O) should be particularly appropriate ; high T_w 's increase v_{18} and reduce H₂O-trapping effects. Measurements in

TFR 600 [12] have confirmed the rapid reduction of the oxide layer under these conditions.

Another method is to decrease c , for instance by appropriate coatings (large k_r , eq. (1)), catalysing the recombination of atomic hydrogen as it impinges onto the surface. This is probably the cause of the beneficial influence of titanium.

4. Hydrogen recycling. — A discussion of the steps involved in the hydrogen exchange at surfaces shows that the simple expressions for the rates of penetration v_1 , diffusion v_D and release v_r as defined in § 2 are applicable when surface coverage effects are negligible, provided that :

- H₂ physisorption and desorption is rapid and
- atomic H exchange between surface and lattice sites is fast.

The hydrogen recycling phenomenon ⁽³⁾ can then be described in zero'th order approximation by very simple equations [9].

During discharges, $v_1 - v_r = -D \partial c / \partial x$. In the first phase c and hence v_r are small : impinging atoms are *absorbed* into the wall wherein they diffuse rapidly ($\partial c / \partial x$ is very large). As the discharge proceeds, c and the recycling coefficient v_r / v_1 increase ; most impinging atoms are released back. The concentration gradient points outwards, towards the surface where c is maximum. The hydrogen which does not recombine on the surface diffuses inwards.

Between discharges, $v_1 = 0$ ($v_r = D \partial c / \partial x$) ; the concentration gradient points inwards (H diffuses towards the surface) ; the concentration at the surface ($c_0 \neq 0$) ensures the H₂ outgasing rate. This explains (§ 3) the production of CH₄ and H₂O between discharges.

After a number of discharges in H₂, a considerable amount of H is dissolved in the wall. If a discharge in deuterium follows, the impinging D either diffuses into the bulk or recombines. Since $(c_{HD}) \neq 0$ at the surface, both D₂ and HD formed. H appears into the plasma at a rate

$$v_r' = 2 \sigma k_r c_H^2 + 2 \sigma k_r' c_H c_D,$$

which is larger than expected from the outgasing rate. Many recycling events occur during one discharge so that the amount of H in the plasma at the end of the pulse is large : in Dite [20], the ratio H/(H + D) at the end of such a pulse was ≈ 65 %. The evolution of this isotopic exchange during consecutive discharges, including the effect of changing the time between

⁽²⁾ The time during which H *knows* that it was initially ionic or neutral is shorter.

⁽³⁾ We discuss recycling on deoxidized surfaces : only H₂ returns to the plasma. If the surface is oxidized, H₂O is also released. Recycling in tokamaks is derived from the density of electrons : (not protons) . O atoms, source of numerous electrons, make the interpretation of recycling in poorly cleaned-down tokamaks difficult.

discharge (readjustment of the D and H profiles in the solid), is easily understood by this model.

To explain the recycling phenomenon, it has previously been postulated [1] that hydrogen *diffuses* (not recombines) out of the lattice. The concentration gradient points then inwards, even during discharges, and c at the surface remains negligible (⁴). The interpretation of experimental results is then intricate : trapping centers close to the surface, from which hydrogen cannot be thermally released have to be assumed, as must be an ion-induced detrapping mechanism which specifically empties hydrogen-filled trapping centers.

To check our model, and to measure σk_r , the system of equations was first extended self-consistently to the case of the solubilisation of H starting from H₂ at a pressure p_{H_2} . A fourth rate, $v_2 = 2 \sigma k_s p_{H_2}$ must be included ; k_s is a second phenomenological rate constant, related to k_r and to the solubility constant K_s ($k_s = k_r K_s^2$) by equilibrium consideration. Degasing experiments, permeation experiments both stationary and nonstationary, using H₂ alone or mixtures of H₂ and H⁰ upstreams, have been carried out. The parameter domain was varied from the purely diffusion-limited regime, via that where diffusion and surface effects play equivalent roles, to the one where pure surface effects dominate (see e.g. [4]). Seven independent methods of measuring σk_r resulted. Good agreement was found between them.

5. Surface embrittlement. — An *internal* pressure p^* within a metal exposed to atomic H particles can be defined by combining eq. (1) with the solubility law $c = K_s p^{1/2}$:

$$p^* = \alpha_1 \varphi_1 / 2 \sigma k_r K_s^2 ; \quad (2)$$

p^* builds up at internal surfaces (grain boundaries, lattice defects) of the wall. For the example given, $p^* \approx 3 \times 10^5$ atm. In present-day devices, p^* is lower because of the small duty cycle ($\leq 1\%$), but still

(⁴) Pushing this postulate *ad absurdum* leads to positive gradients and to a negligible c value also when metals are charged with hydrogen in an H₂ atmosphere.

high enough for surface cracking and embrittlement effects to be feared. These have been observed in simulation experiments in the form of irregular gas release (bursts) [17], increase of the surface hardness of probes exposed to H-atoms, alterations of the surface topography [20, 21]. In addition, the increase of c according to eq. (1) has been directly measured [4] in our permeation experiment.

Embrittlement effects are expected to affect the contamination in two ways : the sudden localised release of H₂ at high pressure offers a favourable condition for the occurrence of unipolar arcs [22, 23, 24]. The opening of cracks is moreover probably accompanied by the ejection of metallic dust.

6. Conclusions : role of T_w and of surface layers. —

This brief discussion brings forth the following points :

— A number of impurity release mechanisms depend on c , the concentration of H dissolved into the surface layers. Of these :

— the release of H₂O, CH₄ can be controlled by appropriate surface prehandling (reduction of carbides and oxides) or by coatings (reduction of c). T_w is important (v_{18} has an activation energy of 20.3 kcal [6]).

— Surface embrittlement can be avoided by raising the wall temperatures ; this increases k_r and K_s , i.e. decreases p^* (eq. (2)). Appropriate coatings catalysing the surface recombination of H, again play a beneficial role [21].

— Because they decrease c which otherwise rises to high values, thin coatings, can depress the tritium inventory in walls of devices such as JET, TFTR when they start operation with DT mixtures.

— In order to discuss quantitatively the results obtained in confinement experiments, more information should be given by the tokamak physicists on the status of the wall during the discharge : liner contamination by limiter material or vice versa, T_w , state of oxidation, of carburisation, surface roughness. These informations are of but moderate importance as long as only points a)-c) of the introduction are discussed. They become crucial when phenomena associated to point d) are envisaged.

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