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Surface problems in magnetic confinement systems

F. Waelbroeck, I. Ali-Khan, K. J. Dietz (*) and P. Wienhold
Institut für Plasmaphysik der Kernforschungsanlage Jülich GmbH,
Association EURATOM-KFA, 5170 Jülich, Postfach 1913, FRG

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 H₂ dissolves endothermally as interstitial atoms into SS and inconels, whereas H₀ and H+ dissolve exothermally; at wall temperature $T_w \leq 600 \, ^\circ C$, evaporation as atoms is negligible.

2. Concentrations c of thermalized and $c_n$ of non-thermal «hot» H in the wall. — In a toroidal device with a minor radius of 20 cm, a density of $10^{14} \, \text{cm}^{-3}$ 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 $\phi_1 \approx 4 \times 10^{16} \, \text{cm}^{-2} \, \text{s}^{-1}$. Less than 10 % of these ($\phi_n \leq 4 \times 10^{15} \, \text{cm}^{-2} \, \text{s}^{-1}$) have energies $E > 100 \, \text{eV}$. A fraction $\alpha_i$ of $\phi_1$ penetrates into the lattice. At high [2] and low [3, 4] energies, $\alpha_i \approx 1$; for $2 \, \text{eV} < E < 200 \, \text{eV}$, $1 > \alpha_i > 0.3$ values are expected [5]. Each second, $v_i = \alpha_i \phi_1$ atoms penetrate; some diffuse deeper into the wall ($v_d = -D \, \partial c / \partial x$), others return to the surface, recombine and are released ($v_r = 2 \, \sigma k, c^2$) as H₂ molecule; $\sigma$ is the ratio of the surface on which recombination occurs to the geometric surface of the wall, $k$, is a phenomenological rate constant. In the quasi-stationary state, when diffusion becomes negligible, $v_1 = v_r$, i.e. (1):

$$c = (\alpha_i \phi_1 / 2 \, \sigma k, c^2)^{1/2}.$$  

(1)

Using the value of $\alpha_i \phi_1$ given above and for $k_7$ 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 H₂ 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 D⁰ atoms have an impact velocity of $\approx 3 \times 10^7 \, \text{cm} \, \text{s}^{-1}$ and a range of $\approx 10^{-5} \, \text{cm}$. They lose their kinetic energy in a time

(*) JET Project, Abingdon, UK.
3. Chemical reactions. — Pressure measurements and gas analyses in hydrogen, using devices incorporating hot filaments are known to be delicate: some H$_2$ dissociates into atoms which stick to walls; H$_2$O, hydrocarbons, even CO and CO$_2$ 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 carbidies and oxides respectively and to $c^2$, i.e., in the stationary case, to $v_1$ (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 pulse, are redeposited on the wall. Plasma contamination rate is eliminated via the pumping ports, limiting the contamination rate.

As shown in § 4, $c$ does not decrease at all the way down to zero at the end of the discharge: some CH$_4$ and H$_2$O 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 H$_2$O 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 $\rho_1$ and minimize dissociation and ionisation of H$_2$O) should be particularly appropriate; high $T_\nu$'s increase $v_{18}$ and reduce H$_2$O-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 $v_0$ and release $v_c$ as defined in § 2 are applicable when surface coverage effects are negligible, provided that:

- H$_2$ physisorption and desorption is rapid and
- atomic H exchange between surface and lattice sites is fast.

The hydrogen recycling phenomenon (2) can then be described in zeroth order approximation by very simple equations [9].

During discharges, $v_1 - v_c = -D \frac{\partial c}{\partial x}$. In the first phase $c$ and hence $v_c$ 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 $v_c/v_1$ 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, $v_1 = 0$ ($v_c = D \frac{\partial c}{\partial x}$); the concentration gradient points inwards (H diffuses towards the surface); the concentration at the surface ($c_0 \neq 0$) ensures the H$_2$ outgasing rate. This explains (§ 3) the production of CH$_4$ and H$_2$O between discharges.

After a number of discharges in H$_2$, 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 ($c_0 \neq 0$) at the surface, both D$_2$ and HD formed. H appears into the plasma at a rate

$$v'_c = 2 \frac{\sigma}{k} c_R + 2 \frac{\sigma}{k} c_H c_D,$$

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) We discuss recycling on deoxidized surfaces: only H$_2$ returns to the plasma. If the surface is oxidized, H$_2$O 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 (*) . 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 $\sigma k$, the system of equations was first extended self-consistently to the case of the solubilisation of H starting from $H_2$ at a pressure $p_{H_2}$. A fourth rate, $v_2 = 2 \sigma k p_{H_2}$ must be included; $k_s$ is a second phenomenological rate constant, related to $k$, and to the solubility constant $K_s (k = k, K^2)$ by equilibrium consideration. Degassing experiments, permeation experiments both stationary and nonstationary, using $H_2$ alone or mixtures of $H_2$ and $H^0$ 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 $\sigma k$, 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 $c = K_s p^{\frac{1}{2}}$:

$$p^* = \alpha\frac{1}{2} \sigma k, K_s^2;$$

$p^*$ builds up at internal surfaces (grain boundaries, lattice defects) of the wall. For the example given, $p^* \approx 3 \times 10^5 \text{ atm}$. In present-day devices, $p^*$ is lower because of the small duty cycle ($\leq 1\%$), but still 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 $H_2$ 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 $H_2O$, $CH_4$ can be controlled by appropriate surface prehandling (reduction of carbides and oxides) or by coatings (reduction of $c$). $T_w$ is important ($\gamma_1 = 9$ has an activation energy of 20.3 kcal [6]).

— Surface embrittlement can be avoided by raising the wall temperatures: this increases $k$, and $K$, 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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