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STUDIES OF CHROMIUM GETTERING

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ABSTRACT

Preliminary results have shown that hydrogen pumping by chromium is a surface effect. Unlike with titanium, the getter material used in many present day tokamaks, there is no significant diffusion into the bulk. This feature, which would minimize the tritium inventory, makes chromium a viable alternate to titanium gettering for future tokamaks.

Additional experiments have been carried out to measure the basic characteristics of chromium films for gases of interest in tokamak research. These gases include deuterium, oxygen and nitrogen. A vacuum system is described which allowed precise control of the test gas, a constant wall temperature and determination of the projected getter surface area. A quadrupole mass spectrometer, rather than simply a total pressure gauge, was utilized to measure the partial pressure of the test gas as well as the residual gas composition in the system. A quartz crystal monitor was used to measure film thickness.

Pumping speeds and sticking coefficients are given as a function of surface coverage for each test gas. A comparison will be made with titanium films deposited in the same vacuum system and under similar conditions.

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Introduction

Titanium gettering has been used in many present-day tokamak devices as a method for improving plasma parameters.[1,2,3,4,5] Observed improvements include the lowering of Z_{eff} to near unity and increased confinement time.[4,6] In this process, an active titanium film is deposited over a portion of the torus between tokamak discharges.

Titanium has been used as the getter material in tokamaks because its gettering properties are well-known and suitable titanium sources are commercially available. Although titanium gettering has been beneficial, there are some potential problems with its long term use. Since titanium has a large pumping capacity for hydrogen, a large quantity of hydrogen may become trapped in the walls of the torus. This may lead to embrittlement and flaking.[7] Furthermore, future tokamak devices, including fusion reactors, operating on a deuterium-tritium (DT) fuel cycle cannot tolorate this large tritium inventory.

Chromium gettering provides a promising alternate to titanium gettering. While titanium pumps large amounts of hydrogen, the hydrogen pumping capacity of chromium is only about one monolayer adsorbed on the surface.[8] A recent tokamak experiment showed that hydrogen recycling increases markedly with chromium gettering.[9]

Although the behavior of titanium getter films has been carefully studied and commercially produced titanium getter pumps are widely used, there is insufficient available data on chromium to allow a comparison of the predicted behavior. Hence, the purpose of this experiment is to investigate the getter pumping capabilities of chromium for gases of interest in fusion research. Titanium gettering was also investigated utilizing the same experimental apparatus and technique in order to compare the behavior of chromium and titanium as getters under similar experimental conditions.

The necessary data base for gettering is given in terms of the sticking coefficient as a function of surface coverage. Therefore, measurements of the pumping speed of chromium and titanium getter films were made for the various test gases as a function of surface coverage. These gases include: (1) deuterium, since this is the working gas of most tokamak experiments, (2) oxygen, because it is the most abundant gaseous impurity in tokamaks, and (3) nitrogen, to establish a baseline. In order to make meaningful measurements several parameters must be controlled:

(1) Substrate temperature.

(2) The flux of gas molecules to the getter surface.

(3) Getter surface area.

Additionally, vacuum conditions and surface cleanliness play very important roles in determining the value of the sticking coefficient.

Experimental Apparatus

A schematic of the experimental apparatus is shown in Figure (1). The system consisted of two parts: the UHV main experimental chamber and the working gas reservoir. The gas leak valve (6), connected the two parts. The chamber was baked at 250°C for 48 hours following pump-down from atmospheric pressure and a base pressure of 4×10^{-9} Torr was achieved by the 60 ℓ/s ion pump before gettering. The volume of the chamber was 7.8 liters. In order to maintain a constant substrate temperature a section of the chamber was equipped with cooling coils which provided a constant temperature of 10°C on the external surface. The power flux to the inner walls during deposition of the getter films could produce a 1°C maximum temperature gradient across the walls.

Deposition of the getter films was provided by sublimation from commercially available resistance heated filaments: chromium on tungsten and a titanium-molybdenum alloy. A system of baffles defined the gettered area.

Film thickness was monitored by a water cooled quartz crystal oscillator with a resolution of approximately 5 monolayers. The deposited films had an approximate thickness of 30 monolayers.

Gas analysis was provided by utilizing a Balzers QMG 511 quadrupole mass spectrometer (RGA). This was used for simultaneously monitoring both the residual gas composition and the partial pressure of the test gas. Calibrations of the RGA were made for all test gases.

The test gas was admitted to the system through a controlled leak from the test gas reservoir which consisted of the volume bounded by the gate valve (6), the piezo-electric valve (11) and the orifice (9) which limited the pumping speed of the turbo pump to 1 ℓ/s. The pressure in the gas reservoir was controlled by a Veeco APC-100 pressure controller which utilized a feedback system to control the piezo-electric valve, thus regulating the flow of gas into the reservoir. The leak rate into the main chamber was controlled by molecular flow through the orifice (12) having a conductance of ∿ 0.6 ℓ/s. By controlling the gas pressure in the reservoir, the leak rate into the main chamber could be preset, then upon opening the solenoid operated gate valve, the appropriate leak was established in the main chamber. Tests showed that the leak rate remained constant to within 0.1% for at least 10 hours. The leak rate for this study was constant at 5.0×10^{-5} Torr ℓ/s, except for H_2 on titanium.

Experimental Procedure

After outgassing the filaments at reduced power levels to avoid sublimation, the current was increased and the deposition rate was monitored by the quartz crystal film thickness monitor. Typically, 3-5 minutes were required to deposit about 30 monolayers. After the deposition was complete, the leak rate, $Q_{(leak)}$ was established by opening the gate valve to the gas reservoir.

The pumping speed of the getter film was determined from the observed rate of increase in the partial pressure, p, of the test gas, as measured by the RGA. The ion pump functioned as an auxiliary pump, thus avoiding the accumulation of impurities which were not pumped by the getter film. Thus,

$$(1)\quad p = \frac{Q_{(leak)}}{S_{(get)} + S_{(aux)}}$$

Where $S_{(get)}$ is the pumping speed of the getter film and $S_{(aux)}$ is the pumping speed of the auxiliary pump.

The rate of gas adsorption $Q_{(get)}$, effective sticking coefficient α, particle flux to the getter film Γ, and the gettered area A are related by:

$$(2)\quad Q_{(get)}(t) = p(t)\ S_{(get)}(t) = \alpha(t)\ \Gamma(t)A$$

and

$$(3)\quad \Gamma = \frac{n\bar{v}}{4} = \frac{p(t)}{\sqrt{2\pi mkT}}$$

Where n is density, v is velocity, m is the ion mass, k is Boltzmann's constant, T is temperature in °K, and p is the pressure in Torr. With Equations (1), (2), and (3) we can express the sticking coefficient $\alpha(t)$ as a function of the measured parameters $p(t)$, $Q_{(leak)}$, and A:

$$(4)\quad \alpha(t) = \frac{3.5 \times 10^{19} \left(Q_{(leak)} - S_{(aux)}\ \ p(t)\right)\sqrt{2\pi mkT}}{A\ p(t)}$$

Finally, we want to express α as a function of the surface coverage θ which is the ratio of the total number of adsorbed particles to the adsorbing surface,

$$(5)\quad \theta(t) = \frac{Q_{(leak)}t - S_{(aux)} \int_0^t p(t)\ dt}{A}$$

$\alpha(t)$ and $\theta(t)$ were evaluated by numerical methods from Equations (4) and (5).

The pumping speed of the auxiliary ion pump did not remain constant throughout the course of an experiment. Measurements at the end of each experimental run with each test gas at equilibrium pressure showed variations of as much as 50%. This measured value was used in the data analysis, although the error would have been insignificant except when the getter film approached saturation and $S_{(get)}$ approached $S_{(aux)}$.

Results

In order to measure sticking coefficients and surface coverage, the gettered area must be known. Because of the microscopic roughness of the substrate and getter film, a distinction must be made between the geometric surface area and the true surface area. For example, measured ratios between true surface and measured surface for nickel have been reported to vary by more than an order of magnitude; a typical value for new rolled nickel is 5.8.[10] A value for the true gettered surface to the geometric surface was determined in this study by assuming a value of unity for the initial sticking coefficient, α_o, of oxygen on titanium as shown in Figure 2. A similar value of α_o for O_2 on titanium has been measured by others.[11] With this normalization the surface coverage of N_2 at saturation is close to the expected theoretical value of 7×10^{14} molecules per cm^2. We assumed that, unlike H_2 and O_2, N_2 is adsorbed only up to one monolayer. In this sense nitrogen served to establish a baseline to provide an estimate of the actual surface area. The sticking coefficient of O_2 remains very high until the surface coverage approaches one monolayer. Actually, in the growth of an oxide film on a clean surface at room temperature, more than one monolayer may be adsorbed.[12,13] The value of the initial sticking coefficient for H_2 on titanium was influenced by the relatively high residual pressure (3×10^{-8} Torr) during the titanium deposition, but agrees with published values measured under similar conditions.[11] We do not understand the difference between the behavior of H_2 and D_2 on titanium shown in Figure 2; it may be due to the difference between adsorption and diffusion. A somewhat similar behavior for D_2 on titanium has been reported elsewhere.[14]

Results for the sticking coefficients of O_2, N_2, and D_2 on chromium are shown in Figure 3 as functions of the surface coverage. As expected, the highly active oxygen has an initial sticking coefficient in excess of 0.9, and remains high until a surface coverage of approximately one monolayer is approached. The value of the initial sticking coefficient for D_2 was probably influenced by the higher pressure (1×10^{-8} Torr) during sublimation. Subsequent Auger analysis of this filament indicated strong impurity (O_2) contamination on its surface.

From these results, we find that chromium gettering of deuterium, nitrogen, and oxygen on chromium is strictly a surface effect.

Conclusions

Because of differing experimental conditions, it is difficult to compare quantitatively the results of different gettering experiments. However, our results on titanium gettering agrees generally with those presented by Gupta and Leck.[11] Our results also indicate that the gettering properties of titanium and chromium are very similar for O_2 and N_2 and seem to be controlled by surface adsorption. Finally, as a main objective of these studies, we have shown that the getter pumping of D_2 by chromium is also restricted to surface adsorption, unlike with titanium where D_2 is adsorbed on the surface and subsequently diffuses into the bulk. These properties make chromium an attractive candidate for use as a getter pump in present-day tokamaks and even more attractive for use in future devices which will be tritium fueled. Chromium sublimation sources suitable for application in a tokamak are presently being fabricated in preparation for a test in the ISX-B device at the Oak Ridge National Laboratory.

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Figure Captions

Figure 1. Schematic diagram of the experimental apparatus; (1) 60 ℓ/s ion pump, (2) quadrupole mass spectrometer, (3) baffles to define getter area, (4) film thickness monitor, (5) cooling coils, (6) solenoid operated gate valves, (7) Schultz-Phelps gauge, (8) thermocouple gauge, (9) orifice to conductance limit turbo pump, (10) test gas supply, (11) piezo-electric valve, (12) orifice for controlled leak, (13) getter filament, (14) sorption pump, and (15) ion guage.

Figure 2. Sticking coefficients as functions of titanium surface coverage for D_2, H_2, N_2, and O_2.

Figure 3. Sticking coefficients as functions of chromium surface coverage for D_2, N_2, and O_2.

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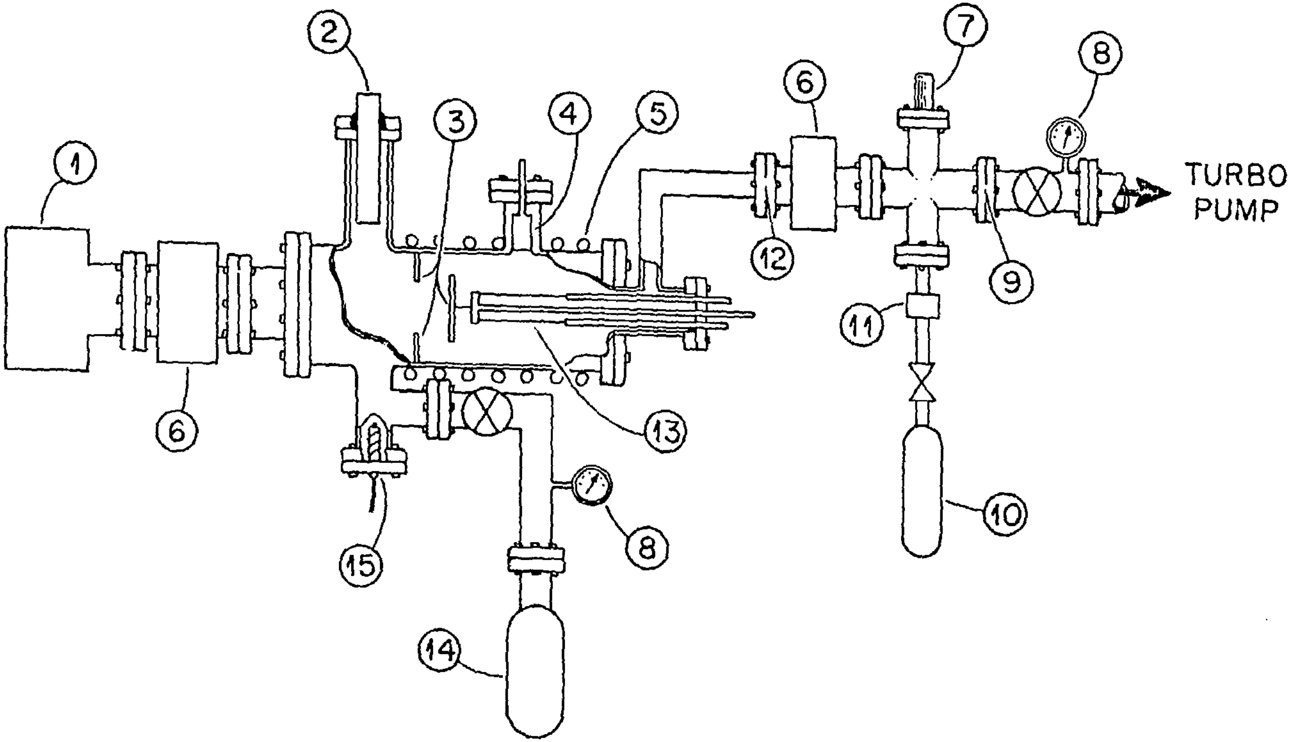

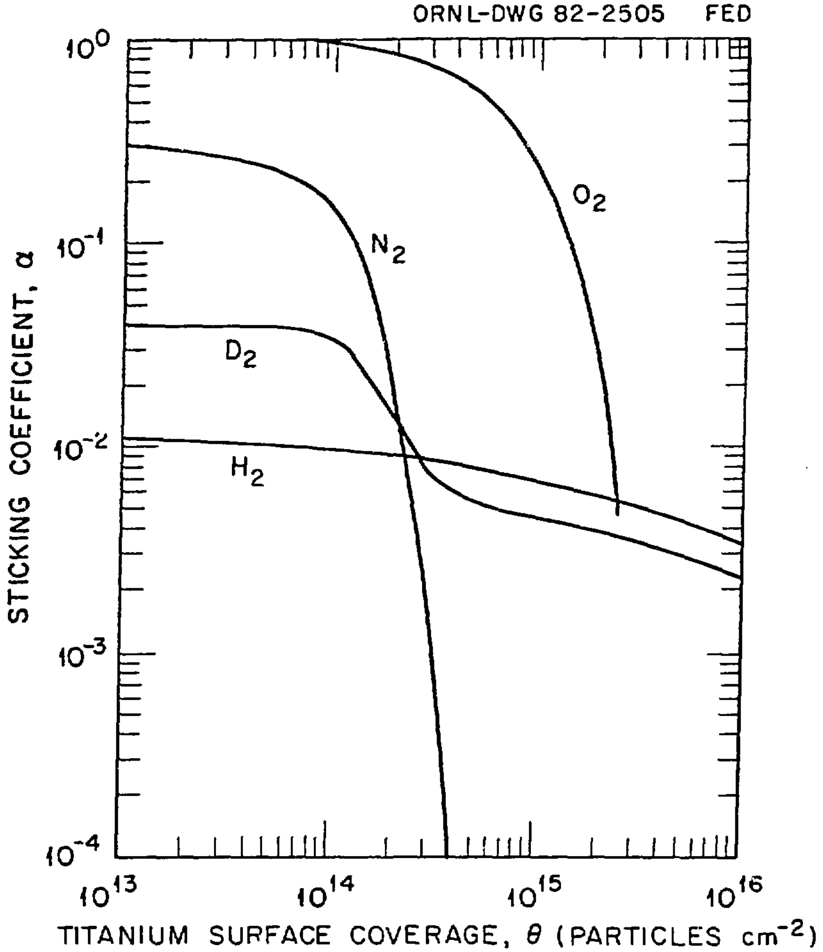
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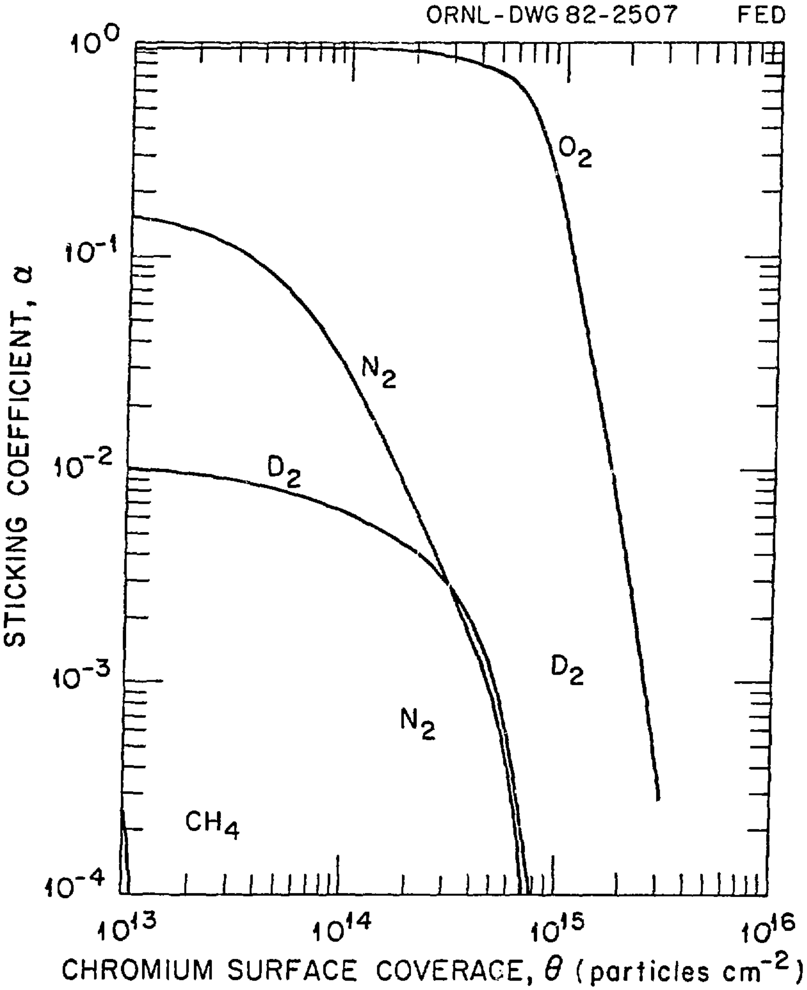
10^0
10^{-1}
10^{-2}
10^{-3}
10^{-4}
STICKING COEFFICIENT, α
O_2
N_2
D_2
H_2
10^{13}
10^{14}
10^{15}
10^{16}
TITANIUM SURFACE COVERAGE, θ (PARTICLES cm^{-2})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10^0
10^{-1}
10^{-2}
10^{-3}
10^{-4}
STICKING COEFFICIENT, α
O_2
N_2
D_2
D_2
N_2
CH_4
10^{13}
10^{14}
10^{15}
10^{16}
CHROMIUM SURFACE COVERAGE, θ (particles cm^{-2})