



A study of the photodesorption process for cryosorbed layers of H₂, CH₄, CO or CO₂ at various temperatures between 3 and 68 K

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Abstract

The vacuum performance of the large hadron collider (LHC) at CERN will depend critically on the photodesorption of gas by synchrotron radiation and the re-adsorption of these molecules back onto the cold surface. The results of photon-induced molecular desorption by synchrotron radiation with a critical photon energy of 284 eV for H₂, CH₄, CO or CO₂ cryosorbed on a stainless steel surface are presented. Most measurements have been carried out in a temperature range from about 3 to 20 K. Measurements for CO₂ were also performed at 68 K. The specific method used for this study has been to pre-deposit a known quantity of gas onto a cold surface, to irradiate the surface with a known photon dose and to measure the quantity of gas remaining on the cold surface by recording the pressure during warm-up. The average photodesorption yields of all gas species were found to increase with increasing surface coverage and to reach a saturation value. For H₂ this value is approximately 0.5 for a coverage exceeding 10¹⁷ molecules/cm², for CH₄ and CO₂ saturation occurs at about 0.5 molecules/photon at ~10¹⁹ molecules/cm², while the corresponding final value for CO is about 0.04 molecules/photon at this coverage. © 1999 Elsevier Science Ltd. All rights reserved.

1. Introduction

In a previous publication [1] a study was made of the photodesorption of CO₂ condensed at about 68 K. However, since the LHC beam screen will operate between 5 and 20 K it is necessary to know the photodesorption yields of gases such as CH₄, CO, and CO₂ which may be condensed in thick layers at those temperatures. In addition, hydrogen may be condensed in thick layers on the magnet cold bore operating at 1.9 K and give rise to a large desorption yield in specific locations without a beam screen. Therefore, to study these condensed gas species the “open geometry” experimental installation described previously for measurements at room temperature and at 68 K was modified to extend the operation to temperatures from 3 to 20 K.

2. Experimental method

The experiment was installed on the low intensity synchrotron radiation beam line [2] on the VEPP-2M

storage ring at the Budker Institute of Nuclear Physics in Novosibirsk, Russia. The photon beam was collimated to a square section 30 × 30 mm² and the incident photon flux during the experiment, illuminating the sample at perpendicular incidence, was typically in the range of 10¹⁵ photons/s with a critical photon energy in the range of 250–300 eV.

Since temperatures of interest in this experiment ranged from 3 to 20 K the set-up was modified as shown in Fig. 1. To make this large temperature excursion possible, the substrate (S) consisting of a 30 mm diameter, 1 mm thick stainless steel plate, was brazed to a thick OFHC Cu cylinder (C) which had both, a heater (H) and a thermometer (Th) incorporated into it. To keep helium consumption low even during the frequent periods when the substrate had to be heated to room temperature, the Cu cylinder was mounted on a stainless steel support (ST) to which a strip of high purity indium (In) was attached. In this way, a variable thermal resistance between the helium bath and the substrate could be obtained as high purity indium has in this respect the very useful property that its thermal conductivity at room temperature is about one order of magnitude less than at liquid helium temperature. Finally, to obtain a small and well-defined measurement volume ($V \approx 0.11$) and to

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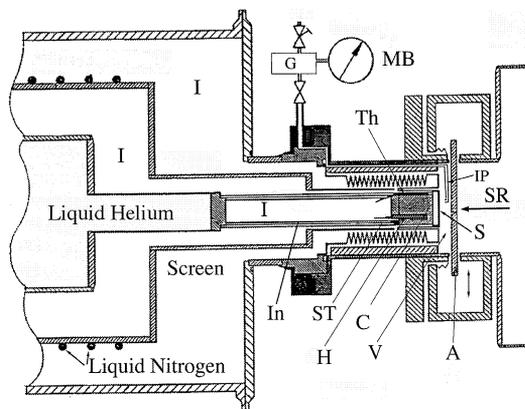


Fig. 1. Experimental set-up at the synchrotron radiation beam line (SR) with the vacuum valve (A) shown in closed position. A detailed description of the system is given in the text.

minimise the heat in-flow from the room temperature parts, a long and narrow thermal leakage path was created by two tightly fitting coaxial pipes and a set of bellows which were welded to the stainless steel substrate on one end and to the room temperature flange on the other end. These elements had the additional function of separating the insulation vacuum (I) from the vacuum in the measurement volume. To reduce thermal losses the helium bath was surrounded by a liquid nitrogen cooled screen. To initiate a measurement, the calibrated volume ($G = 1$ l) was filled at room temperature with the respective gas H_2 , CH_4 , CO or CO_2 to a pressure, measured with the membrane gauge (MB), and this gas was then injected through the injection pipe (IP) to be condensed onto the cold substrate (S). The temperature of the substrate was measured with a thermometer (Th) and controlled by electric heaters (H) incorporated into the copper block (C). While temperatures between 5.5 and 20 K were obtained through thermal contact with the liquid helium bath via the previously described supporting tube with the attached indium strip (ST), the reduced temperature of 3 K was achieved through direct thermal contact of the substrate with the sub-cooled liquid helium bath. This modification is not shown in Fig. 1.

For the desorption measurement, the all-metal valve was opened and the substrate, with its known coverage of condensed gas, exposed to photons. The evolution of the desorbed molecular species in the experimental chamber was measured using a quadrupole residual gas analyser. To avoid any significant amount of re-pumping of desorbed gas back onto the cold substrate, the external pumping speed was considerably larger than the intrinsic pumping speed of the cold surface – approximately 40 l/s at room temperature for CO. For comparison, the external pumping speed which was provided in this set-up by a combination of sputter-ion and Ti-sublimation pumps and connected to the experimental chamber by

a 80 mm long 100 mm diameter duct corresponded to about 1000 l/s for CO. At the end of the measurement the all-metal valve was closed. The pressure in the combined volume $V + G$ during the warm-up yields the condensed gas remaining on the substrate.

The basic result which can be obtained through this experiment is the average removal coefficient $\langle \eta_r \rangle$ for cryosorbed molecules, i.e. the quantity of cryosorbed gas removed from the cold substrate during a known exposure to photons. For this purpose three quantities have to be measured: the initial amount of gas condensed on the substrate, Q_i (molecules), the accumulated photon dose, Γ (photons) and the amount of gas evaporated from the substrate during warm-up after the irradiation, Q_f (molecules). Both quantities, Q_i and Q_f are determined in a straight forward way from the initial pressure in volume G and the final pressure in volume $G + V$ after the irradiation while the system is at room temperature.

Thus, the average removal coefficient $\langle \eta_r \rangle$ (molecules/photon) is given by

$$\langle \eta_r \rangle = (Q_i - Q_f)/\Gamma.$$

The average surface coverage of condensed gas $\langle s \rangle$ (molecules/cm²) for each measurement is defined as: $\langle s \rangle = (Q_i + Q_f)/(2A)$, where A is the irradiated surface area. By repeating this measurement for different initial quantities of condensed gas, the dependence of the average removal coefficient on the average surface coverage was obtained for each gas species.

The term average removal coefficient $\langle \eta_r \rangle$ has been used in this work to underline the basic approach of this procedure which measures the removal of condensed gas by a given photon irradiation rather than the molecular desorption yield (molecules/photon) through the increase in pressure. It has been discussed in detail in a previous study [3], that the determination of the molecular desorption yield from the pressure increase due to photon stimulated desorption in a vacuum system with cryosorbing walls requires the knowledge of the pumping speed including the usually unknown sticking probability on the wall.

As an additional complication of such a measurement, some of the condensed gas species may be cracked into their atomic constituents and form new molecular species before being desorbed into the gas phase. Hence, the quantitative interpretation of the pressure in terms of the desorption yield of individual molecular species requires a very extensive analysis.

3. Results

A summary of the results of this study is shown in Fig. 2. To facilitate the comparison with previous measurements with a “closed geometry”, the H_2 data

[3, 4] have been converted into the corresponding recycling coefficient assuming a sticking probability of 0.1. During several of the measurements with CH_4 , CO and CO_2 , the substrate temperature was varied in the range from 5.5 to 20 K to see whether a temperature dependence of the recycling yield can be observed. However, since in all these cases no significant change of the system pressure with temperature was found, it may be concluded that the measured recycling yield has no strong dependence on the substrate temperature.

3.1. H_2

The measurements with H_2 were done, as indicated before, using a modified set-up with respect to Fig. 1 with the sample in direct thermal contact with the sub-cooled helium bath. In the range of coverage studied, 4×10^{16} – 5×10^{18} molecules/ cm^2 , the average removal coefficient for cryosorbed H_2 was found to have a constant value of about 0.5 molecules/photon. The data points derived from Refs. [2, 3] assuming a sticking probability of 0.1 were obtained for lower surface coverage and agree quite closely with the new set of data as may be seen from the overlap of the two measurements at about 5×10^{16} molecules/ cm^2 .

3.2. CH_4

The average removal coefficient for cryosorbed CH_4 at about 15 K is seen to increase with surface coverage and to reach about 0.4 molecules/photon at a surface cover-

age of 10^{19} molecules/ cm^2 . No significant difference was observed when changing the temperature from 5.5 to 20 K.

In a measurement without pre-condensed CH_4 on the substrate, the quantity of gas released during the warm up following the irradiation was approximately 4×10^{-3} – 1.2×10^{-2} mbar l. From this result one can derive a lower limit for the determination of the average surface coverage between 1 and 3×10^{16} molecules/ cm^2 as well as a lower limit for the average removal coefficient for CH_4 between 5×10^{-4} and 1.5×10^{-3} molecules/photon.

It was also observed that the H_2 partial pressure during photon irradiation with condensed CH_4 increased slightly as compared to the bare substrate. This effect may be attributed to cracking of CH_4 .

3.3. CO

The average removal coefficient for cryosorbed CO was found to increase with surface coverage as shown in Fig. 2 and to reach a value of about 0.04 molecules/photon at a surface coverage of 1×10^{19} molecules/ cm^2 . Again, no variation of the removal coefficient with temperature was observed when changing the temperature between 5.5 and 15 K.

In an independent experiment using the substrate without pre-condensed CO , the quantity of gas released during the warm up following an irradiation was measured to be between 5 and 8×10^{-3} mbar l. From this result a lower limit for the determination of the surface coverage of

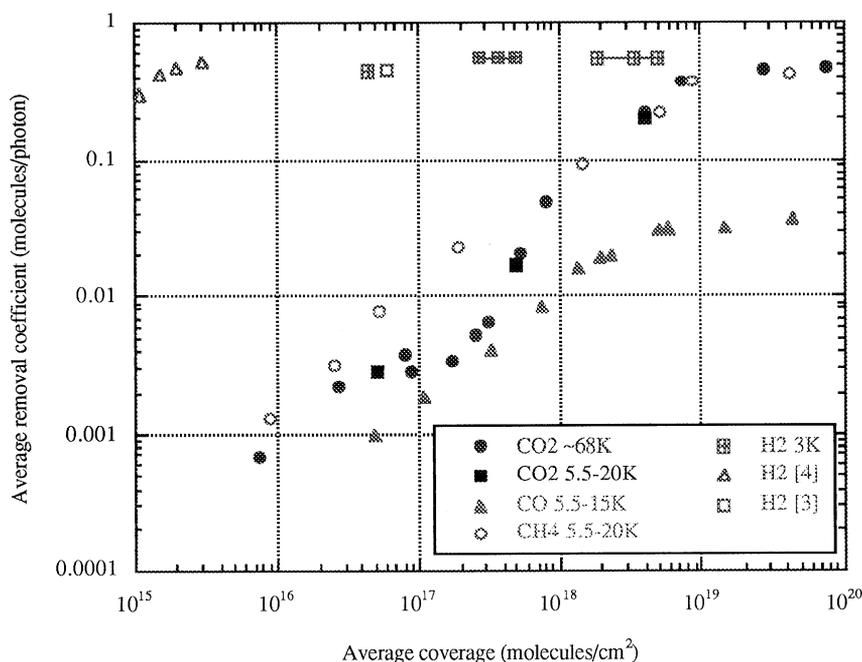


Fig. 2. Average removal coefficient as a function of the average surface coverage. Data for H_2 from Refs. [3, 4] have been included in this figure.

about $1\text{--}1.5 \times 10^{16}$ molecules/cm² and a lower limit for the average removal coefficient for CO between 7×10^{-4} and 1×10^{-3} molecules/photon can be derived.

During the desorption of the condensed CO only two peaks were observed in the residual gas spectrum corresponding to mass 2 and 28, respectively. While the signal for mass 28 increased with CO surface coverage, the signal for mass 2 remained the same as for the bare substrate without pre-condensed CO suggesting that the origin of H₂ is desorption from the underlying substrate.

3.4. CO₂

Three sets of measurements with cryosorbed CO₂ were made, one at ~ 15 K and two at approx. 5.5 K. These data have been presented together with earlier results at 68 K [1] in Fig. 2. During the photon exposure, the temperature was increased to about 20 K without a noticeable effect on the pressure. In line with several previous observations [1, 5], cracking of CO₂ into CO and O₂ could again be seen to dominate the desorption process.

4. Summary

For the four gases studied, the average removal coefficient shows a very similar behaviour, initially increasing with surface coverage and reaching a constant, maximum value for a sufficiently large surface coverage. For surface coverages below 10^{16} molecules/cm², the removal coefficients for all gases other than H₂ are so small that they could not be distinguished from the desorption of the bare substrate.

The maximum values of the removal coefficients for the four gas species were approximately 0.4–0.5 molecules/photon for H₂, CH₄ and CO₂ and 0.04 molecules/photon for CO.

All measurements for CO₂ between 5.5 and 20 K show the same variation with coverage and, in addition, are consistent with earlier results at 68 K as well as with measurements in the “closed-geometry” experimental set-up [3].

In the temperature range 5.5–20 K, no dependence of the removal coefficients on the substrate temperature could be observed.

5. Implications for the LHC vacuum system

In view of the operation of the LHC vacuum system [6], the results of this study, which were, however, obtained at perpendicular rather than grazing photon incidence, show that H₂ is the most critical gas species since its coverage on the inner wall of the beam screen must be

maintained well below a monolayer, i.e. $\ll 10^{15}$ molecules/cm². For the other gas species the following conclusions may be drawn:

(i) Even in the unlikely situation of thick, condensed layers of CH₄, CO and CO₂ on the cold vacuum chamber of the LHC, the dynamic pressure will be dominated by the primary desorption coefficients of these gas species from the bare substrate [7], i.e. from the cold beam screen. In contrast to what has been found with H₂, for which the removal (recycling) coefficient can reach values close to unity at a rather small coverage of 10^{15} molecules/cm², these heavier gases do not desorb strongly and thus do not require large pumping speeds to obtain the required low operating pressure. Therefore, a beam screen without pumping holes to the 1.9 K magnet cold bore could provide adequate vacuum conditions for these gas species.

(ii) Since the removal coefficients for the four gas species studied in this experiment do not depend to any significant extent on the temperature of the substrate, one may conclude that, as long as the gas can be efficiently cryosorbed on the 1.9 K cold bore, the temperature of the beam screen is not a critical parameter.

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