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**PHOTON STIMULATED DESORPTION AND THE EFFECT OF CRACKING OF
CONDENSED MOLECULES IN A CRYOGENIC VACUUM SYSTEM**

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Cracking of the molecular species CO₂ and CH₄ has been observed in recent studies and these findings have been incorporated in a more detailed dynamic gas density model for the LHC. In this paper, we describe the relevant physical processes and the parameters required for a full evaluation.

It is shown that the dominant gas species in the LHC vacuum system with its beam screen are H₂ and CO. The important result of this study is that while the surface coverage of cryosorbed CH₄ and CO₂ molecules is limited due to cracking, the coverage of H₂ and CO molecules may increase steadily during the long term operation of the machine.

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Photon Stimulated Desorption and the Effect of Cracking of Condensed Molecules in a Cryogenic Vacuum System

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Introduction

Over most of its circumference, the LHC vacuum system consists of a cold bore tube inside the 1.9 K cryomagnets combined with a perforated beam screen which is inserted in the cold bore to intercept beam induced heat loads from synchrotron radiation (SR), from image currents of the circulating beam and from photoelectrons. The experimental work on photon stimulated desorption in a vacuum system with cryosorbing walls started in 1993 at the Budker Institute of Nuclear Physics (Novosibirsk, Russia) and at the Brookhaven National Laboratory (Upton, NY, USA) in the context of the SSC project. During these early studies it was demonstrated that three physically distinct effects may occur: photon stimulated desorption of

chemically bound molecules, photon stimulated desorption of condensed, and thus only weakly bound molecules and finally the thermal vapour pressure of the condensed gas molecules. Recent studies, both at BINP and at CERN have highlighted in addition to these effects the importance for the LHC of cracking of cryosorbed molecules by SR. This effect, which had not been taken into account in earlier studies, will be addressed in this paper.

Photon induced desorption in a vacuum system with cryosorbing walls

In contrast to a room temperature vacuum system, where the gas molecules are readily removed by external pumps, in a cryogenic system molecules have generally a high probability to stick to the wall after only a few wall collisions. External pumping from the ends of a long vacuum vessel is rather inefficient and in most cases for the LHC, pumping from the ends of a vacuum vessel can be neglected entirely. As a consequence a steadily growing layer of condensed gas may accumulate on the wall during machine operation. A complete description of the processes, which determine the gas density during an extended period of operation, must include the following processes:

Desorption of strongly bound, chemisorbed molecules from the surface and from near-surface layers of the vacuum chamber, described by a primary desorption coefficient.

Desorption of weakly bound, physisorbed molecules from a surface layer of condensed molecules, described by a recycling coefficient. In fact, a molecule which has been desorbed from its strongly bound state, as described by the primary desorption coefficient, may readily re-condense in a weakly bound state where it has a considerably higher desorption probability. This 'recycling effect' of physisorbed molecules in a closed vacuum system depends strongly on the surface density of cryosorbed molecules and may lead to a large increase of the volume density in the system.

The thermal equilibrium pressure between surface and volume gas densities which can be described by an adsorption isotherm for the respective gas species. The sticking probability of molecules depends on the surface coverage, on the characteristics of the surface and on temperature.

A further process included in this study is the cracking of condensed gas molecules transforming one molecular species into one or more other species. For instance, CO₂ molecules have been found to produce CO and O₂. Due to this effect, the amount of condensed CO₂ in the system will be reduced while, at the same time, the cracking of this species constitutes an additional source of CO and O₂.

Incorporating these physical processes into the previous model for the cryogenic vacuum system [1], the balance for each individual gas species, i , can be written as:

$$V \frac{dn_i}{dt} = (\eta_i + \eta'_i + \chi_i) \dot{\Gamma} - \alpha_i S_i (n_i - n_{e_i}) - C_i n_i \quad (1)$$

$$A \frac{ds_i}{dt} = \alpha_i S_i (n_i - n_{e_i}) - (\eta'_i + \kappa_{i \rightarrow k+m}) \dot{\Gamma} \quad (2)$$

with: n the volume molecular density; s the surface molecular density; V the vacuum chamber volume and A the vacuum chamber wall area. $\dot{\Gamma}$ is the photon intensity; η the primary photo-desorption yield and η' is the recycling coefficient, $S_i = A \bar{v}_i / 4$ gives the ideal wall pumping speed, \bar{v}_i is mean molecular speed for the gas i ; and α is the molecular sticking probability. n_e is the thermal equilibrium gas density of the condensed gas. Furthermore, C represents the distributed pumping speed, in the LHC provided by the pumping slots in the beam screen [2]. The effect of cracking of molecules is described by the parameters $\kappa_{i \rightarrow k+m}(s_i)$ is the cracking efficiency of type i molecules into type k and type m and $\chi_i(s_j)$ describes the inverse process, the efficiency of producing type i molecules by cracking of the type j parent molecules. The two quantities χ and κ are connected through the relation:

$$\chi_i(s_j) = a_{i,j} \kappa_{j \rightarrow i+n}(s_j),$$

involving the coefficients $a_{i,j}$.

The behaviour of the vacuum system of the LHC depends on many parameters of which not all are known with sufficiently precision. It should be noted that most parameters in this model depend implicitly on the surface coverage. The limited amount of available experimental data and the choices made for this study will be reviewed in Section 3.

The system equations (1) and (2) describe the two cases which are particularly relevant for the LHC vacuum system: a bare cold bore tube, i.e. a beam pipe without a beam screen, and a cold bore which contains a beam screen with pumping holes. These two cases are characterised by the conditions $C = 0$ and $C > 0$ respectively. Since in this study one is not concerned with the fast transient behaviour but rather in the slow evolution over many hours and even months of photon irradiation, the following approximations for a ‘quasi-static’ vacuum system have been made:

$$\frac{dn_i}{dt} \approx 0 \quad \text{and} \quad \frac{ds_i}{dt} \neq 0 \quad (3)$$

LHC cold bore without a beam screen

The gas density inside a long, cold vacuum system can be obtained by neglecting the gas flow towards both ends of the system. This approach approximates a ‘closed system’ which becomes independent of boundary conditions and gives a uniform longitudinal density distribution. With the assumption of a quasi-static system the gas density for each gas species is given by:

$$n_i \approx \frac{(\eta_i + \eta'_i(s_i) + \chi_i(s_j))\dot{\Gamma}}{\alpha_i S_i} + n_e$$

Due to the dependence of most of the parameters on the surface coverage, the gas density will slowly evolve during the exposure to SR. Furthermore, due to the cracking process, the different cryosorbed species are transformed from one type into another and their partial pressures become mutually interdependent.

The slowly changing surface density $s(t)$ of the cryosorbed molecules can be computed stepwise starting from the initial status of the system:

$$s_i(t) = s_i(0) + \frac{1}{A} \int_{t=0}^t (\eta_i + \chi_i(s_j) - \kappa_i(s_i)) \dot{\Gamma} dt .$$

A very general estimate of the maximum gas density in the system can be obtained by assuming that the recycling coefficient as well as the cracking yield of all gas species increase with surface coverage until they reach their respective maximum values η_{max} , κ_{max} and χ_{max} [3,4,5,6]. Then the density would have an upper bound:

$$n_i \leq \frac{\left(\eta_i + \eta'_{i_{max}} + \sum_{j \neq i} \chi_{i_{max}}(s_j) \right) \dot{\Gamma}}{\alpha_i S_i} + n_e$$

In the more specific case when there is no pre-condensed gas at the beginning of the SR irradiation one arrives at a slightly different estimate since in this case the rate of cracking of molecules can never exceed the primary production rate and therefore $\kappa_{j \rightarrow i+k} = a_{i,j} \chi_i(s_j) \leq \eta_j$. From this condition follows:

$$n_i \leq \frac{\left(\eta_i + \eta'_{i_{max}} + \sum_{j \neq i} \frac{\eta_j}{a_{i,j}} \right) \dot{\Gamma}}{\alpha_i S_i} + n_e .$$

A further case occurs for a species which is not the result of cracking by some parent molecules but which is exclusively produced by primary desorption with SR. For this particular

species two conditions apply: $\chi_i(s_m) = 0$ and $\kappa_{i \rightarrow j+k}(s_i) \neq 0$. Therefore, when an initially bare surface is irradiated with photons, $\kappa_i(s_i)$ will increase until it reaches its maximum $\kappa_{i \max}(s_i) = \eta_i$ when the rate of production of molecules by the primary desorption is in balance with the recycling rate and rate of cracking and thus $\frac{ds_i}{dt} = 0$. Defining the parameter r_i , as the ratio of the maximum of both, the recycling coefficient and of the cracking coefficient, $r_i = \eta'_{i \max} / \kappa_{i \max}$, one obtains the condition $\eta'_i < \eta'_{i \max} = r_i \kappa_{i \max} = r_i \eta_i$. With this approximation the gas density has the upper bound:

$$n_i \leq \frac{\eta_i(1 + r_i) \dot{\Gamma}}{\alpha_i S_i} + n_e.$$

LHC cold bore with a beam screen

The configuration with a beam screen and pumping holes is characterised by $C > 0$, hence by a non-vanishing distributed pumping speed in addition to any wall pumping effect. The quasi-static gas density in this system is given by:

$$n_i = \frac{(\eta_i + \eta'_i + \chi_i) \dot{\Gamma} + \sigma_i S_i n_e}{\alpha_i S_i + C_i}$$

Contrary to the situation with a bare cold bore, the gas density and the surface density on the beam screen will always be limited by the distributed pumping C of the holes in the screen. The surface density can be computed with the expression:

$$s_i(t) = s_i(0) + \frac{1}{A} \int_{t=0}^t [(\eta_i + \chi_i(s_i) - \kappa_i(s_j)) \dot{\Gamma} - C_i n_i] dt$$

Under conditions where the thermal equilibrium density n_e can be neglected, the slowly varying gas density can be expressed as:

$$n_i(t) = \frac{(\eta_i + \chi_i - \kappa_i) \dot{\Gamma}}{C_i} - \frac{A}{C_i} \frac{ds_i}{dt}.$$

During a long SR exposure the gas density will increase and when the surface coverage has reached a constant value such that $\frac{ds}{dt} = 0$, which in turn implies a constant gas density independent of the wall pumping speed, one obtains:

$$n_i = \frac{(\eta_i + \chi_i - \kappa_i) \dot{\Gamma}}{C_i}.$$

Since at this equilibrium $\kappa_{j \rightarrow i+k} = a_{i,j} \chi_i \leq \eta_j$, it has an upper limit

$$n_i \leq \frac{\left(\eta_i + \sum_{j \neq i} \frac{\eta_j}{a_{i,j}} \right) \dot{\Gamma}}{C_i} \text{ and for } \eta_j = 0, \text{ it reduces to } n_i \leq \frac{\eta_i \dot{\Gamma}}{C_i}$$

Experimental parameters

Primary desorption yield

The primary desorption yield, η , and its variation with the accumulated photon dose, D , has been studied at low temperature extensively by different experiments [1,2]. For a SR spectrum with a critical photon energy of about 50 eV a satisfactory parameterisation of the desorption yield for a OFHC copper vacuum chamber and for grazing photon incidence, is given by

$$\eta = \eta_0 (D_0 / D)^{0.33}$$

where η_0 is the photodesorption yield at the given photon dose D_0 . The temperature of the test chamber for these measurements was 78 K. Since experimental data only exist for a photon dose between 10^{19} to 10^{22} photons/m, an extrapolation to a larger photon dose, e.g. for the LHC with a typical linear photon flux of 10^{17} photons/(m·s), has been obtained using the parameters in Table 1.

Table 1. Primary photodesorption yield at 78 K and 10 K

	$\eta(78 \text{ K})$ D= 10^{19} photons/m	$\eta(10 \text{ K})$ D= 10^{19} photons/m	$\eta(10 \text{ K \& } 78 \text{ K})$ D= 10^{24} photons/m
H ₂	$2 \cdot 10^{-3}$	$4 \cdot 10^{-4}$	$5 \cdot 10^{-5}$
CH ₄	$2 \cdot 10^{-5}$	—	$9 \cdot 10^{-7}$
CO	$2 \cdot 10^{-4}$	—	$5 \cdot 10^{-6}$
CO ₂	$3 \cdot 10^{-5}$	—	$5 \cdot 10^{-7}$

It should be noted that in the LHC a dose of 10^{24} photons/m corresponds to approximately 4 months of continuous operation. Comparing the data for H₂ at 78 K with measurements between 5 to 10 K, the initial desorption yield at 5 K is less by about a factor of 5, however, at a high accumulated photon dose the desorption yield seems to be less dependent on temperature [2].

Recycling and cracking of molecules

Molecules condensed on a cryogenic surface may be desorbed by SR either as the original molecule, or may be cracked and subsequently form new species. In experiments, which reproduce the closed geometry of the LHC vacuum system, it has been possible to observe only the compound quantity $(\eta + \eta' + \chi)/\alpha$ rather than the individual contributions, since the wall pumping can not be neglected. From such a measurement the ratio η/α for H₂ was obtained and was found to increase with surface coverage and to saturate at a value of about 0.5 H₂ molecules/photon for a coverage larger than $3 \cdot 10^{15}$ molecules/cm² [3,4]. For CH₄, CO and CO₂ only an upper limit could be deduced [4]:

$$(\eta + \eta')/\alpha \approx 10^{-3} \text{ molecules/photon.}$$

More recent experiments, which were performed in an 'open geometry' set-up [5,6], circumvent the problem of re-condensation of molecules and hence do not require the knowledge of the sticking probability α . With this experiment the 'removal coefficient' η'_r , i.e. the number of pre-condensed molecules desorbed per incident photon, was measured for H₂, CH₄, CO and CO₂ in a temperature range from ~3 K for H₂ up to 68 K for CO₂. The removal coefficient was found to increase with surface coverage and to reach a saturation value of 0.55 molecules/photon for H₂ at $3 \cdot 10^{15}$ molecules/cm². The saturation values for CH₄, CO and CO₂ were found to be 0.45, 0.4 and 0.04 molecules/photon at a surface coverage of $\sim 10^{19}$ molecules/cm². Since the removal coefficient includes both, desorption and cracking of molecules, it corresponds to $\eta'_r = \eta' + \kappa$. Nevertheless, since the pre-condensed gas species is known in this experiment, it is possible to deduce the ratio η'/κ from the partial pressures of a newly appearing species. The main findings of this experiment have been that the desorption of a thick layer of condensed CO₂ and CH₄, equivalent to many hundred monolayers, involves predominantly cracking of these molecules, e.g.:



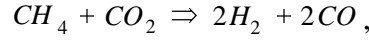
The cracking process has been found to exceed the desorption (recycling) rate of these molecules by a factor of 10. No dependence on temperature could be observed in the range from 3 K to 20 K for H₂ and up to 68 K for CO₂. To estimate the parameters for these two gas species one may assume that, e.g. cracking of CO₂ generates one molecule of CO and one atom of O, hence

$$\chi_{\text{CO}}(s_{\text{CO}_2}) = \kappa_{\text{CO}_2 \rightarrow \text{CO} + \text{O}} \quad \text{and} \quad \chi_{\text{O}_2}(s_{\text{CO}_2}) = 0.5 \kappa_{\text{CO}_2 \rightarrow \text{CO} + \text{O}}.$$

Similarly, cracking of CH₄ would produce two H₂ molecules and one C, hence

$$\chi_{H_2}(s_{CH_4}) = 2\kappa_{CH_4 \rightarrow 2H_2 + C} \text{ and } \chi_C(s_{CH_4}) = \kappa_{CH_4 \rightarrow 2H_2 + C}.$$

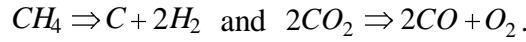
Finally, it may be necessary to include the process:



which would imply :

$$\chi_{CO}(s_{CO_2} + s_{CH_4}) = \kappa_{CO_2 \rightarrow CO + O} + \kappa_{CH_4 \rightarrow 2H_2 + C}.$$

In a cryogenic vacuum system H_2 , CH_4 , CO and CO_2 represent the main species which are desorbed by SR and two of them may be cracked by photons to form new molecules, e.g. :



As a consequence, an additional source of H_2 , CO and O_2 appears in the vacuum system. Since in practice $\eta_{CO_2} \approx \eta_{CH_4}$ one may assume that a sufficient amount of these molecules are produced such that the overall process

$$k_1 CH_4 + k_2 CO_2 \Rightarrow 2k_1 H_2 + (k_1 + k_2) CO + \left(\frac{k_2}{2} - k_1\right) O_2$$

is well balanced. Estimates of the maximum values of the recycling yield and of the cracking parameters are summarised in Table 2.

Table 2. Upper bounds for the parameters of recycling and cracking.

	H_2	CH_4	CO	O_2	CO_2
$\eta'_{r \max}$	0.55	0.4	0.04	—	0.45
η'_{\max}	0.55	0.04	0.04	no data (0.04)	0.04
κ_{\max}	—	$\kappa_{CH_4 \rightarrow 2H_2 + C}$ ≈ 0.36	—	—	$\kappa_{CO_2 \rightarrow CO + O}$ ≈ 0.41
χ_{\max}	$\chi_{H_2}(s_{CH_4})$ ≈ 0.72	—	$\chi_{CO}(s_{CO_2} + s_{CH_4})$ $\approx 0.41 + 0.36$	$\chi_{O_2}(s_{CO_2})$ ≈ 0.2	—

In the absence of data for the recycling effect of O_2 , the same value as for CO has been assumed provisionally for the calculations.

Sticking probability

The sticking probability of molecules on either the bare cold bore at 1.9 K or on the cold beam screen at ~20 K is a crucial parameter for the understanding of a cryogenic vacuum system as

shown in the preceding sections. The sticking probability for H₂ on a beam screen with a stainless steel or a copper coated surface in the temperature range from 4.2 to 11 K was found to lay between 0.01 to 0.6 [1,2]. The sticking probability for CO under similar conditions is not known but may be expected to fall in the same range. Measurements of the sticking probability for CO₂ [4] indicate a sticking probability of 10⁻⁴ on a bare copper surface at ~77 K. However, with an increasing amount of pre-condensed CO₂ the sticking probability was found to increase and to approach unity as observed for hydrogen.

Dynamic gas density in the LHC

Vacuum system with a beam screen

For the LHC vacuum system with a beam screen there is a minimum net pumping speed by the pumping slots independent of the sticking probability of molecules on the surface of the beam screen. Assuming that the thermal vapour pressure of the gas species concerned always remains sufficiently low that it can be neglected with respect to the SR induced dynamic pressure, the upper limits of the gas densities for H₂, CO and O₂ can be estimated from the expressions :

$$n_{H_2} \leq \frac{(\eta_{H_2} + 2\eta_{CH_4}) \dot{\Gamma}}{C_{H_2}}$$

$$n_{CO} \leq \frac{(\eta_{CO} + \eta_{CO_2} + \eta_{CH_4}) \dot{\Gamma}}{C_{CO}}$$

$$n_{O_2} \leq \frac{0.5\eta_{CO_2} \dot{\Gamma}}{C_{O_2}}.$$

Similarly the gas density for CH₄ and CO₂ may be derived from the expressions:

$$n_{CH_4} \leq \frac{\eta_{CH_4} (1 + r_{CH_4}) \dot{\Gamma}}{\sigma_{CH_4} S_{CH_4} + C_{CH_4}}$$

$$n_{CO_2} \leq \frac{\eta_{CO_2} (1 + r_{CO_2}) \dot{\Gamma}}{\sigma_{CH_4} S_{CH_4} + C_{CO_2}}.$$

where by r is the ratio $r_i = \eta'_i \max / \kappa_i \max$. Numerical estimates for a typical set of parameters are presented in Table 3.

Table 3, Upper limits of the gas density in the LHC with a beam screen.

Temperature of beam screen	n(H ₂) [H ₂ /m ³]	n(CH ₄) [CH ₄ /m ³]	n(CO) [CO/m ³]	n(O ₂) [O ₂ /m ³]	n(CO ₂) [CO ₂ /m ³]
T = 5 K	8.2·10 ¹⁴	2.3·10 ¹³	3.6·10 ¹³	2.4·10 ¹³	5.6·10 ¹³
T = 20 K	4.1·10 ¹⁴	1.2·10 ¹³	1.8·10 ¹⁴	1.2·10 ¹³	2.8·10 ¹³

Evolution of the gas density in the LHC

The gas density in the LHC vacuum system with a cold bore at 1.9 K and a beam screen at 20 K has been calculated using the numerical data from Table 1. At 1.9 K the saturated vapour pressure of all gases including hydrogen can be neglected, while at the temperature of the beam screen the saturated vapour pressure of hydrogen is very large, which implies that only a negligible amount of H₂ can be cryosorbed at this temperature. The assumed photon flux is 10¹⁷ photons/(sec·m) corresponding to 'nominal' operating conditions of the LHC. The evolution of the gas density of the surface coverage and of the recycling coefficient as a function of the accumulated photon dose are shown in Figures 1, 2 and 3. Here it has been assumed that the sticking probabilities are 0.5 for H₂ and ~1 for all other gases. It can be seen that the dynamic vacuum is rather sensitive to the effect of cracking of CH₄ and of CO₂. Furthermore, the surface coverage for these gases is limited by the cracking.

Figure 1 illustrates that at the relatively low temperature of 5 K, the H₂ density increases to a maximum value after about 15-20 minutes of irradiation and decreases for further irradiation. The initial increase is due to the finite pumping capacity of the beam screen and the maximum is reached when the secondary desorption yield is compensated by the wall pumping speed and the surface coverage reaches its maximum, see Figure 3.

The evolution of the gas density for CH₄ and CO₂ is maximum at the start of irradiation and decreases as a function of accumulated dose due to photon scrubbing. The surface coverage is limited by cracking and the secondary desorption of these gases is negligible.

It is interesting to observe in Figure 1 that the density of CO initially decreases due to the photon scrubbing of the surface. However, from a photon dose of about 10²² photons/m onwards it increases again because of the additional contribution from cracking which in turn

increases the surface coverage and results in a higher recycling rate. Nevertheless, since the recycling coefficient for CO remains relatively low up to a rather high surface coverage of many 1000 monolayers the gas density does not reach a saturation level even at an accumulated photon dose of 10^{24} photons/m. This dose corresponds to approximately four months of LHC operation.

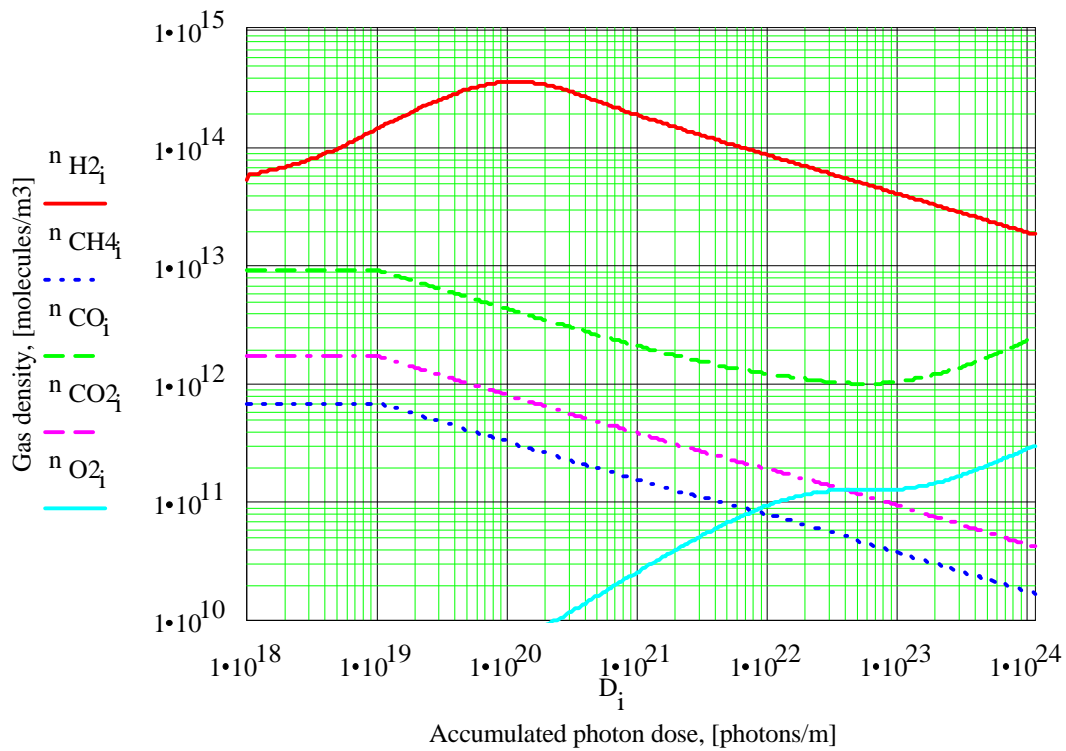


Figure 1. Gas density (molecules/m³) for H₂, CH₄, CO, O₂ and CO₂ as a function of the linear accumulated photon dose in the LHC with a beam screen at 5K.

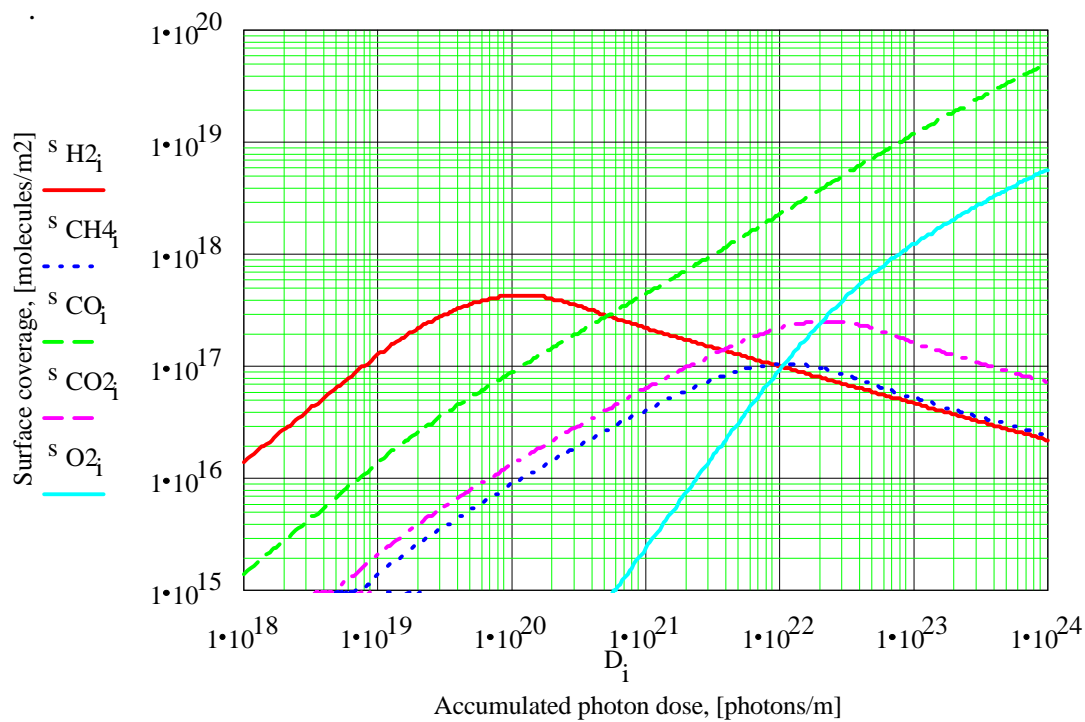


Figure 2. Surface density (molecules/m²) for H₂, CH₄, CO, O₂ and CO₂ as a function of the linear accumulated photon dose in the LHC with a beam screen at 5K.

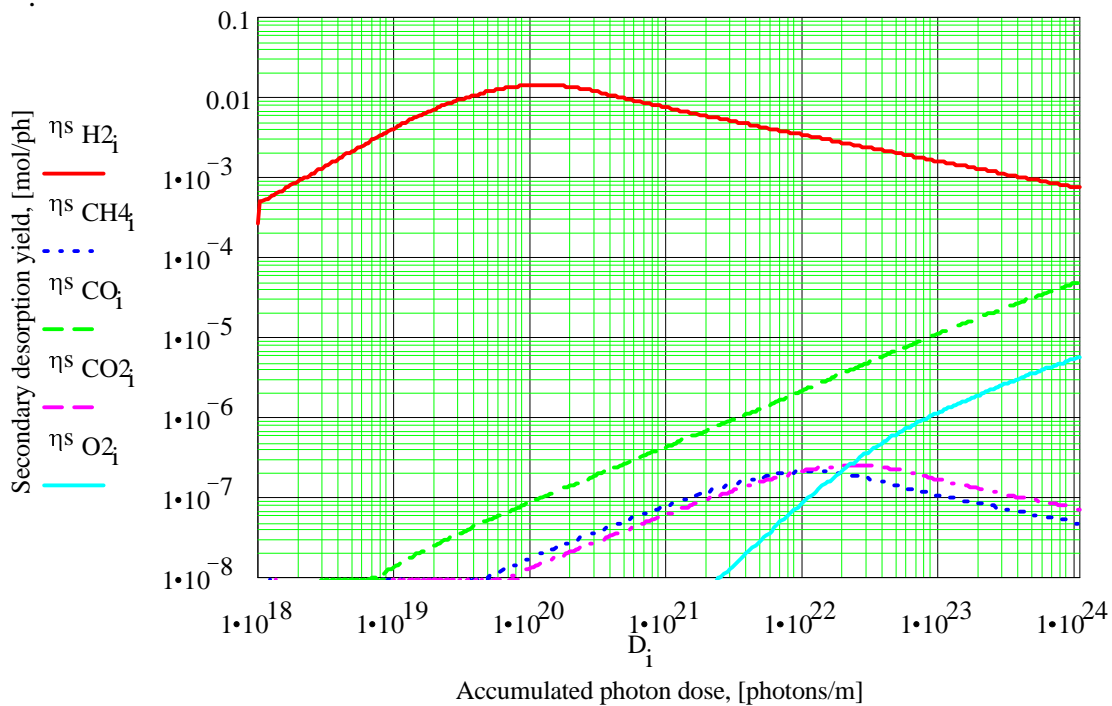


Figure 3. The recycling coefficient (molecules/photon) for H₂, CH₄, CO, O₂ and CO₂ as a function of the linear accumulated photon dose in the LHC with a beam screen at 5K.

For the LHC beam vacuum a very important requirement is that the energy dissipation due to beam losses by nuclear scattering on the rest gas, which are directly related to the beam lifetime, must not exceed the cooling capacity of the 1.9 K cryogenic system. This criterion defines a ‘lifetime limit’ and a minimum beam lifetime in the LHC [4]. The tolerable losses budget of 0.1 W/m correspond to an average hydrogen density of 10^{15} molecules/m³. For heavier molecules this density has to be divided by a factor 5.4 for CH₄, 7.8 for CO and 12.2 for CO₂. Figure 4 shows the total equivalent H₂ density as a function of the photon exposure. It can be seen that the lifetime limit will not be reached even after many months of LHC operation.

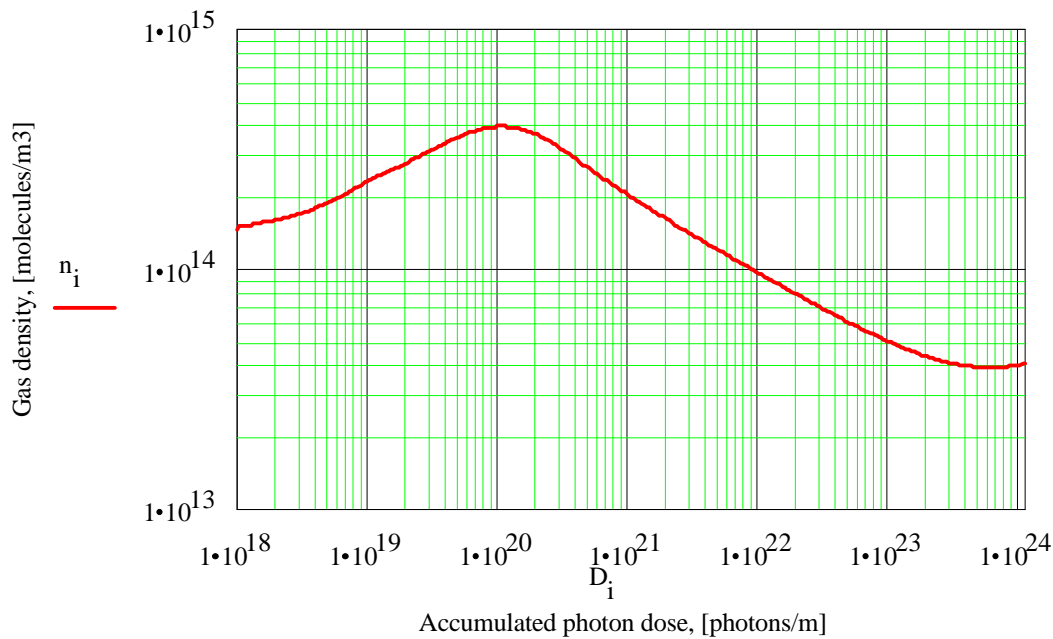


Figure 4. Hydrogen equivalent gas density with respect to the lifetime limit

Conclusions

In this study the dominant vacuum processes in a vacuum chamber with cryosorbing walls and exposed to synchrotron radiation have been studied based on the most reliable experimental parameters available. With respect to the cryogenic vacuum system of the LHC with its beam screen, it can be shown that the dominant gas species are H₂ and CO. Due to the pumping slots

in the beam screen, there is a continuous transfer of molecules from the inside of the beam screen to the outer cold bore surface where all molecules can be cryosorbed permanently and with a negligible vapour pressure.

The amount of cryosorbed molecules of CH₄, O₂ and CO₂ is limited due to the combined effects of recycling and cracking of these molecules by SR. The surface coverage of CO will increase steadily during the LHC operation since its recycling coefficient is rather small. CO will therefore accumulate until the system is warmed up and this gas can be removed by external pumps. During warm up after a long period of operation the main gases to be pumped will be H₂, CO and perhaps O₂.

This study also confirms that the total gas density inside that vacuum system will remain below the beam lifetime limit also when considering the effect of cracking, except in the unrealistic case of a very thick pre-condensed layer of CO or CO₂.

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