Reduction of the Photoelectron Induced Gas Desorption in the PETRA Vacuum System by in situ Argon Glow Discharge Cleaning

by

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Introduction

In the large electron positron storage ring PETRA (Positron-Electron-Tandem-Kraftbeschleuniger-Anlage), the pressure in the aluminium vacuum chamber must be such that up to 90 mA of electrons and 90 mA of positrons with energies between 5 and 19 GeV can be stored for periods of the order of some hours without undue loss from beam-residual gas interactions.

In addition to the static thermal outgassing of the vacuum chamber, the pumping system must cope with gas desorbed from the chamber walls by photoelectrons liberated by synchrotron radiation \(^1,2,3\). This electromagnetic radiation, in PETRA amounting to 4 MW and distributed over 2000 m, is concentrated mainly in the bending magnets and has a spectral distribution extending from infrared to X-rays. Such a large machine (bending radius 192.05 m) means that the synchrotron radiation strikes the chamber at a rather small angle (\( \approx 20 \times 10^{-3} \) radians) thus enhancing the photoelectron production and hence the gas desorption. It is imperative therefore that the vacuum chamber inner surfaces be as clean as possible to minimize the initial photoelectron-induced gas desorption and reduce, if not eliminate, the running time of the machine necessary to clean the surfaces and lower the gas desorption to an acceptable level.

In this paper we calculate the photon energy distribution of the synchrotron radiation for the 5 - 19 GeV energy range of PETRA and from published values of the experimentally measured photoelectric yield of Al we calculate the total number of photoelectrons produced.

From measurements of the desorption coefficient (number of molecules desorbed per incident electron) of an Al chamber before and after in situ glow discharge cleaning \(^2,4\), the photoelectron induced pressure increases in PETRA are predicted.
Calculation of the number of photoelectrons

To calculate the total number of photoelectrons produced by the incident photons the photoelectron yield and the number of photons must be known over the complete energy range of the incident radiation, i.e., from 10 eV to 10^6 eV. In addition, the photoelectric yield as a function of angle of incidence must be known.

The glancing angle of incidence of the synchrotron radiation in PETRA is not constant but runs from a maximum of 24.4 x 10^-3 radians in part of the bending magnet chamber to a minimum of 16.5 x 10^-3 radians at the end of the straight section between two bending magnets. This spatial dependence of the glancing angle of incidence $\delta$ is shown in Fig. 1. The average value of $\delta$ is 20 x 10^-3 rad.

Between 90° and 10° the photoelectric yield is a $1/\sin\delta$ dependence. Recent unpublished measurements here at DESY 5) indicated that below 10° the yield departs from this $1/\sin\delta$ law and decreases with decreasing glancing angle of incidence. Thus use of the $1/\sin\delta$ dependence will, if anything, overestimate the yield. Lacking any better angular dependence, we have used the $1/\sin\delta$ dependence which will then correspond to the worst possible case. For $\delta = 20 \times 10^{-3}$ radians the ratio of the yield at that glancing angle of incidence to the yield at normal incidence is 50.

From the literature we have constructed the curve shown in Figure 2 of the normal incidence photoelectric yield from Al or Al_{0.3} for photon energies between 7 and 10^6 eV. Between 200 and 1100 eV and 3 x 10^4 and 10^6 eV no data were available. Using the measured absorption coefficient $\mu$ for Al in these energy ranges (Figure 3) and assuming that the yield may be given by 6)

$$\text{photoelectric yield} = \mu \cdot \varepsilon \cdot c$$

where $c$ is the photon energy, the dashed regions of Figure 2 were obtained.

In fact, the experimental data between $2 \times 10^3$ and $3 \times 10^4$ eV could be reasonably well described by the same expression. For the photoelectric yield at 20 \times 10^{-3} radians glancing angle of incidence Fig. 2 has of course to be multiplied by 50.

The number of photons of synchrotron radiation per eV per second per mA of beam $d^2 N_{ph}/d\varepsilon dt$ is given by 19)

$$\frac{d^2 N_{ph}}{d\varepsilon dt} = \frac{6.95 \times 10^{13}}{R^2} \int K_{5/3}(x) \, dx$$

(1)

where $R$ is the electron energy in GeV, $\varepsilon$ is the photon energy in eV, $\varepsilon_c$ is the critical energy $= 2.218 \times 10^3 \frac{R}{R}$ (eV), $R$ is the bending radius in metres, $y = \varepsilon/\varepsilon_c$ and $K_{5/3}$ is a modified Bessel function of second kind and order 5/3.

In Figure 4 is shown the spectral distribution of photons computed from equation (1) for energies of 5, 14 and 19 GeV.

The number of photoelectrons per eV per second per mA of beam (at normal incidence) was obtained by multiplying the photoelectric yield of Figure 2 by the photon spectral distribution of Figure 4. By integrating over the photon energy, multiplying by 50 to account for the glancing angle of incidence, multiplying by the total beam current (electrons + positrons) shown in Figure 5 for PETRA 20) and dividing by the total length of chamber struck by synchrotron radiation (~ 2000 m), the total photoelectron current in mA from each metre of chamber is obtained as a function of beam energy. This is shown in Figure 6 where it is seen that the maximum number of photoelectrons produced is 250 mA m^-1 at 14 GeV.
Most of these photoelectrons have energies less than about 30 eV \(^2\)
but to simulate the worst possible desorption in the laboratory we
have chosen a bombarding energy of 300 eV.

The dynamic pressure in PETRA

In the following section it is shown that the desorption coefficient \(\eta\)
for one species \(g\) in molecules per electron may be written

\[
\eta_g = \frac{5.23}{I} \times S_g \times \Delta P_g(Abs) \tag{2}
\]

Using this equation we are therefore now in a position to calculate the
absolute rise in pressure \(\Delta P(Abs)\) as a function of beam energy using
the calculated photoelectron current \(I\) of Figure 6 and the values of
the pumping speed \(S_g\) of the distributed ion pumps for various \(\eta\) values.
The photoelectrons may desorb twice - once on leaving the surface and a
second time on re-entering. This means that in the calculation
of \(\Delta P(Abs)\) the total photoelectron current \(I\) obtained from Figure 6
must be multiplied by 2. For the distributed ion pumps the pumping
speeds for CO, CO\(_2\) and CH\(_4\) were assumed to be equal but the pumping speed
for H\(_2\) to be a factor of two higher \(^{21}\). In Figure 7 is shown the
pumping speed of the distributed ion pumps for CO as a function of beam
energy.

In Figure 8 is shown the expected rise in absolute pressure in PETRA
as a function of beam energy for \(\eta\) values between \(10^{-4}\) and \(10^{-7}\) mol. el\(^{-1}\).
Thus, to keep the pressure rise in the \(10^{-9}\) torr range, \(\eta\) values \(<\ 10^{-5}\)
mol. electron\(^{-1}\) are necessary.

The beam lifetime is determined mainly by Bremsstrahlung interactions
of the stored beam with the nuclei of the residual gas \(^{22}\). The probability
\(W\) that an electron be scattered within one radiation length is given by \(^{22}\)

\[
W = \frac{4}{3} \ln \frac{\gamma}{\Delta \gamma} - \frac{5}{6}
\]

where \(\gamma = \frac{E}{m_e c^2}\) is the beam energy referred to the electron rest mass
and \(\Delta \gamma\) is the maximum tolerable energy spread of the stored electrons.
For PETRA \(\frac{\gamma}{\Delta \gamma} = 500\) and thus gives a value for \(W\) of 7.45.

The beam lifetime \(\tau\) is given by \(^{22}\)

\[
\tau = \frac{X_0}{\rho c}
\]

where \(X_0\) is the radiation length of the residual gas in g cm\(^{-2}\)
\(\rho\) is the residual gas density in g cm\(^{-3}\)
and \(c\) is the velocity of light in cm s\(^{-1}\).
Thus, for a pressure \(P\) in torr of a gas of molecular weight \(N\), the
expression for the lifetime in hours becomes

\[
\tau = 2.12 \times 10^{-8} \frac{X_0}{\rho P} \tag{3}
\]

The radiation length \(X_0\) depends on the square of the atomic number of
the residual gas molecules \(^{23}\) and may be written as follows:

\[
X_0^{-1} = \frac{4}{137} \frac{N}{A} \frac{\rho}{r_0^2} \frac{2}{2(\gamma+3)} \left[ \ln \left( \frac{1912}{137} \right) \left( \frac{\rho}{2(\gamma+3)} \right)^2 \right]
\]

where \(N\) is the Avogadro's number
\(A\) is the atomic weight
\(r_0\) is the classical electron radius
and \(J\) is a correction factor which includes contributions from the
electrons and deviations from the Born approximation.
From published radiation lengths for the different elements \(^{23}\), the radiation lengths of different gases and their relative influence on the beam lifetime may be calculated. The relative beam lifetimes are shown in Table 1 where it can be seen that, with reference to CO, for the same effect on the beam lifetime, the partial pressure of H\(_2\) would have to be 23.2 times higher than the CO partial pressure. However, a partial pressure of Argon only 0.36 that of CO can be tolerated.

From Equation (3) the absolute beam lifetime in \(1 \times 10^{-8}\) torr CO is calculated to be 2.87 hours. But, at \(2 \times 10^{-8}\) torr and a typical residual gas composition of 90% H\(_2\) and 10% CO, the beam lifetime rises to 10.25 hours.

Experimental

The experimental arrangement for measuring the electron induced desorption coefficients from an Al vacuum chamber is shown schematically in Figure 9.

The stainless steel part of the vacuum system was bakeable to 300°C and the Al chamber to 150°C.

The system was pumped by a 600 l/s \(^{2}\) ion pump and during bakeout by a turbomolecular pump. The actual measuring chamber was pumped via a diaphragm with a 10 mm hole giving a conductance of 9.27 l/s for H\(_2\).

The total pressure was measured with a calibrated Bayard-Alpert ionisation gauge and the partial pressure with a quadrupole mass spectrometer (Balzers model 111A). The sensitivity of the quadrupole in torr (H\(_2\) equivalent) A\(^{-1}\) was determined for H\(_2\), CH\(_4\), CO, CO\(_2\) and Ar by injecting these gases one at a time into the system via a leak valve and measuring the quadrupole ion current and the corresponding total pressure in H\(_2\) equivalent.

A tungsten wire 0.25 mm in diameter and 90 cm long, stretched along the axis of the Al chamber, heated by alternating current and biased negatively to 300 V with respect to the chamber, gave a maximum electron emission current of \(\sim 40 \text{ mA}.\) The surface area of chamber bombarded by electrons was \(2.4 \times 10^{-3} \text{ cm}^2.\)

Argon glow discharge cleaning of the Al chamber was accomplished by introducing via the leak valve \(\approx 10^{-2}\) torr of pure argon (99.99%) and pumping continuously with the turbomolecular pump. The glow discharge was initiated by applying about \(+350\) V to the W wire, the resulting discharge current being about 0.5 A. Even distribution of the discharge could be checked by feeling the temperature of the Al chamber along its length and also, but less reliably, by usual inspection through the end window. After about 1 hour at 0.5 A and 350 V, the chamber attained a temperature of \(\approx 80^\circ\text{C}.\)

During an electron bombardment measurement, the Al chamber was water cooled. Bombarding current densities between \(6.0 \times 10^{-8}\) and \(2 \times 10^{-7}\) A cm\(^{-2}\) were used. The lowest possible current density was always used to avoid excessive desorption and hence cleaning of the surface.

The Al chamber was cleaned following the procedure described in Appendix A.

The desorption coefficient \(n\) is defined as

\[
    n = \frac{\Delta Q}{I/e} \text{ mol. electron}^{-1}
\]

where \(\Delta Q\) is the number of molecules desorbed
\(I\) is the bombarding electron current in amps
\(e\) is the electronic charge.

For a gas \(g\), for which the pumping speed of the system is \(S_g\) (l/s) and the corresponding rise in absolute pressure is \(\Delta P_g\) (Abs), the expression for \(n\) may be written
\[ \eta = \frac{5.23}{1} \sum_{s} \Delta P (\text{Abs}) \]

where the summation is taken over all gases desorbed and the factor 5.23 takes account of the conversion from molecules \( e^{-1} \) to torr \( e^{-1} \) \( \Delta P (\text{Abs}) \) and also \( \eta \). If we write

\[ \Delta P (\text{Abs}) = \Delta P (N_2 \text{ equiv}) \times R_g \]

and

\[ \Delta P (N_2 \text{ equiv}) = P \times \Delta P_{\text{total}} (N_2 \text{ equiv}) \]

where \( \Delta P_{\text{total}} (N_2 \text{ equiv}) \) is the total change in pressure read on the gauge (in \( N_2 \) equivalent) on desorption and \( \Delta P (N_2 \text{ equiv}) \) is the partial change in pressure of gas \( g \) (in \( N_2 \) equivalent) and the fraction of \( \Delta P_{\text{total}} (N_2 \text{ equiv}) \) due to desorption of gas \( g \) is \( P_g \) it can be shown \(^{24} \) that for a desorbed species \( g \) the desorption coefficient \( \eta_g \) is given by

\[ \eta_g = \frac{5.23}{1} \frac{\Delta P_{\text{total}} (N_2 \text{ equiv})}{\Delta P (N_2 \text{ equiv})} \times R_g \times P \times \Delta P_{\text{total}} (N_2 \text{ equiv}) \]

(3)

Results and discussion

On bombarding the Al chamber surface with 300 eV electrons only four species were desorbed, \( H_2 \), \( CH_4 \), \( CO \) and \( CO_2 \). In Figure 10 is shown the value of the measured desorption coefficients \( \eta_g \) for these four gases for various treatments.

In the as received state (after normal chemical cleaning) the largest desorption coefficient was \( \eta_{H_2} = 8.7 \text{ mol. el}^{-1} \), followed by \( \eta_{CO} = 3 \text{ mol. el}^{-1} \) and \( \eta_{CO_2} = 2.1 \text{ mol. el}^{-1} \) with the smallest being \( \eta_{CH_4} = 0.23 \text{ mol. el}^{-1} \).

A bakeout at \( 150^0 \text{C} \) for 24 hours did little to ameliorate the situation and resulted in \( \eta_{H_2} = 3.9 \text{ mol. el}^{-1} \), \( \eta_{CO} \) and \( \eta_{CO_2} = 1 \text{ mol. el}^{-1} \) and \( \eta_{CH_4} = 0.12 \text{ mol. el}^{-1} \).

Bakeout temperatures of \( 150^0 \text{C} \) are only effective in desorbing in a reasonable time gases in states whose binding energies are less than about 1.0 eV, thus the populations of the more tightly bound states remain relatively unchanged. Since in the first approximation changes in \( \eta \) for a certain species reflect changes in the relative surface coverage of that species and some of the binding energies of gases on Al almost certainly have values in excess of 1.0 eV the relative ineffectiveness of the \( 150^0 \text{C} \) bakeout in reducing \( \eta \) may be understood in terms of large quantities of tightly bound (>1.0 eV) gas on the Al surface.

The magnitude of these desorption coefficients and their relative change after bakeout are in general agreement with other electron and ion desorption measurements on Al. \(^{25,24} \).
0.3% CO₂ and 1.7% Ar; however after several days pumping the relative concentrations of H₂O, CO and Ar tended to decrease.

To further reduce the desorption coefficients the surface was electron bombarded with 11.5 mA at 300 eV for 205 minutes (1.6 × 10⁻⁵ A cm⁻²), then for a further 20 hours at 40 mA (3.3 × 10⁻⁶ A cm⁻²). The nH₂, after the second cleaning, reduced to 7.8 × 10⁻⁶ mol. el⁻¹ and nCO and nCO₂ were all below the limit of measurement, i.e. = 1 × 10⁻⁷ mol. el⁻¹.

It was observed that leaving the W filament hot for about 80 minutes resulted in an increase of nH₂ by about a factor of 6 over the previous measurement. After electron scrubbing to return to the initial n value the filament was left off for 15 hours and this time the value of nH₂ increased by only a factor of 1.4, the base pressure being similar in both cases. It is well known that H₂ may be dissociated at an appreciable rate at the surface of W whose temperature is > 1000°K forming reactive atomic hydrogen which may be readily adsorbed on any neighboring surface. It is this process which may be taking place in our system where the W surface is continually replenished with atomic hydrogen which in turn forms molecular hydrogen and gives rise to an artificially high nH₂!

Upon exposure of this clean surface to air a drastic recontamination takes place with nH₂ = 1.8 × 10⁻¹ mol. el⁻¹, nCO₂ = 1.2 × 10⁻¹ mol. el⁻¹, nCO = 5.1 × 10⁻² mol. el⁻¹ and nCH₄ = 4.1 × 10⁻³ mol. el⁻¹. Compared to the as received state, a memory effect is observed but a sequence of further discharge cleaning and exposure to air always resulted in a recontamination to this same level. Thus, after each exposure to air, the system must be redischarged to regain the low desorption coefficients.

An argon discharge after this exposure to air and without baking the system once more resulted in a clean surface with nH₂ = 2.7 × 10⁻⁴ mol. el⁻¹, nCH₄ = 2.6 × 10⁻⁶ mol. el⁻¹, nCO₂ = 2.1 × 10⁻⁶ mol. el⁻¹ and nCO = 2.3 × 10⁻⁷ mol. el⁻¹.

The desorption coefficients were further reduced after bombarding with 39 mA at 300 eV for 22 hours (3.5 × 10⁻⁶ A cm⁻²) resulting in nH₂ = 6.9 × 10⁻⁶ mol. el⁻¹, nCH₄ = 4.2 × 10⁻⁷ mol. el⁻¹, nCO₂ = 2.5 × 10⁻⁷ mol. el⁻¹ and nCO < 1 × 10⁻⁷ mol. el⁻¹.

It is interesting to calculate the self cleaning time of PETRA to see if discharge cleaning is really a worthwhile exercise. Laboratory measurements have shown that the desorption coefficient η (for CO on undischarged surfaces) may be written as

\[ η = 4.83 \times 10^{-7} \times D^{-0.63} \]

where D is the electron dose in A hours cm⁻². If we consider always 1 metre of PETRA chambre, this expression may be written as

\[ η = 7 \times 10^{-5} \left( \frac{I \times t}{A} \right)^{-0.63} \tag{5} \]

where I is the time in hours and \( I \) is the total desorbing photoelectron current per metre of chamber in amps. The total desorbing photoelectron current is the total calculated photoelectron current multiplied by two to account for desorption on leaving and re-entering the chamber wall. From equation (2) and assuming only CO is involved

\[ η = \frac{5.23 \times S \times ΔP}{A} \tag{6} \]

where S is the pumping speed in l s⁻¹ and A is the total desorbing photoelectron current in l s⁻¹ in amps and ΔP is the rise in pressure in torr.

For CO S = 120 l s⁻¹. Therefore equation (6) becomes

\[ η = \frac{6.276 \times 10^{2} \times ΔP}{A} \tag{7} \]
Allowing a ΔP_max of 1 x 10^{-8} torr of CO with a desorbing photoelectron current of 0.5 A m^{-1} we get from equation (7) the following condition for \( n \)

\[
 n < 1.25 \times 10^{-5} \text{ mol. electron}^{-1}
\]

To clean the PETRA vacuum chamber to this value an electron beam at the injection energy of 7 GeV can be used. Under these conditions the beam current is \( \approx 30 \text{ mA} \) and the desorbing photoelectron current is \( \approx 50 \text{ mA m}^{-1} \). By rearranging equation (3) the cleaning time \( t \) is given by

\[
 t = \frac{2.55 \times 10^{-7}}{1.587}
\]

Inserting \( I = 50 \text{ mA} \) and \( n = 1.25 \times 10^{-5} \text{ mol. electron}^{-1} \) into this expression, a cleaning time of 308 hours (13 days) is obtained. Thus, to sufficiently clean the vacuum chamber the machine must be run continuously for about 2 weeks.

We have shown that an in situ pure argon discharge in either a baked or unbaked Al chamber can easily reduce the desorption coefficients for CH_{4}, CO and CO_{2} to the low \( 10^{-6} \) or \( 10^{-7} \) mol. electron\(^{-1} \) range. Since the high H_{2} desorption coefficients are an artefact of our measuring procedure it is anticipated that they too, after discharge, are in the \( 10^{-6} \) to \( 10^{-7} \) mol. electron\(^{-1} \) range.

In the laboratory the discharge was run from the central electrode, such a system being of course excluded in an actual machine. A possible solution exists in using the anodes of the distributed ion pumps where, by putting \( \pm 350 \text{ V} \) on the anodes and by judicious regulation of the argon pressure, the glow discharge may propagate throughout the whole magnet vacuum chamber. Suitable protection for the pump anode supporting insulators should ensure that no shorting occurs due to coating by sputtered metal.

The ion dose required to achieve desorption coefficients \( \approx 10^{-7} \) mol. electron\(^{-1} \) is about 1 A.h m\(^{-2} \) of chamber. Since discharge currents \( \approx 3 \) to 4 amps are easily produced, a 5 m magnet chamber may be cleaned in about one hour. In addition, any number of magnet chambers may be cleaned in parallel depending on the number of power supplies available. An additional advantage of the discharge is that the power dissipated in the chamber causes heating. For example, in the insulated, uncleared laboratory test chamber 0.5 Ar m\(^{-1} \) resulted in a temperature rise of \( \approx 80^\circ \text{C} \), thus simultaneously cleaning and baking the system.

Conclusions

The photoelectron current in PETRA was calculated to be 250 mA m\(^{-1} \) with two 100 mA beams at 14 GeV.

This photoelectron current desorbs gas from the chamber walls resulting in pressure increase. To keep these pressure increases within reasonable bounds, desorption coefficients \( < 10^{-5} \) mol. electron have to be achieved.

Relying on machine self-cleaning to reach these low desorption coefficients means a continuous running time of \( \approx 2 \) weeks at the injection energy of 7 GeV.

Laboratory experiments have shown that in situ argon glow discharge cleaning can easily produce a surface with desorption coefficients in the range \( 10^{-6} \) to \( 10^{-7} \) mol. electron\(^{-1} \) after an ion dose of 1 A.h m\(^{-2} \) = \( \approx 8 \times 10^{18} \) ions cm\(^{-2} \). In addition, because of the power dissipated in the chamber it is baked.

After the discharge the residual gas composition in the prototype PETRA Al chamber was typically 90% H_{2}, 9.1% H_{2}O, CO and CO_{2} and 1.7% Ar with the relative concentration of H_{2}O, CO, CO_{2} and Ar lending to decrease after several days pumping.
To discharge clean in situ actual magnet chambers mounted in the machine, it is proposed that the discharge be run from the anodes of the distributed sputter ion pumps whence, via the pumping ports, it may propagate throughout the whole vacuum chamber.

References:

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Acknowledgements

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<table>
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Appendix A

The cleaning procedure for the Al chamber consisted of the following steps:

1) Cleaning in detergent solution (5% VR 9518 - Henkel Co.) at 60°C.
2) Rinsing in demineralized water.
3) Immersion in 6% NaOH at 60°C for 5 minutes.
4) Rinsing in demineralized water.
5) Immersion in 15% HNO₃ at 20°C for 5 minutes.
6) Rinsing with demineralized water at 96°C.
7) Drying in air.

![Diagram of beam direction and position of magnets](image-url)
Fig. 2 The normal incidence photoelectric yield in electrons per absorbed photon for Al and Al₂O₃ for photon energies between 7 and 10⁶ eV. The curve was composed from the following references:
A (refs. 7) and 8), B (refs. 7) and 9), C (refs. 9) and 10),
D (ref. 11), E (refs. 12) and 13) and F (ref. 13).

Fig. 3 The linear absorption coefficient u/cm⁻¹ for Al between 1 and 10⁶ eV. The curve is composed from measurements from the following references:
A (ref. 14), B (ref. 15), C (ref. 16),
D (ref. 17) and E (ref. 18).
Fig. 4 The spectral distribution of the synchrotron radiation computed from equation (1) for photon energies between $10^2$ and $10^6$ eV for various electron energies.

Fig. 5 The total beam current (electrons plus positrons) in PETRA as a function of energy.
Fig. 6 The total number of photoelectrons (from positron and electron synchrotron radiation) in mA per metre of chamber for synchrotron radiation incident at $20 \times 10^{-3}$ radians glancing angle as a function of energy.
Fig. 8 The calculated rise in absolute pressure in PETRA as a function of energy for various $n$ values.

Fig. 9 Schematic diagram of the electron desorption equipment.
Fig. 10. The desorption coefficient $n$ for $H_2$, $CH_4$, CO and $CO_2$ as a function of the treatment of the chamber.