Tritium migration along the cryopumping section

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The transport section of the Karlsruhe Tritium Neutrino experiment (KATRIN) must provide the dramatic reduction of tritium flow and gas density from the end of a 10-m-long windowless gaseous tritium source throughout several stages of a differential pumping system. The final stage of this section, the cryogenic pumping section (CPS) based on pumping of tritium on argon frost at 4.5 K, should provide the flow ratio between inlet and outlet in the range of 10^7. Cryosorbed tritium may decay, emitting a few keV electrons. These electrons in their turn cause the electron-stimulated desorption of cryosorbed argon and tritium, which is redistributed along the CPS (migration process). This effect was modeled with the use of the method of angular coefficients. The main result is that the tritium migration process does not affect the CPS performance at KATRIN for a given inlet flow. Meanwhile, if the flow chosen is larger, the migration effect could be dominant. © *2008 American Vacuum Society.* [DOI: 10.1116/1.2816945]

I. INTRODUCTION

The Karlsruhe Tritium Neutrino experiment (KATRIN) is a large vacuum system and aims to measure the electron neutrino mass from the β decay of tritium with unprecedented sensitivity.[1,2] To achieve this purpose, the tritium gas flow has to be significantly reduced along the transport line by means of a modular differential pumping system. A detailed description of the KATRIN vacuum system, its requirements, and its challenges can be found elsewhere.[2] The final element of the transport line is the cryogenic pumping section (CPS), where tritium is pumped on argon frost. The difficulty of predicting the behavior of this section is related to tritium radioactivity: when cryosorbed tritium decays it emits a few keV electrons,[2] which cause desorption and readsorption of argon and, what is most of interest, tritium, i.e., even when tritium molecules are cryosorbed with high sticking probability and some large sojourn time, there is still a possibility that they might be desorbed due to the decay of neighbor tritium molecules. This process is called the tritium migration process.

Available test particle Monte Carlo programs allow modeling of a complicated vacuum chamber, but do not allow studying time dependent processes like tritium migration. The aim of the present work was to study on a simple tubular model whether the tritium migration is significant in the KATRIN CPS and should be included in a full model (i.e., writing a special program) or if it can be neglected and modeling with available test particle Monte Carlo programs will provide accurate results.

II. METHOD OF ANGULAR COEFFICIENT

Consider a tubular vacuum chamber of diameter d and length L with sorbing walls with sticking probability α. The tube is placed between two large volumes with gas densities n_1 and n_2 ($n_1 \gg n_2$). The numerical model of the inner part of the tube consists of inlet and outlet disks and N rings as shown in Fig. 1. A longitudinal nondimensional coordinate $\chi = l/d$ is used in the model. A nondimensional ring length is $\mathsf{s} = \Delta l/d$.

The transmission probabilities between different elements can be described in the method of angular coefficient as the following:[3]

(1) The probability φ_{DD} for a molecule from one disk to reach another co-axial disk of the same diameter on distance χ is described as

$$\varphi_{DD}(\chi) = (\sqrt{\chi^2 + 1} - \chi)^2. \tag{1}$$

(2) The probability φ_{RD} for a molecule from the inner part of a ring of length ζ to reach a co-axial disk of the same diameter on the distance χ is

$$\varphi_{RD}(\chi) = \frac{\chi^2 + 0.5}{\sqrt{\chi^2 + 1}} - \chi. \tag{2}$$

(3) The probability φ_{DR} from a disk to a ring is

$$\varphi_{DR}(\chi) = 4\mathsf{s}\ \varphi_{RD}(\chi). \tag{3}$$

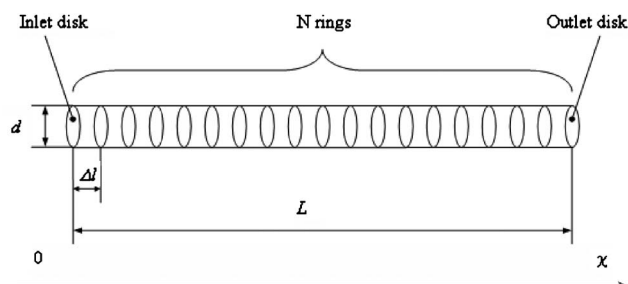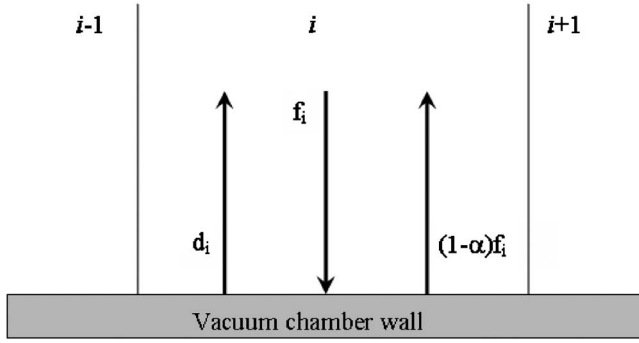(4) The probability φ_{RR} from a ring to a ring is

FIG. 1. Model layout.

FIG. 2. Incoming and outgoing gas flow to the surface i.

FIG. 3. Calculation of electron-stimulated desorption of tritium.

$$\varphi_{RR}(\chi) = \text{s}\left[1 - \frac{\chi(\chi^2 + 1.5)}{(\chi^2 + 1)^{1.5}}\right]. \tag{4}$$

The transmission probability matrix $W[N+2, N+2]$ of the model is described as the following:

$$w_{i,j} = \begin{cases} w_{0,0} = w_{N+1,\ N+1} = 0, \\ w_{N+1,0} = w_{0,N+1} = \varphi_{DD}(L/d), \\ w_{i,0} = w_{N+1-i,\ N+1} = \varphi_{DR}((j-0.5)\text{s}) \quad \text{for } i = 1, \dots, N, \\ w_{0,j} = w_{N+1,\ N+1-j} = \varphi_{RD}((j-0.5)\text{s}) \quad \text{for } i = 1, \dots, N, \\ w_{i,j} = \varphi_{DR}(|i-j|\text{s}) \quad \text{for } i, j = 1, \dots, N. \end{cases} \tag{5}$$

The sticking probability vector is

$$\alpha_i = \begin{cases} 1 & \text{for } i = 0 \text{ and } i = N+1, \\ \alpha & \text{for } i = 1, \dots, N. \end{cases} \tag{6}$$

III. CALCULATION OF FLOW RATE INITIAL REDUCTION FACTOR

These calculations can be done in the same way as it is described in Refs. 4–7 but applied in some different way; therefore, it is described below.

In general, the gas flow through the surface i, Q_i, consists of three parts: oncoming flow f_i and two outgoing parts (see Fig. 2): desorbed (or injected) part, d_i, and reflected part, $(1 - \alpha_i)f_i$:

$$Q_i = d_i - \alpha_i f_i, \quad i = 0, \dots, N+1. \tag{7}$$

The flux f_i of molecules arriving to the ith surface is equal to

$$f_i = \sum_j w_{i,j} q_j, \quad i, j = 0, \dots, N+1, \tag{8}$$

where q_j is a flux of molecules from the jth pumping surface: $q_j = d_j + (1 - \alpha_j)f_j$. Then Eq. (7) can be rewritten as

$$f_i = \sum_j w_{i,j}(d_j + (1 - \alpha_j)f_j), \quad i, j = 0, \dots, N+1,$$
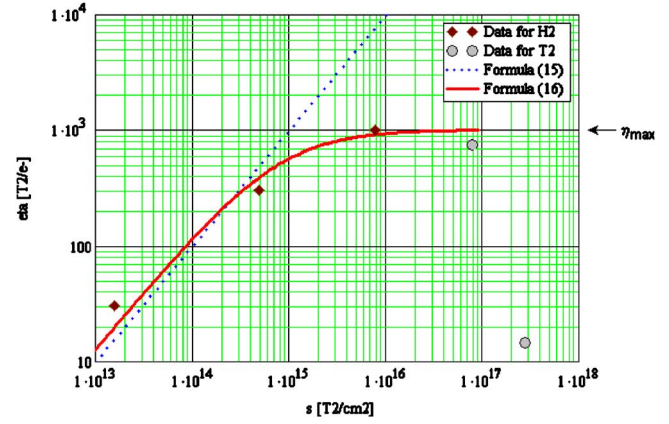
or

$$f_i - \sum_j w_{i,j}(1 - \alpha_j)f_j = \sum_j w_{i,j}d_j. \tag{9}$$

When desorption and sticking probability are defined, then this is a system of $N+2$ linear equations for $N+2$ unknowns f_i, which can be rewritten in matrix form as

$$(\mathbf{E} - \mathbf{W} \cdot \text{diag}(1 - \boldsymbol{\alpha})) \cdot \mathbf{f} = \mathbf{W} \cdot \mathbf{d}, \tag{10}$$

where $\mathbf{E}$ is an identity matrix, $\mathbf{W}$ is a matrix of the transmission probabilities defined above, $\text{diag}(1 - \boldsymbol{\alpha})$ is a diagonal matrix element of vector $(1 - \alpha)$, $\mathbf{f}$ is a vector of arriving flows, and d is a vector of desorption. Solving the equation type $\mathbf{A} \cdot \mathbf{x} = \mathbf{b}$ for $\mathbf{x}$ is a built-in function in many available math packages, for example in MathCAD. Having a solution for fluxes f_i for the given distribution of gas sources d_i, the gas flow Q_i at every surface i can be calculated with formula (7).

In the case of the CPS, initially the only desorbing surface is the inlet surface: $d_0 \neq 0$ and $d_i = 0$ for $i = 1, \dots, N+1$. Then the *section transmission probability* is equal to

$$T = \frac{f_{N+1}}{d_0}. \tag{11}$$

The amount of tritium entering the CPS during time period Δt is equal to $Q\Delta t$, where Q is the tritium flux entering the CPS. This gas is cryosorbed on an argon frost with surface coverage distribution s_i, which can be calculated from a solution for f_i as the following:

$$s_i\left[\frac{T_2}{m^2}\right] = \frac{\alpha_i f_i}{A_i}, \quad i = 1, \dots, N, \tag{12}$$

where A_i is the area of ith ring. The number of molecules cryosorbed on each ring is

$$n_i[T_2] = \alpha_i f_i, \quad i = 1, \dots, N. \tag{13}$$

IV. A MODEL FOR CALCULATION OF A FLOW RATE REDUCTION FACTOR CONSIDERING A TRITIUM MIGRATION EFFECT

The cryosorbed tritium molecules must have sufficient bounding energy that equilibrium gas density and therefore
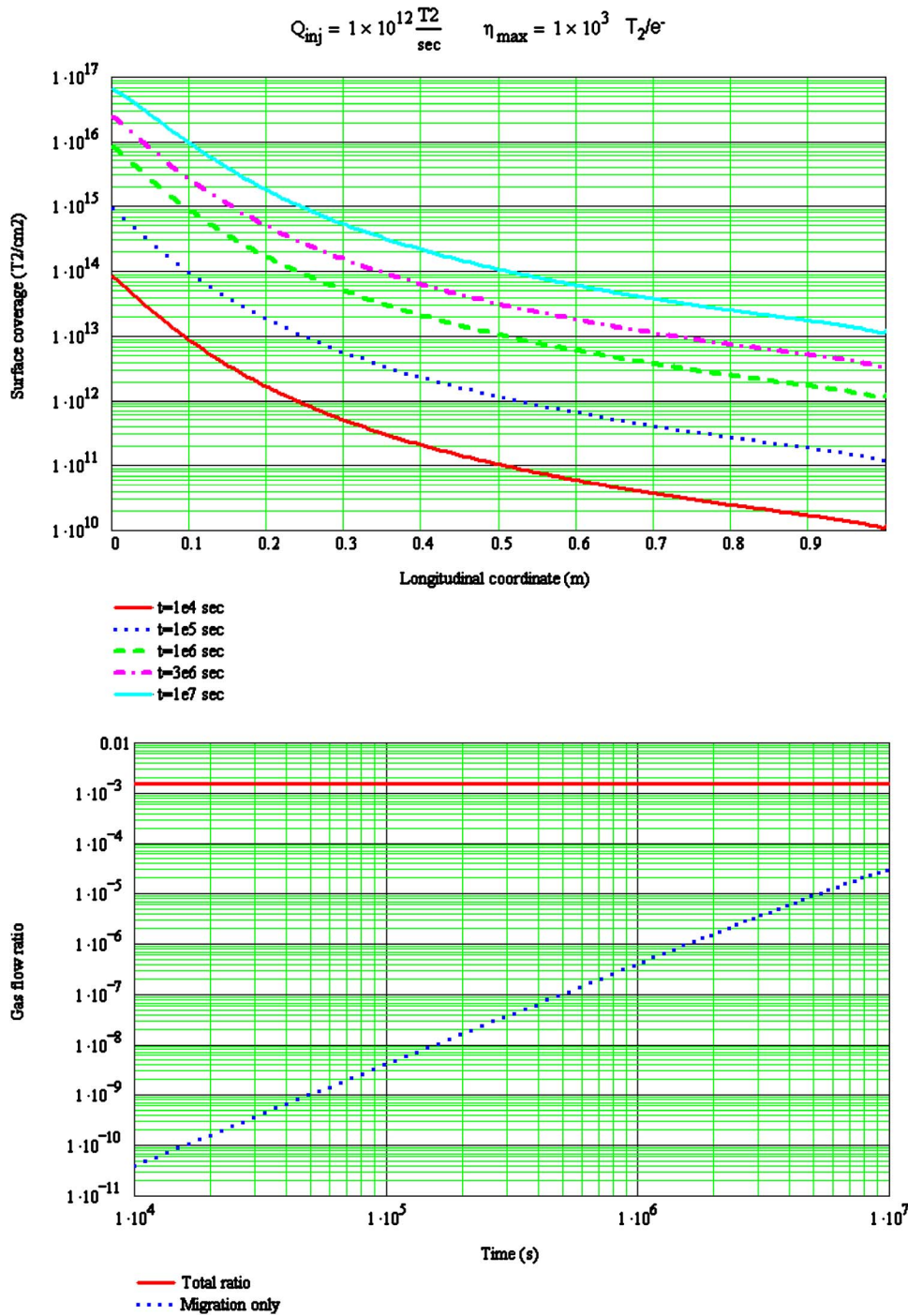
$$Q_{inj} = 1 \times 10^{12} \frac{T2}{sec} \qquad \eta_{max} = 1 \times 10^3 \ T_2/e^-$$

FIG. 4. Surface coating as a function of longitudinal coordinate and time (upper graph) and gas flow ratio (bottom graph) for $\eta = 10^3$ T_2/e^- and inlet flow $Q = 10^{12}$ T_2/s.

tritium thermal desorption and readsorption can be neglected. Meanwhile, some of these molecules will decay, emitting a few keV electrons and a few eV beta particles, which will move along the solenoid magnetic field lines causing desorption of argon and tritium. The number of decays during time Δt on each ring can be calculated as

$$\Gamma_i = n_i(1 - 2^{-\Delta t/\tau}) = \alpha_i f_i(1 - 2^{-\Delta t/\tau}), \quad i = 1, \dots, N. \quad (14)$$

There is a quite limited number of publications on electron-stimulated desorption from cryosorbed gases and mixtures but all at normal incident angle. The most relevant is Ref. 8, where the desorption yields are reported for argon and argon-hydrogen mixture. The desorption yield of argon depends on the surface coverage and electron energy with highest measured value of ~ 20 Ar/e^-. The desorption yield of hydrogen depends on the percentage of hydrogen in argon-hydrogen mixture linearly between 0.8% and 25% of H_2 and not changing between 25% and 100% of H_2 with the highest measured value of $\sim 10^3$ H_2/e^- (at electron energy of 300 eV). The desorption yield of hydrogen reduces for layers thicker than 100 monolayers of H_2. This result is shown in Fig. 3 with diamonds. Ion-induced desorption at the eV energy range is much less than these values (see, for example,
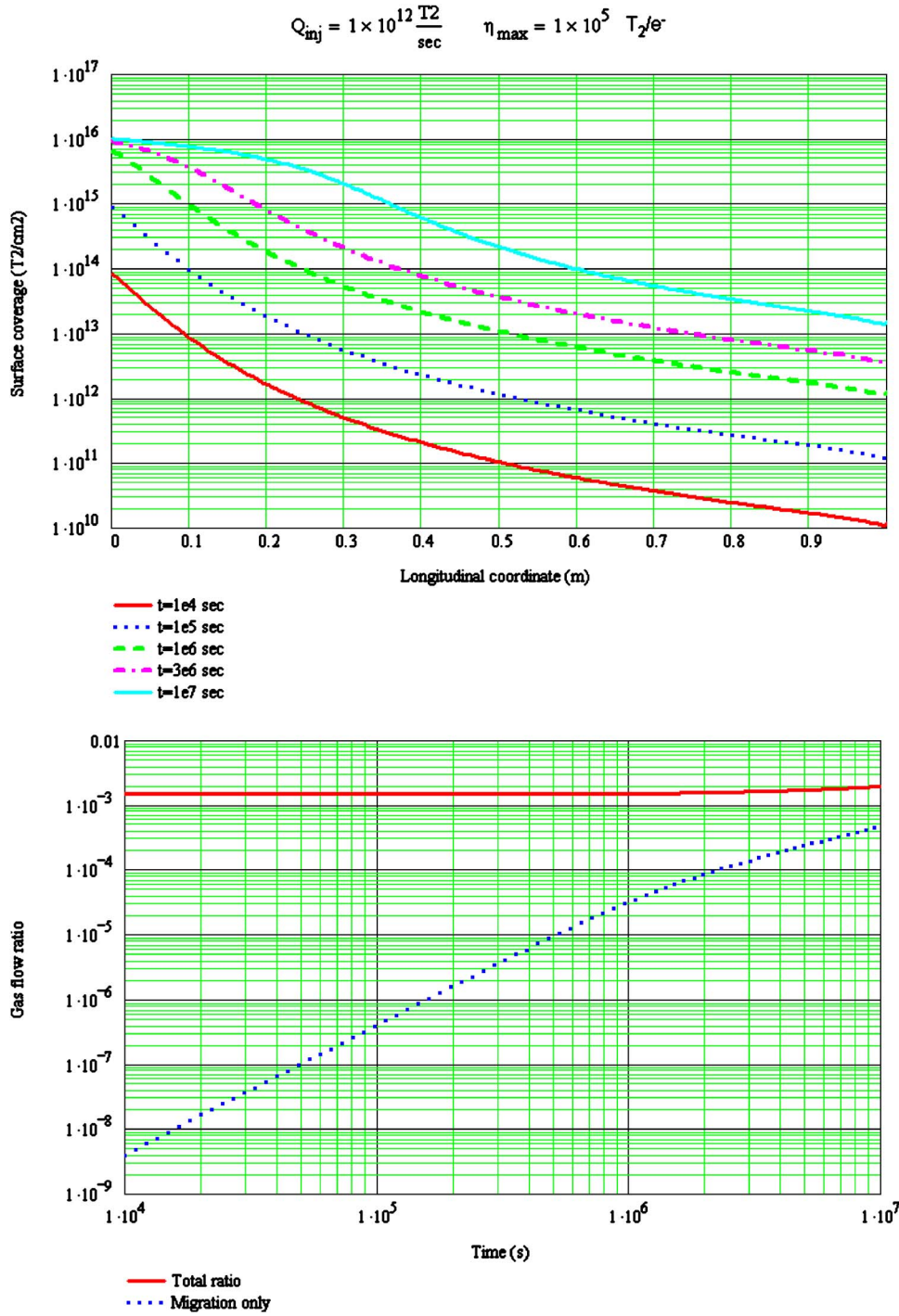
$$Q_{inj} = 1 \times 10^{12} \frac{T2}{sec} \qquad \eta_{max} = 1 \times 10^5 \ T_2/e^-$$

FIG. 5. Surface coating as a function of longitudinal coordinate and time (upper graph) and gas flow ratio (bottom graph) for $\eta = 10^5$ T_2/e^- and inlet flow $Q = 10^{12}$ T_2/s.

Refs. 9 and 10) and can be considered as negligible compared to the electron-stimulated desorption.

The upper limit of electron-stimulated desorption from argon frost can be calculated also from the average electron energy of about 6 keV per tritium decay, assuming all energy transferred to cryodeposit. If the energy is mainly transferred to the adsorbent, with a sublimation energy for solid argon of $E_{subl}(\text{Ar}) = 8048$ J/mol,[11] up to 60 000 Ar atoms per tritium decay could be evaporated. Meanwhile, the sublimation energy for both hydrogen and tritium sorbed on argon should be much less, ~0.5 to 2 kJ/mol, and strongly depends on

temperature and surface coverage, therefore one may expect that the amount of desorbed hydrogen and/or tritium from argon frost should be larger and can be described as

$$\eta(s_{T_2}) = 6 \times 10^4 \frac{s_{T_2}}{s_{Ar}} \frac{E_{subl}(\text{Ar})}{E_{subl}(T_2)}. \tag{15}$$

The electron-stimulated desorption yields calculated with formula (15) are shown with a blue dotted line in Fig. 3 for the ratio $E_{subl}(\text{Ar})/E_{subl}(T_2) = 8$, which is a sensible approximal value for the temperature of about 4.2 to 4.5 K. Having
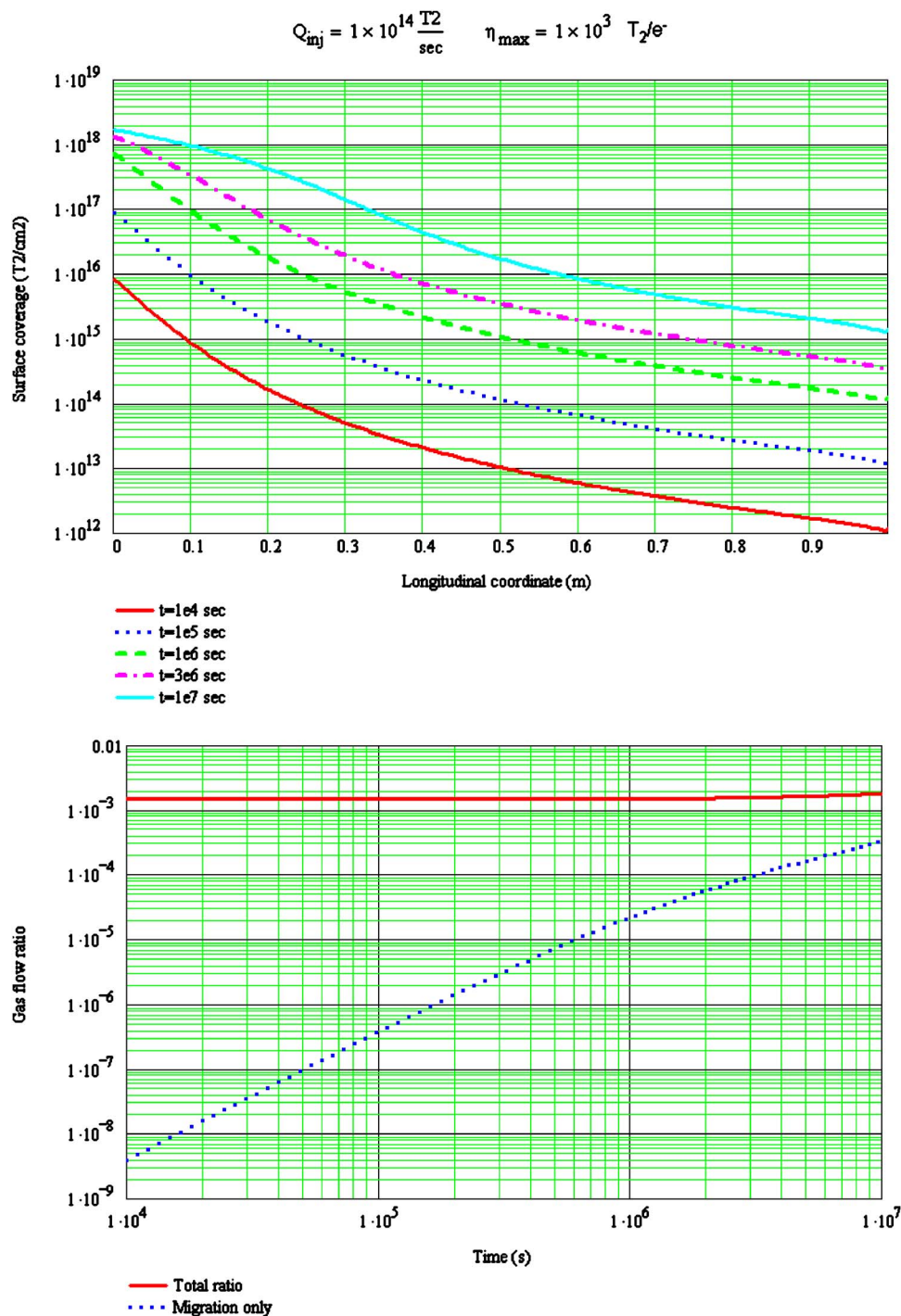
$$Q_{inj} = 1 \times 10^{14} \frac{T2}{sec} \qquad \eta_{max} = 1 \times 10^3 \; T_2/e^-$$

FIG. 6. Surface coating as a function of longitudinal coordinate and time (upper graph) and gas flow ratio (bottom graph) for $\eta = 10^3 \; T_2/e^-$ and inlet flow $Q = 10^{14} \; T_2/s$.

an approximate character of this curve, the match of the data and the curve in Fig. 3 is reasonable for the T_2 surface coverage less than a monolayer, but not for the higher T_2 surface coverage.

Other useful results were obtained during the neutrino mass experiments at Mainz University. One week of effective lifetime was reported[12] for 40-monolayer tritium condensate on an aluminum substrate at 2.8 K. This gives a desorption rate of 750 T_2 per decay. In a later publication,[13]

one year of effective lifetime was reported for a 140-monolayer tritium condensate on a graphite substrate at 1.9 K, which gives the desorption rate of 15 T_2 per decay. There are no data for tritium and hydrogen that lead to the expectation of the maximum tritium desorption yield much larger than $10^3 \; T_2$ molecules/decay.

Following this, the tritium desorption yield can be described as a function of T_2 surface coverage s as the following:

$$Q_{inj} = 1 \times 10^{14} \frac{T2}{sec} \qquad \eta_{max} = 1 \times 10^5 \ T_2/e^-$$
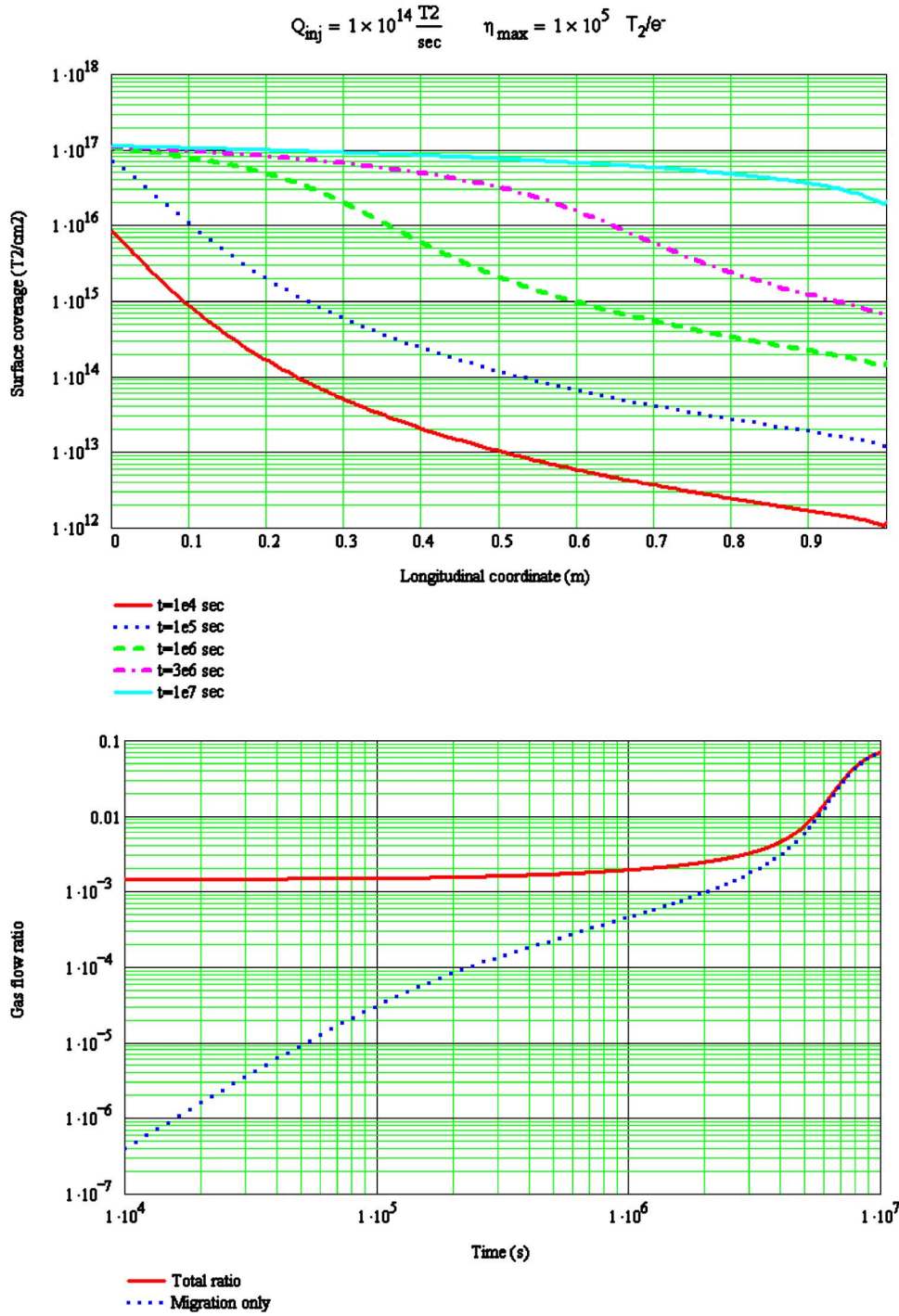

FIG. 7. Surface coating as a function of longitudinal coordinate and time (upper graph) and gas flow ratio (bottom graph) for $\eta = 10^5 \ T_2/e^-$ and inlet flow $Q = 10^{14} \ T_2/s$.

Legend (upper graph):
- t=1e4 sec
- t=1e5 sec
- t=1e6 sec
- t=3e6 sec
- t=1e7 sec

Legend (lower graph):
- Total ratio
- Migration only

$$\eta(s) = \eta_{max} \frac{s}{s + s_m}, \tag{16}$$

where s_m is a T_2 surface coverage corresponding to the end of the linear growth of the tritium desorption yield $s_m \approx 4 \times 10^{14} \ T_2/cm^2$. The graph calculated with formula (16) for $\eta_{max} = 10^3 \ T_2/decay$ is shown by a solid red line in Fig. 3. This formula is used in the following analysis because the coverage higher than $10^{17} \ T_2/cm^2$ is not expected.

The number of molecules desorbed during time Δt can be calculated for each ring as

$$d_i = \eta(s_i)\Gamma_i = \eta_{max} \frac{\alpha_i f_i}{1 + \dfrac{s_m A_i}{\alpha_i f_i}}(1 - 2^{-\Delta t/\tau}), \quad i = 1, \dots, N. \tag{17}$$

For studying the migration process as a function of time, two simultaneous processes are treated in the calculation model as two discrete processes happening in turn with each time interval Δt:
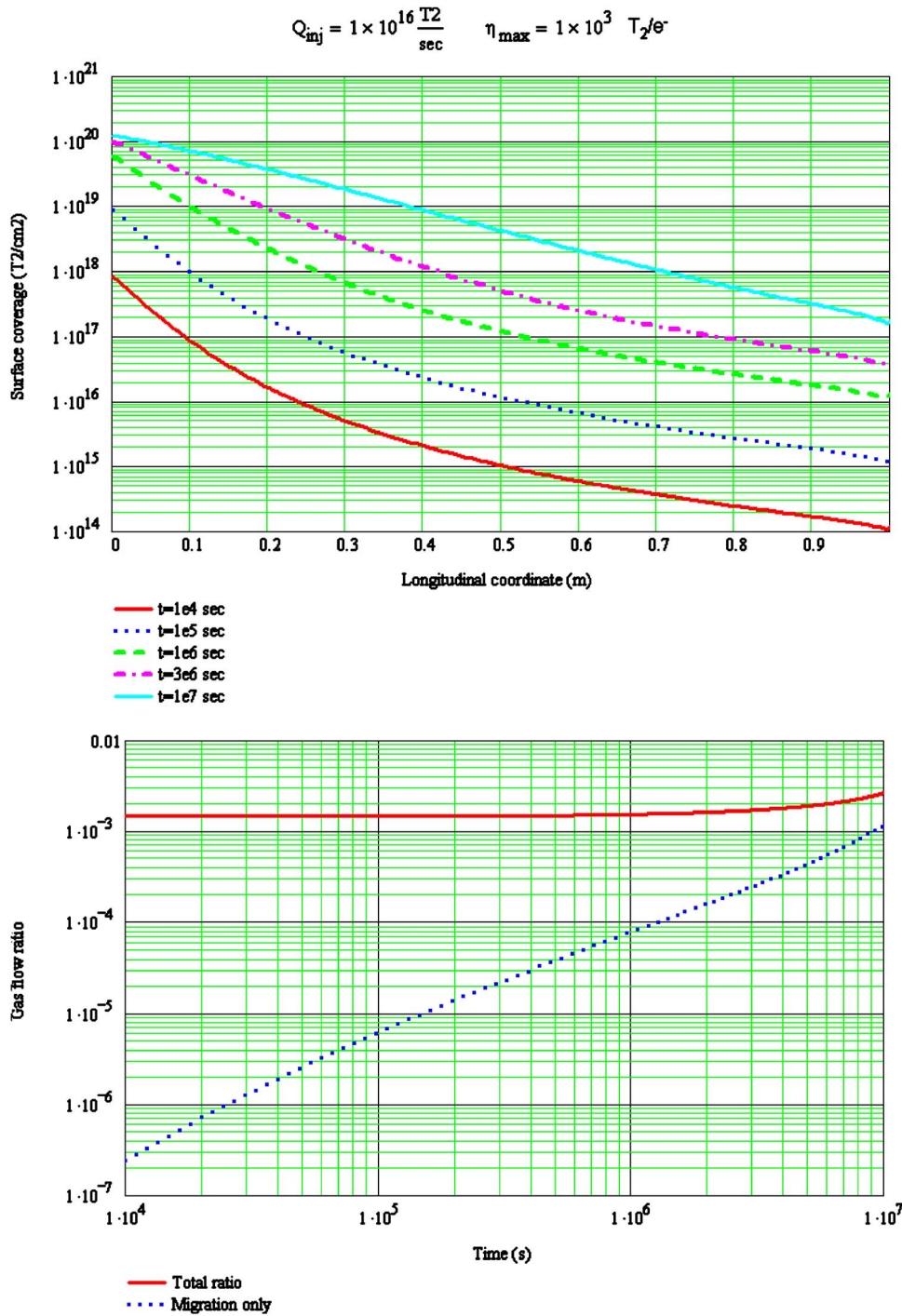
$$Q_{inj} = 1 \times 10^{16} \frac{T2}{sec} \qquad \eta_{max} = 1 \times 10^3 \ T_2/e^-$$

FIG. 8. Surface coating as a function of longitudinal coordinate and time (upper graph) and gas flow ratio (bottom graph) for $\eta = 10^3$ T_2/e^- and inlet flow $Q = 10^{16}$ T_2/s.

- Gas injection, when the amount of tritium gas entered into the CPS during time between t_0 and $t_0 + \Delta t$, is distributed following solution for f_i from Eq. (10), which gives surface coverage distribution s_i and the number of decays during time Δt on each ring.
- Tritium decay and electron-stimulated desorption. Assuming that all decays happen simultaneously at the end of time period $t = t_0 + \Delta t$, the number of molecules desorbed due to the tritium decays is calculated with formula (17). This gives a new desorption vector $\mathbf{d}'$ for Eq. (10) and a new solution $\mathbf{f}'$.

The final surface coverage distribution s_i' at the end of time period $t = t_0 + \Delta t$ is calculated as the following:

$$n_i' = n_i - \Gamma_i - d_i' + \alpha_i f_i',$$

$$s_i' = \frac{n_i'}{A_i}, \quad i = 1, \ldots, N. \tag{18}$$

In the next time periods, the iteration cycles are described as follows:

Given inlet flow Q,

$$\mathbf{d}_0 = \begin{cases} d_{0,0} = Q \; \Delta t, \\ d_{0,j} = 0 \quad \text{for} \; j = 1, \ldots, N+1. \end{cases}$$

Solving $(\mathbf{E} - \mathbf{W} \times \text{diag}(1-\boldsymbol{\alpha})) \times \mathbf{f}_0 = \mathbf{W} \times \mathbf{d}_0$,

$$\mathbf{n}_0 = \text{diag}(\boldsymbol{\alpha}) \times \mathbf{f}_0, \quad \mathbf{s}_0 = \frac{\mathbf{n}_0}{A},$$

for $k = 1, \ldots, M$,

$$\boldsymbol{\Gamma}_k = \mathbf{n}_{k-1}(1 - 2^{-\Delta t/\tau}) = \text{diag}(\boldsymbol{\alpha}) \times \mathbf{f}_{k-1}(1 - 2^{-\Delta t/\tau}),$$

$$\mathbf{d}_k = \begin{cases} d_{k,j} = 0 \quad \text{for} \; j = 0 \; \text{and} \; j = N+1, \\ d_{k,j} = \eta(s_{i-1,j})\Gamma_{k,j} = \eta_{\max} \dfrac{n_{k-1,j}}{1 + \dfrac{s_m A}{n_{k-1,j}}}(1 - 2^{-\Delta t/\tau}) \quad \text{for} \; j = 1, \ldots, N+1. \end{cases}$$

Solving $(\mathbf{E} - \mathbf{W} \times \text{diag}(1-\boldsymbol{\alpha})) \times \mathbf{f}_k = \mathbf{W} \times \mathbf{d}_k$,

$$\mathbf{n}_k = \mathbf{n}_0 + \mathbf{n}_{k-1} - \boldsymbol{\Gamma}_k - \mathbf{d}_k + \boldsymbol{\alpha} \times \mathbf{f}_k,$$

$$\mathbf{s}_k = \frac{\mathbf{n}_k}{A}. \tag{19}$$

Here index k corresponds to the iteration number.

V. RESULTS OF CALCULATIONS

Surface coating as a function of longitudinal coordinate and time and a gas flow ratio were calculated for a tube with the following parameters: $d = 75$ mm, $L = 1$ m, and the tritium sticking probability $\alpha = 0.7$. The electron-stimulated desorption of tritium was calculated with formula (16) with two different values of the main parameter in the formula:

1. $\eta_{\max} = 10^3 \; T_2/e^-$ (as shown in Fig. 3), the same as it was measured for hydrogen, and
2. $\eta_{\max} = 10^5 \; T_2/e^-$ to study what can happen if $\eta_{\max}$ is underestimated for some unknown reason.

The inlet tritium flow was taken as $Q = 10^{12} \; T_2/s$. This value was calculated from injected flow at WGTS $Q_{inj} = 1.8$ mbar l/s and a flow rate for DPS1-F and DPS2-F of about 3×10^{-8}. The developing tritium surface coverage along the tube and the tube flow ratio were studied as a function of time up to 10^7 s (115 days, which is reasonable time between warming up the cryostat) and shown in Figs. 4 and 5. One can see that although the surface coating distribution depends on $\eta_{\max}$, the CPS gas flow ratio is almost insensitive to $\eta_{\max}$ in the range 10^3 to $10^5 \; T_2/e^-$ for given geometry and injected gas flow $Q = 10^{12} \; T_2/s$.

The reasonable question is: At what inlet tritium flow is the migration effect significant?

In the case of one turbo-molecular pump failure at DPS1-F or DPS2-F, the inlet flow may increase up to about 30 times. Results of calculations for the inlet tritium flow of $Q = 10^{14} \; T_2/s$ are shown in Figs. 6 and 7. In this case, the migration effect is negligible for $\eta_{\max} = 10^3 \; T_2/e^-$. Mean-

while, the gas flow ratio for $\eta_{\max} = 10^5 \; T_2/e^-$ becomes sensitive to this effect after about 10 days of continuous injection; it changes ten times after ~ 70 days of continuous injection.

In the case of significant failure at DPS1-F or DPS2-F, the inlet flow may increase even higher. Results of calculations for the inlet tritium flow of $Q = 10^{16} \; T_2/s$ are shown in Figs. 8 and 9. In this case, the migration effect should be considered after ~ 100 days for $\eta_{\max} = 10^3 \; T_2/e^-$ and after ~ 2 days of continuous injection for $\eta_{\max} = 10^5 \; T_2/e^-$; then the gas flow ratio changes 10 times after ~ 9 days and 70 times after ~ 30 days of continuous injection.

The results of these calculations mean the following:

- The tritium migration effect can be neglected at the nominal operation inlet tritium flow of $Q = 10^{12} \; T_2/s$.
- If inlet tritium flow increases due to a turbo-molecular pump failure or other reason up to $Q = 10^{14} \; T_2/s$, the tritium migration effect can be neglected for the expected $\eta_{\max} = 10^3 \; T_2/e^-$, or, in the worst case, if $\eta_{\max}$ is underestimated, the tritium migration effect can be neglected for 10 days for $\eta_{\max} = 10^5 \; T_2/e^-$.
- If inlet tritium flow increases due to a significant failure up to $Q = 10^{16} \; T_2/s$, the tritium migration effect can be neglected for the expected $\eta_{\max} = 10^3 \; T_2/e^-$ for about 100 days, or, in the worst case, if $\eta_{\max}$ is underestimated, the tritium migration effect can be neglected for 2 days for $\eta_{\max} = 10^5 \; T_2/e^-$.
- Since in the case of failure it is very unlikely that injection will continue longer than a few hours, then the tritium migration effect can be neglected in the CPS and the usual Monte Carlo routine with available codes can be used for modeling of the whole CPS.

VI. CONCLUSIONS

An analytical model was developed for studying the tritium migration stimulated by its radioactive decay along the cryogenic tube with an argon frost. Calculations were performed for one straight part of the CPS. Because of the lack
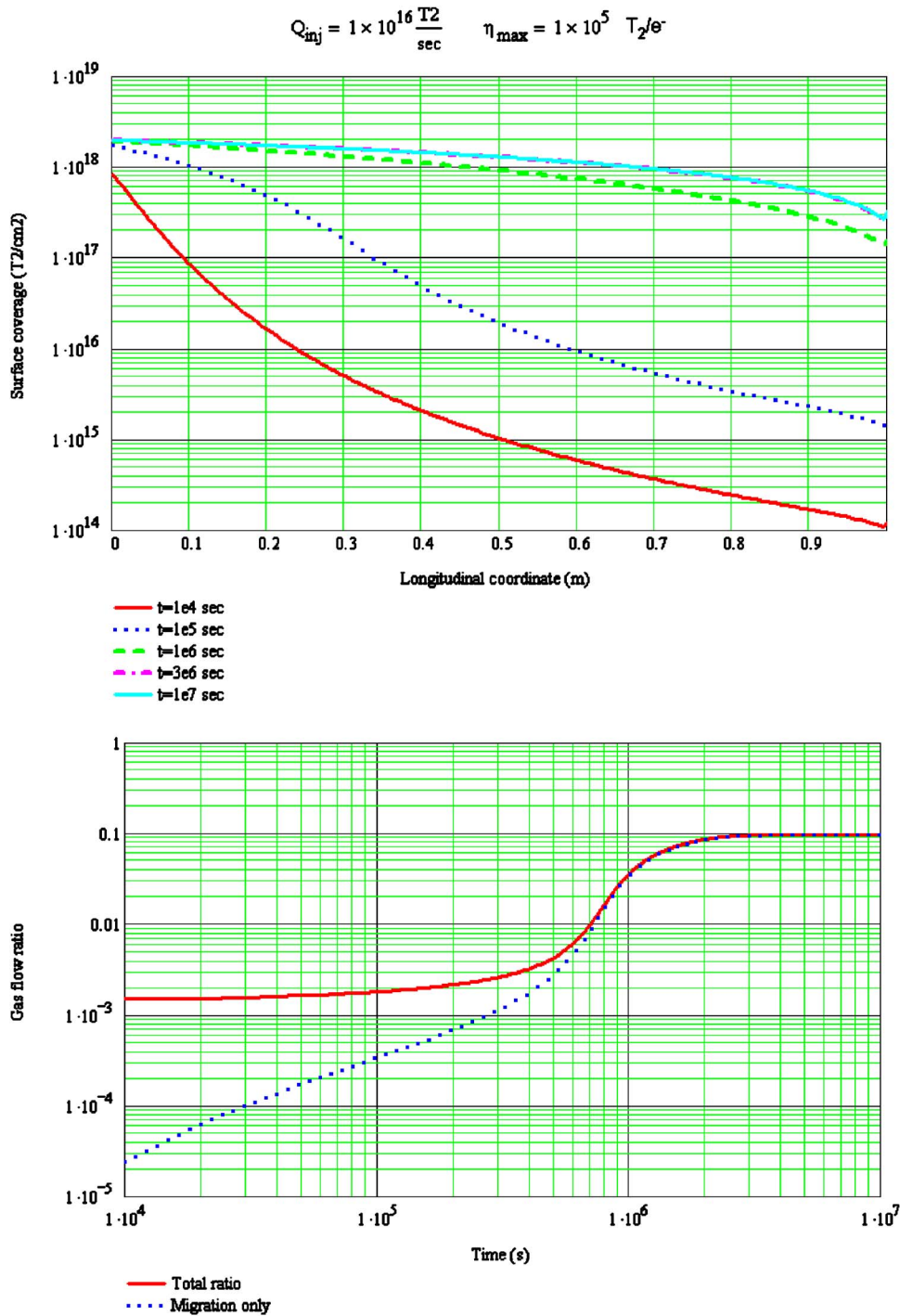
$$Q_{inj} = 1 \times 10^{16} \frac{T2}{sec} \qquad \eta_{max} = 1 \times 10^5 \; T_2/e^-$$

FIG. 9. Surface coating as a function of longitudinal coordinate and time (upper graph) and gas flow ratio (bottom graph) for $\eta = 10^5$ T_2/e^- and inlet flow $Q = 10^{16}$ T_2/s.

of experimental data, the maximum electron-stimulated desorption in the model was varied in a wide range from the likely expected 10^3 T_2/e^- to the maximum possible 10^5 T_2/e^-. The inlet flow was varied from $Q = 10^{12}$ T_2/s at normal operation to $Q = 10^{16}$ T_2/s presenting an accident case. It was found that

- The migration effect can be neglected for the calculation of CPS flow ratio for the inlet flow of $Q = 10^{12}$ T_2/s (normal operation).
- The migration effect should be considered for the calculation of CPS flow ratio for the inlet flow of $Q = 10^{14}$ T_2/s (a

case of failure of one turbo-molecular pump in an upstream differential pumping section) after 10 days of continuous injection.

- The migration effect may significantly affect the CPS flow ratio with the inlet flow of $Q = 10^{16}$ T_2/s (an accident case) after about 2 days of continuous injection.

The main conclusion is that the migration process is insignificant in all operational regimes of CPS operation and allows using the available test particle Monte Carlo programs to model the whole CPS.

It was also shown that the migration effect could be significant when a significant amount of tritium sorbed at the CPS.

ACKNOWLEDGMENTS

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