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Tritium migration along the Cryopumping section

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Abstract.

Transport section of the **Ka**rlsruhe **Tri**tium **N**eutrino experiment (KATRIN) must provide the dramatic reduction of tritium flow and gas density from the end of 10-m long windowless gaseous tritium source throughout a few stages of differential pumping system. The final stage of this section, cryogenic pumping section (CPS) base 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 re-distributed along CPS (migration process). This effect was modelled with use of method of angular coefficients. The main result is that the tritium migration process does not affect the CPS performance at KATRIN.

Keywords: Cryopumping; electron stimulated desorption; migration.

1. Introduction.

The **Ka**rlsruhe **Tri**tium **N**eutrino 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 beam transport line by means of a modular differential pumping system. A detailed description of the KATRIN vacuum system, its requirements and challenges can be found in [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 behaviour of this section related to tritium radioactivity: when cryosorbed tritium decays it emits a few keV electron, which cause desorption and re-adsorption of argon and, what is most of interest, tritium, i.e. even when tritium molecules are cryo-sorbed with high sticking probability and some large sojourn time, there is still possibility that they might be desorbed due to decay of neighbour tritium molecules, this process was called tritium migration process.

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

2. Method of angular coefficient.

Consider a tubular vacuum chamber of diameter d and length L with sorbing walls with sticking probability a. The tube placed between two large volumes with gas densities n_1 and n_2 (n_1»n_2). In the numerical model of the inner part of the tube consist of inlet and outlet disks and N rings as shown on Figure 1. A longitudinal non-dimensional co-ordinate $c = l/d$ is used in the model, a non-dimensional ring length is $V = \Delta l/d$.

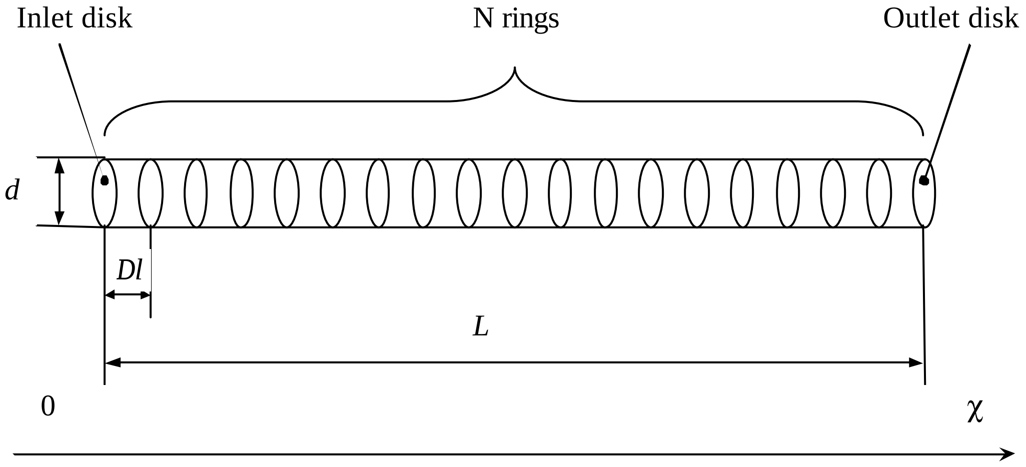

Figure 1. Model layout.

The transmission probabilities between different elements can be described in the method of angular coefficient as the following [1]:

- The probability j_{DD} for molecule from one disk to reach another co-axial disk of the same diameter on distance c is described as

$$j_{DD}(c)=\left(\sqrt{c^2+1}-c\right)^2. \tag{1}$$

- The probability j_{RD} for molecule from the inner part of a ring of length z to reach a co-axial disk of the same diameter on the distance c is:

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

- The probability j_{DR} from a disk to a ring this is:

$$j_{DR}(c)=4V\,j_{RD}(c) \tag{3}$$

where A is the vacuum chamber wall area per unit axial length, and B is a disk area.

- The probability j_{RR} from a ring to a ring this is:

$$j_{RR}\left(c\right)=V\left[1-\frac{c\left(c^{2}+1.5\right)}{\left(c^{2}+1\right)^{1.5}}\right] \qquad (4)$$

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

$$w_{i,j}=\begin{cases} w_{0,0}=w_{N+1,N+1}=0 & \\ w_{N+1,0}=w_{0,N+1}=j_{DD}\left(L/d\right) & \\ w_{i,0}=w_{N+1-i,N+1}=j_{DR}\left(\left(j-0.5\right)V\right) & \text{for } i=1...N \\ w_{0,j}=w_{N+1,\,N+1-j}=j_{RD}\left(\left(j-0.5\right)V\right) & \text{for } i=1...N \\ w_{i,j}=j_{DR}\left(\left|i-j\right|V\right) & \text{for } i,j=1...N \end{cases} \qquad (5)$$

Sticking probability vector is:

$$a_{i}=\begin{cases} 1 & \text{for } i=0 \text{ and } i=N+1 \\ a & \text{for } i=1...N \end{cases} \qquad (6)$$

3. Calculation of flow rate initial reduction factor.

These calculation can be done in the same way as it is described in [3,4,5,6] but applied in some different way, therefore it is described below.

In general, the gas flow trough the surface *i*, Q_i, consist of three part: oncoming flow f_i form all other parts of vacuum system, and two outgoing parts (see Figure 2): desorbed (or injected) part, d_i, and reflected part, $\left(1-a_{i}\right)f_{i}$:

$$Q_{i}=d_{i}-a_{i}f_{i}, \quad i=0...N+1 \qquad (7)$$

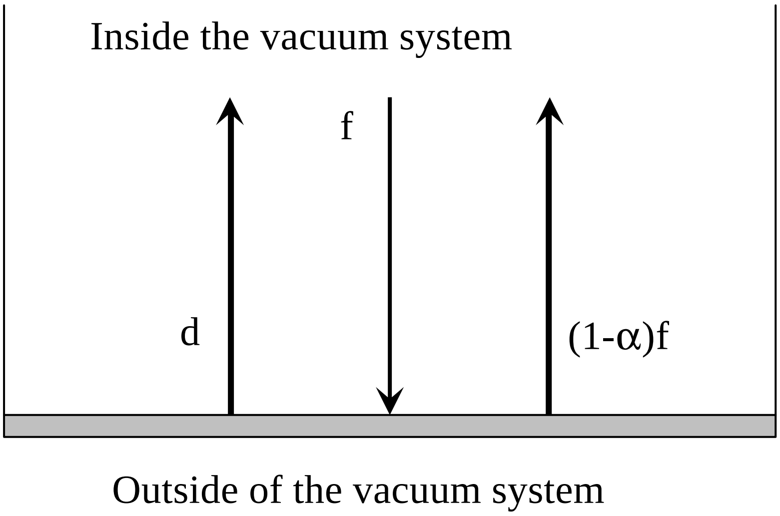

Figure 2. Incoming and outgoing gas flow to the surface

The flux *f* of molecules arriving to *i*-th surface for system is equal to:

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

Where q_j is a flux of molecules from *j*-th pumping surface: $q_j = d_j + (1-\alpha_j) f_j$. When the equation (7) can be re-written as:

$$\begin{aligned} & f_i = \sum_j w_{i,j} \left(d_j + (1-\alpha_j) f_j \right), \quad i, j = 0 \ldots N+1 \\ & or \\ & f_i - \sum_j w_{i,j} (1-\alpha_j) f_j = \sum_j w_{i,j} d_j \end{aligned} \tag{9}$$

When desorption and capture coefficient are defined, then this is a system of N+2 linear equations for N+2 unknowns f_i, which could be re-written in matrix form as:

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

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

(7).

In the case of CPS, initially there is only one desorbing surface — inlet surface: d_0?0 and d_i=0 for i=1…N+1. Then the *section transmission probability* is equal to:

$$T = \frac{f_{N+1}}{d_0}. \qquad (11)$$

Amount of tritium entering the CPS during time period *Dt* is equal to *QDt*, 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:

$$s_i\left[\frac{T_2}{m^2}\right] = \frac{a_i f_i}{A_i}, \quad i = 1...N \qquad (12)$$

where A_i is the area of *i*-th ring. Number of molecules cryosorbed on each ring is:

$$n_i\left[T_2\right] = a_i f_i, \quad i = 1...N \qquad (13)$$

4. 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 tritium thermal desorption and re-adsorption can be neglected. Meanwhile some of these molecules will decay emitting a few keV electrons and a few eV beta particle, which will move along the solenoid magnetic field lines causing desorption of argon and tritium. The number of decays during time *Dt* on each ring can be calculated as:

$$\Gamma_i = n_i\left(1-2^{-\frac{\Delta t}{t}}\right) = a_i f_i\left(1-2^{-\frac{\Delta t}{t}}\right), \quad i = 1...N \qquad (14)$$

There is a quite limited number of publication on electron stimulated desorption from cryosorbed gases and mixtures but all at normal incident angle, the most relevant is paper [7] where the desorption yield are reported for argon and argon-hydrogen mixture. Desorption yield of argon depends on coverage and electron energy with highest measured value of ~20 Ar/e^-. Desorption yields of hydrogen depends on percentage on 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 highest measured value of $\sim 10^3$ H_2/e^- (at electron energy of 300 eV). Desorption yields of hydrogen reduces for layers thicker than 100 monolayers of H2. This result is shown on Figure 3 with diamonds. Ion induced desorption at eV energy range is much less than these values (see, for example [8,9]) and can be considered as negligible one comparing to 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 cryo deposit. If energy transferred to adsorbent mainly, with a sublimation energy for solid argon of $E_{subl}(Ar)$=8048 J/mol [10], when up to 60,000 Ar atoms per tritium decay could be evaporated. Meanwhile, a sublimation energy for both hydrogen and tritium sorbed on argon should be much less, ~0.5-2 kJ/mol, and strongly depends on temperature and surface coverage, therefore one may expect that number of desorbed hydrogen and tritium from argon frost should be larger and can be described as:

$$h\left(s_{T_2}\right) = 6\cdot 10^4 \frac{s_{T_2}}{s_{Ar}} \frac{E_{subl}(Ar)}{E_{subl}(T_2)}. \qquad (15)$$

The electron stimulated desorption yields calculated with formula (15) is shown with blue dotted line on Figure 3 for the ratio $E_{subl}(Ar)/E_{subl}(T_2) = 8$, which is sensible approximal value for the temperature about 4.2-4.5K. Having an approximate character of this curve, the match of the date and the curve on Figure 3 is reasonable for the T2 surface coverage, s(T2), less than a monolayer, but not for the higher T2 surface coverage.

Other useful results were obtained during the neutrino mass experiments at Mainz University One week of effective lifetime was reported in [11] for 40-monolayer tritium condensate on an aluminium substrate at 2.8 K. This gives a desorption rate of 750 T2 per decay. In later publication [12] one year of effective lifetime was reported 140-monolayer tritium condensate on a graphite substrate at 1.9 K, which gives the desorption rate of 750 T_2 per decay. There is no data for tritium and hydrogen to expect maximum tritium desorption yield much larger than 10^3 T2 molecules/decay.

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

$$h(s) = h_{\max} \frac{s}{s + s_m} \qquad (16)$$

where s_m is a T_2 surface coverage corresponding to the end of linear growth of tritium desorption yield ($s_m \approx 4{\cdot}10^{14}$ T_2/cm^2). The graph calculated with formula (16) for $h_{max} = 10^3$ T2/decay is show by red solid lane on Figure 3. This formula will was used in the following analysis as the coverage higher than 10^{17} T_2/cm^2 is not expected.

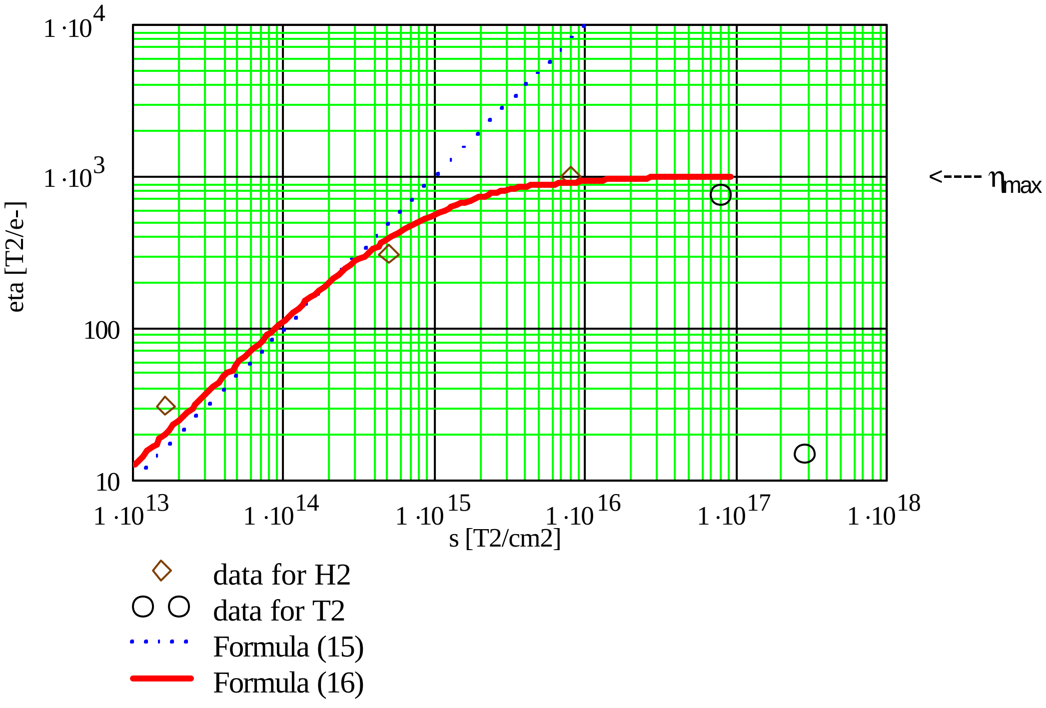

Figure 3. calculation of electron stimulated desorption of tritium.

The number of molecules desorbed during time *Dt* can be calculated for each ring as:

$$d_i = h\left(s_i\right)\Gamma_i = h_{\max}\frac{a_i f_i}{1+\frac{s_m A_i}{a_i f_i}}\left(1-2^{-\frac{\Delta t}{t}}\right),\quad i=1\ldots N \tag{17}$$

For studying of the migration process as a function of time, two simultaneous processes treated in the calculation model as two discrete processes happening in turn with each time interval *Dt*:

- Gas injection, when amount of tritium gas entered into the CPS during time between t_0 and t_0+Dt is distributed following solution for f_i from equation (10), that gives surface coverage distribution s_i and the number of decays during time *Dt* on each ring.
- Tritium decay and electron stimulated desorption. Assuming that all decay happen simultaneously at the end of time period $t= t_0+Dt$ the number of molecules desorbed due to the tritium decays is calculated with formula (17). This gives a new desorption vector **d¢** for equation (10) and a new solution **f¢**.

The final surface coverage distribution s_i¢ at the end of time period $t= t_0+Dt$ is calculated as following:

$$\begin{aligned} n_i' &= n_i - \Gamma_i - d_i' + a_i f_i', \\ s_i' &= \frac{n_i'}{A_i},\quad i=1\ldots N \end{aligned} \tag{18}$$

In the next time periods the itteration cycles are described as:

Given inlet flow Q

$$\mathbf{d}_0 = \left| \begin{array}{l} d_{0,0} = Q\Delta t \\ d_{0,j} = 0 \qquad \text{for j} = 1...N+1 \end{array} \right.$$

Solving $\left(\mathbf{E} - \mathbf{W}\times diag\left(1-\mathbf{a}\right)\right)\times\mathbf{f}_0 = \mathbf{W}\times\mathbf{d}_0$

$$\mathbf{n}_0 = diag\left(\mathbf{a}\right)\times\mathbf{f}_0, \quad \mathbf{s}_0 = \frac{\mathbf{n}_0}{A}$$

for $k = 1...M$

$$\left| \begin{array}{l} \mathbf{G}_k = \mathbf{n}_{k-1}\left(1-2^{-\frac{\Delta t}{t}}\right) = diag\left(\mathbf{a}\right)\times\mathbf{f}_{k-1}\left(1-2^{-\frac{\Delta t}{t}}\right) \\ \mathbf{d}_k = \left| \begin{array}{l} d_{k,j} = 0 \quad \text{for } j = 0 \text{ and } j = N+1 \\ d_{k,j} = h\left(s_{i-1,j}\right)\Gamma_{k,j} = h_{\max}\dfrac{n_{k-1,j}}{1+\dfrac{s_m A}{n_{k-1,j}}}\left(1-2^{-\frac{\Delta t}{t}}\right) \text{for j} = 1...N+1 \end{array} \right. \\ \text{Solving } \left(\mathbf{E} - \mathbf{W}\times diag\left(1-\mathbf{a}\right)\right)\times\mathbf{f}_k = \mathbf{W}\times\mathbf{d}_k \\ \mathbf{n}_k = \mathbf{n}_0 + \mathbf{n}_{k-1} - \mathbf{G}_k - \mathbf{d}_k + \mathbf{a}\times\mathbf{f}_k \\ \mathbf{s}_k = \dfrac{\mathbf{n}_k}{A} \end{array} \right. \tag{19}$$

here index k corresponds to the itteration number

5. Results of calculations.

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

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

The inlet tritium flow was taken as Q=10^{12} T_2/sec. This value was calculated from injected flow at WGTS Q_{inj}=2 mbar·lt/s and a flow rate for DPS1-F and DPS2-F of about $3{\cdot}10^{-8}$. The developing tritium surface coverage along the tube and the tube flow ratio was studies as a function of time up to 10^7 s (115 days reasonable time between warming up the cryostat) and shown on Figure 4 and Figure 5. One can see that that although the surface coating distribution depends on h_{max}, the CPS gas flow ratio almost insensitive to h_{max} in the range 10^3 to 10^5 T_2/e^- for given geometry and injected gas flow Q=10^{12} T_2/sec.

The reasonable question is: what the inlet tritium flow the migration effect is significant at.

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/sec are shown on Figure 6 and Figure 7. In this case the migration effect is negligible for h_{max}=10^3 T_2/e^-, meanwhile it should be considered for h_{max}=10^5 T_2/e^- after about 10 days of continuous injection, the gas flow ratio changes 10 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/sec are shown on Figure 8 and Figure 9. In this case the migration effect should be considered after ~100 days for h_{max}=10^3 T_2/e^- and after ~2 days of continuous injection for and h_{max}=10^5 T_2/e^-, then the gas flow ratio changes 10 times after ~9 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/sec;
- if inlet tritium flow increase due to a turbo-molecular pump failure or other reason up to of Q=10^{14} T_2/sec the tritium migration effect can be neglected for expected h_{max}=10^3 T_2/e^-, or, the worst if η_{max} is underestimated, the tritium migration effect can be neglected for 10 days for h_{max}=10^5 T_2/e^-;
- if inlet tritium flow increase due to a significant failure up to of Q=10^{14} T_2/sec the tritium migration effect can be neglected for expected h_{max}=10^3 T_2/e^- for about

100 day, or, the worst if η_{max} is underestimated, the tritium migration effect can be neglected for 2 days for h_{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 CPS and usual Monte-Carlo routine with available codes can be used for modelling of whole CPS.

6. Conclusion.

A 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 CPS. Because the lack of experimental data the maximum electron stimulated desorption in the model was varied in wide range from the likely expected 10^3 T_2/e^- to maximum possible 10^5 T_2/e^-. The inlet flow was varied from Q=10^{12} T_2/sec at normal operation to Q=10^{16} T_2/sec presenting an accident case.

- The migration effect can be neglected for the calculation of CPS flow ratio for the inlet flow of Q=10^{12} T_2/sec (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/sec (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/sec (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 programmes to model whole CPS.

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

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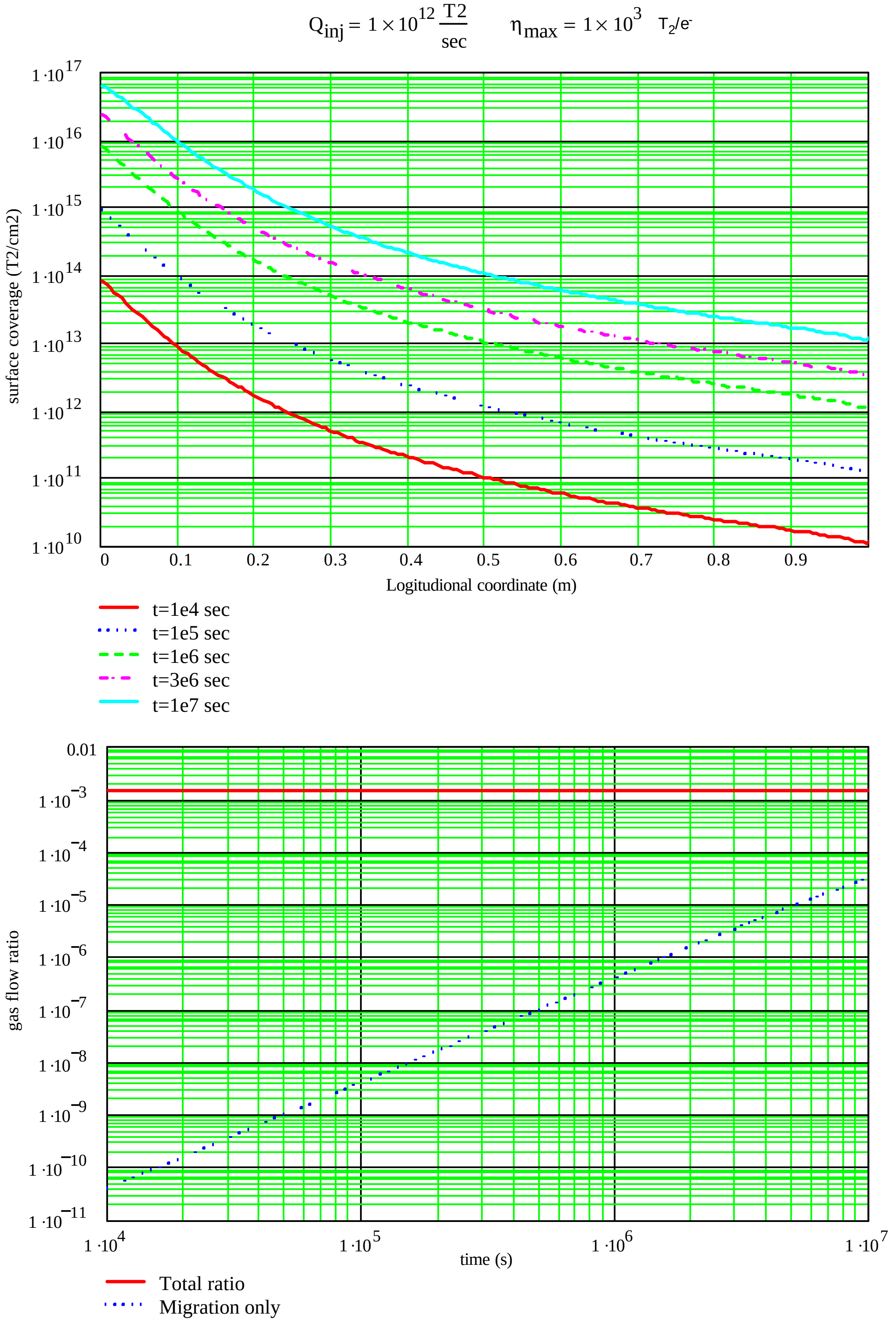

Figure 4. Surface coating as a function of longitudinal coordinate and time (upper graph) and gas flow ratio (bottom graph) for h=10^3 T_2/e^- and inlet flow Q=10^{12} T_2/sec.

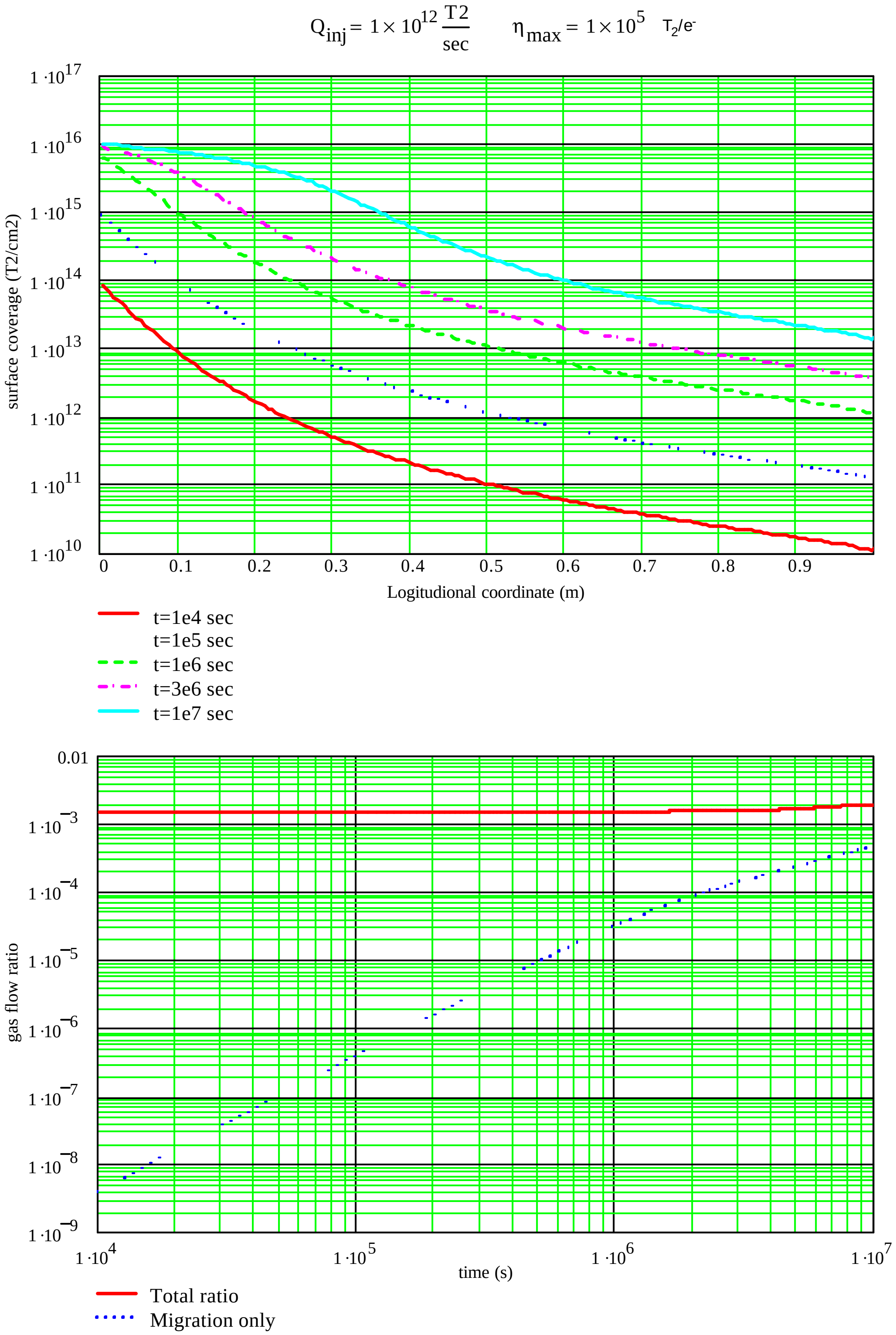

Figure 5. Surface coating as a function of longitudinal coordinate and time (upper graph) and gas flow ratio (bottom graph) for h=10^5 T_2/e^- and inlet flow Q=10^{12} T_2/sec.

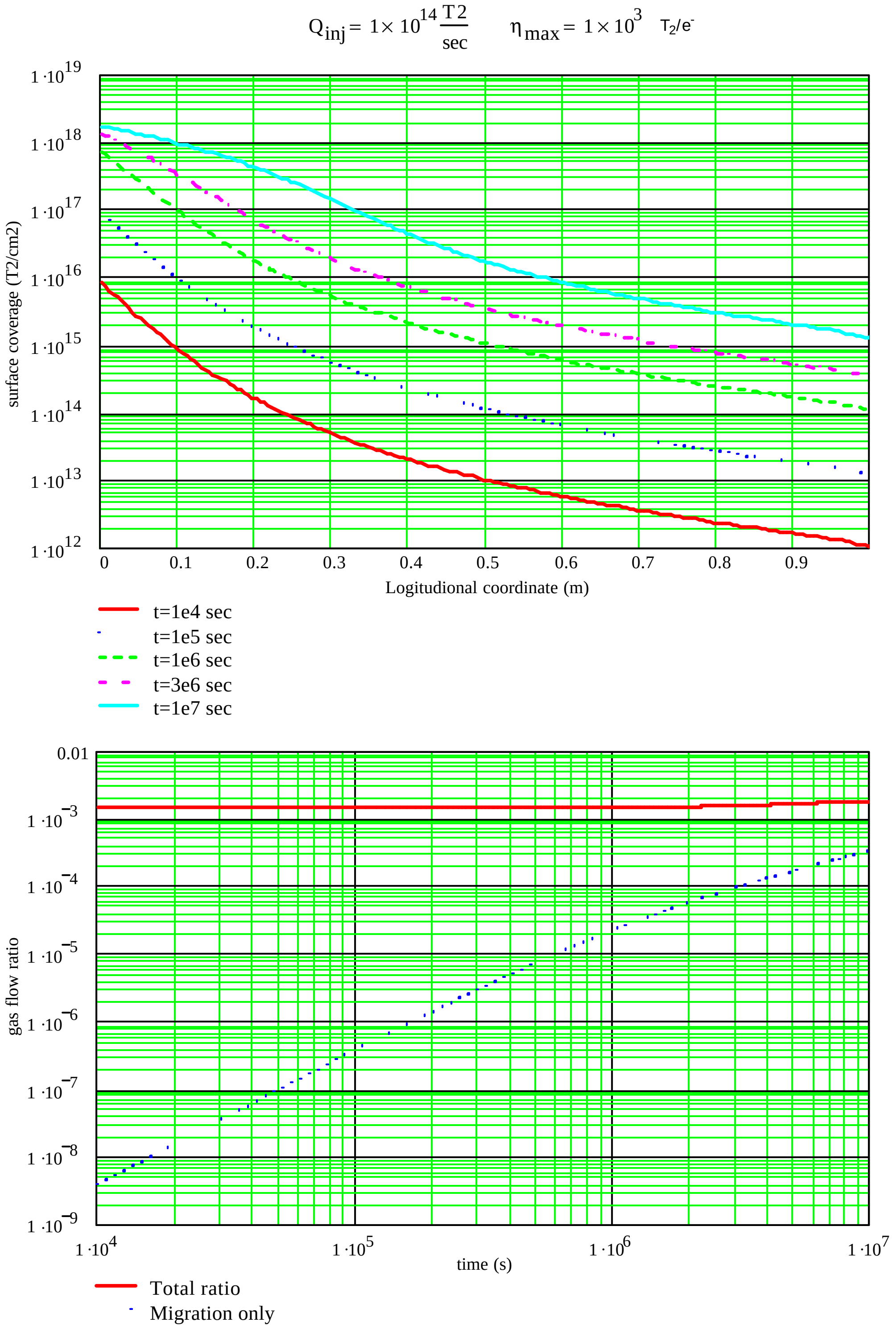

Figure 6. Surface coating as a function of longitudinal coordinate and time (upper graph) and gas flow ratio (bottom graph) for h=10^3 T_2/e^- and inlet flow Q=10^{14} T_2/sec.

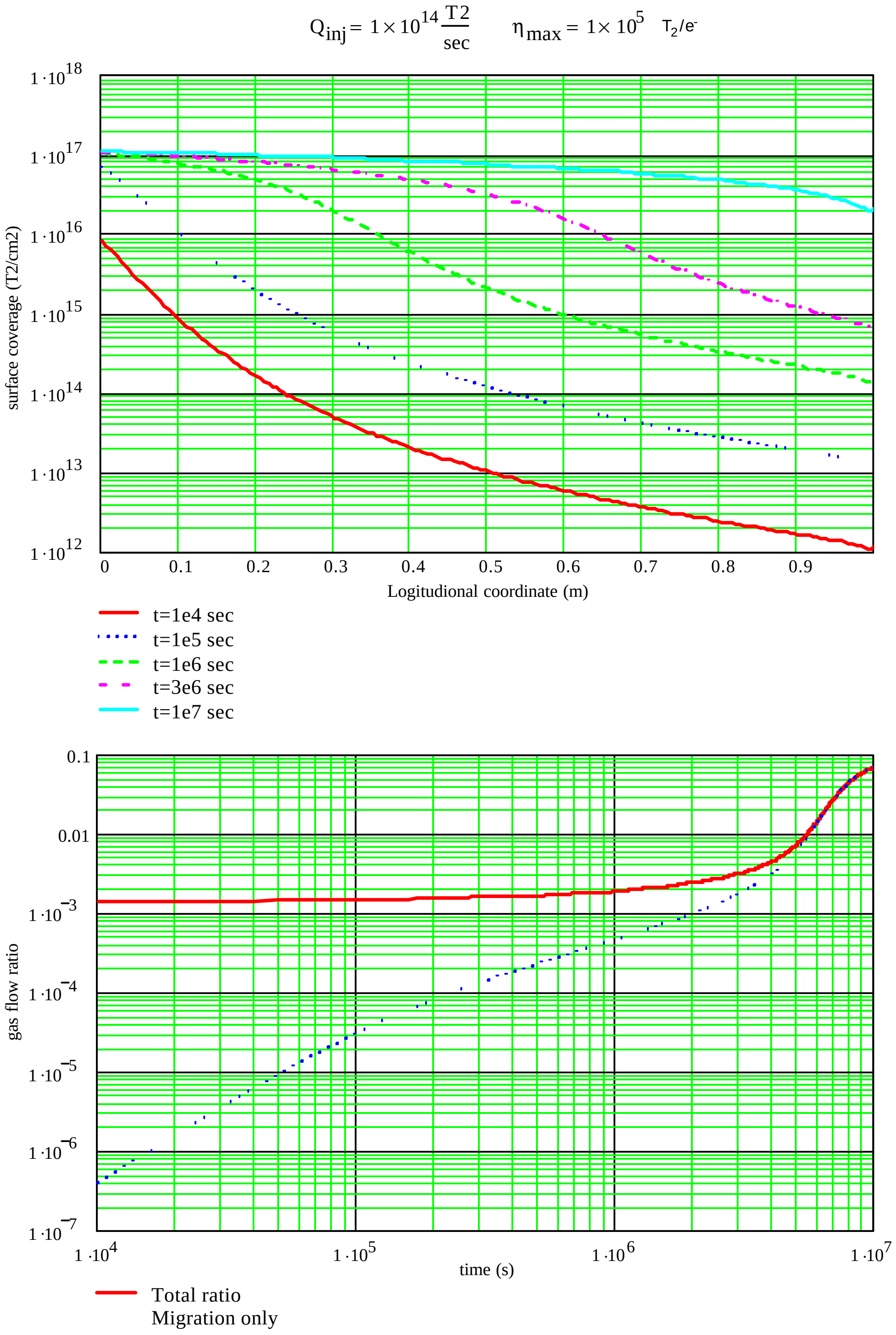

Figure 7. Surface coating as a function of longitudinal coordinate and time (upper graph) and gas flow ratio (bottom graph) for $\eta=10^5$ T_2/e^- and inlet flow $Q=10^{14}$ T_2/sec.

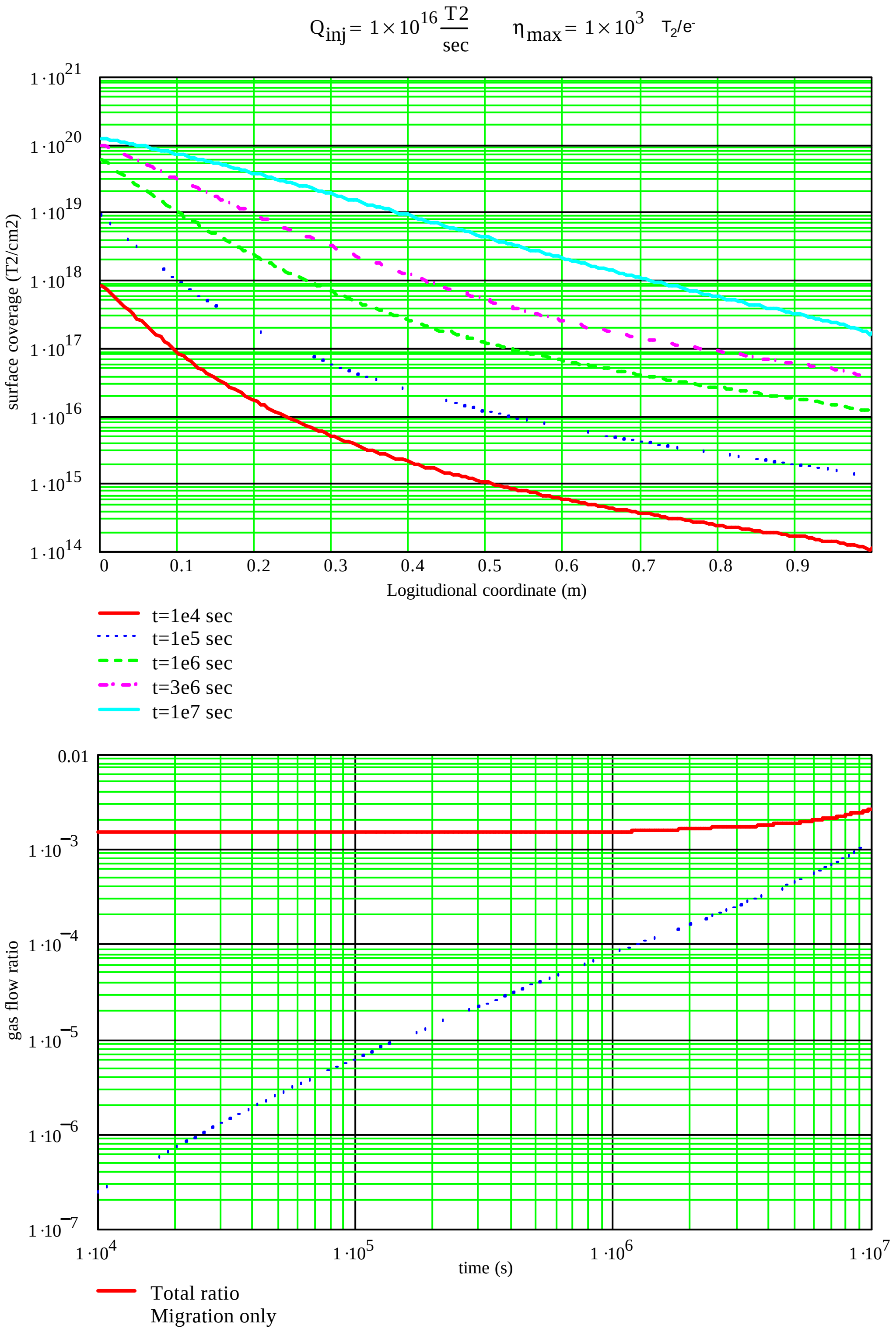

Figure 8. Surface coating as a function of longitudinal coordinate and time (upper graph) and gas flow ratio (bottom graph) for $\eta=10^3$ **T_2/e^- and inlet flow** $Q=10^{16}$ **T_2/sec.**

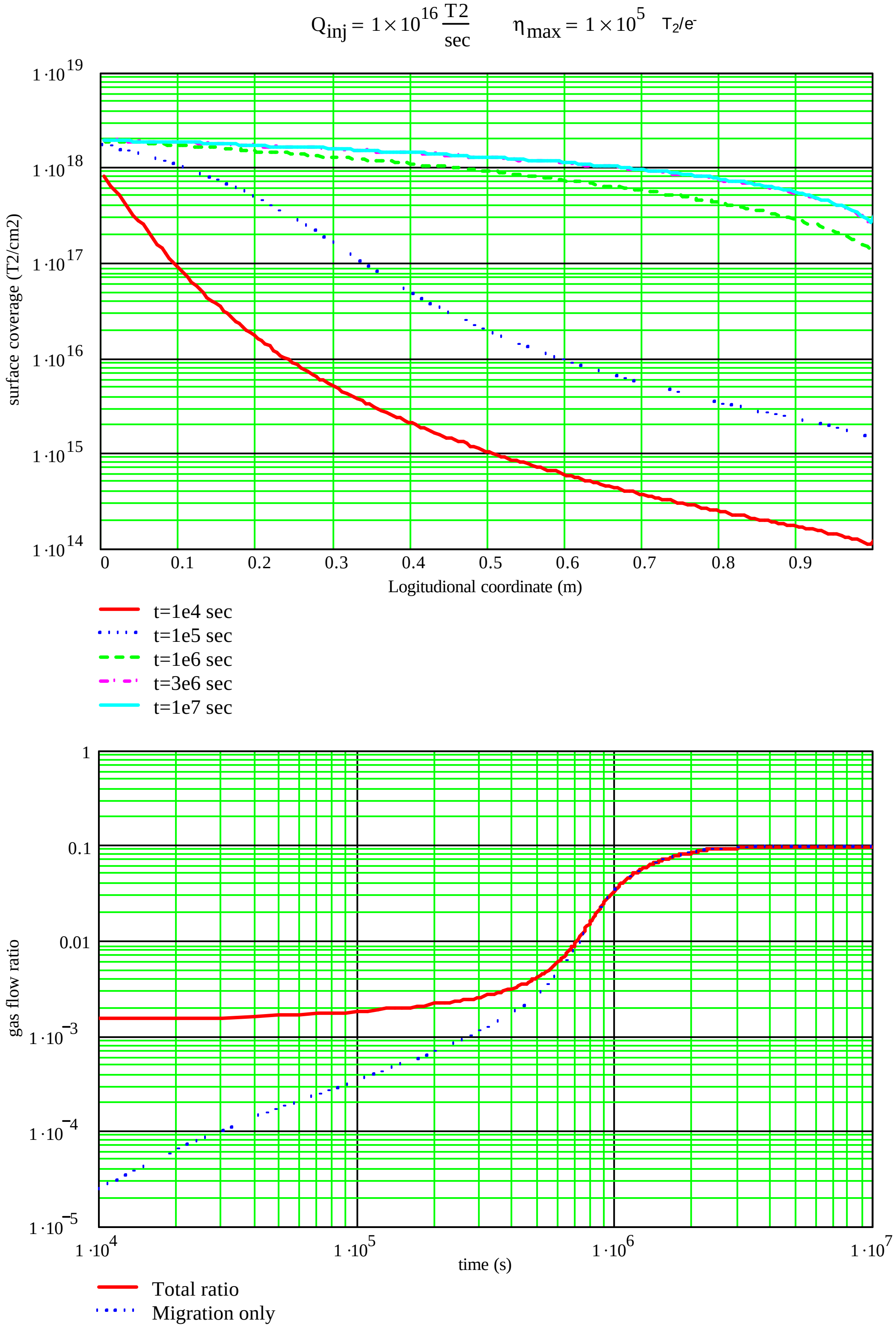

Figure 9. Surface coating as a function of longitudinal coordinate and time (upper graph) and gas flow ratio (bottom graph) for h=10^5 T_2/e^- and inlet flow Q=10^{16} T_2/sec.

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