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Measuring non-condensable gases in steam

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In surgery, medical devices that are used should be sterilized. To obtain surface steam sterilization conditions, not only in the sterilizer chamber itself but also in the loads to be sterilized, the amount of non-condensable gases (NCGs), for instance air, should be very low. Even rather small fractions of NCGs (below 1 %) seriously hamper steam penetration in porous materials or devices with hollow channels (e.g., endoscopes). A recently developed instrument which might detect the presence of residual NCGs in a reliable and reproducible way is the 3M™ Electronic Test System (ETS). In this paper, a physical model is presented that describes the behavior of this instrument. This model has been validated by experiments in which known fractions of NCGs were introduced in a sterilizer chamber in which an ETS was placed. Despite several approximations made in the model, a good agreement is found between the model predictions and the experimental results. The basic principle of the ETS, measuring the heat transfer by condensation on a cooled surface, permits a very sensitive detection of NCGs in harsh environments like water vapor at high temperatures and pressures. Our model may serve to develop adapted and optimized versions of this instrument for use outside the field of sterilization, e.g., in heat exchangers based on steam condensation. © 2013 AIP Publishing LLC

I. INTRODUCTION

Sterilization of medical devices is a crucial part of preventing device-related infections in hospitals. Worldwide, steam sterilization is the most applied sterilization method for surface sterilization of surgical instruments. Surface steam sterilization conditions are derived from sterilization of aqueous liquids, in which the mechanism for the killing of organisms is coagulation. Coagulation requires energy and a wet environment. In surface steam sterilization, steam may provide both the energy and the water. Steam sterilization conditions imply that 100 % water vapor has to be present on the surface at a certain temperature for a certain time. To establish these conditions, the air that is initially present in the sterilizer chamber has to be replaced by saturated steam. This should occur during the first phase of a sterilization process, which is illustrated in Figure 1. During the second phase, the sterilization phase, the established sterilization conditions have to be maintained for a certain time.

If small amounts (about 1 % or more) of non-condensable gases, like air, are present in the sterilization chamber at the start of the sterilization phase, steam sterilization conditions may still be met on the surfaces of the load that are exposed directly to the steam, but the penetration of steam in, for instance, porous materials or narrow channels might be seriously impaired. A possible lack of steam penetration was already recognized half a century ago, and led to the development of the Bowie and Dick (B&D) steam penetration test. This test uses a chemical indicator to judge the penetration of steam through a pack of porous textile towels.

Since the introduction of the B&D test textile loads are less frequently present, whereas loads with surgical instruments have become more common. Furthermore, during the last two decades Minimal Invasive Surgery (MIS) has developed rapidly. Instruments used for MIS often contain hollow channels with diameters of 1 mm or smaller and lengths up to 1 m. The inner surfaces of these channels have to be sterilized as well. Unfortunately, a direct measurement of the steam penetration in such channels is not (yet) possible in practice. Recently, we have performed model calculations and experiments on a test tube mimicking the channels present in medical instruments. This study revealed that a significant fraction of the commonly used sterilization processes may fail to reduce the amount of NCGs to a level which is sufficiently low for sterilization of the inner part of such instruments. No direct relation has been found between the results of a B&D test and the amount of NCGs in the sterilizer chamber.

The concentration of NCGs can be derived indirectly from the steam saturation, which can be calculated from the measured temperature and pressure, but this approach is rather inaccurate for small concentrations of NCGs. On the other hand, small amounts of air can in principle be measured accurately by an Oxygen Analyzer. This instrument, however, has the drawback that it is generally designed to measure a dry gas mixture at a constant (often rather high) temperature. Hence it cannot be used to measure the gas mixture in the sterilizer chamber directly, because this mixture contains a largely varying amount of water vapor and varies in temperature between room temperature and about 410 K. Therefore, samples of the gas mixture should be taken, which next have to be analyzed at a fixed temperature. This hampers a real-time monitoring of the fraction of NCGs. An alternative approach is to measure parameters that are very sensitive to the presence of small amounts of NCGs, such as the speed of condensation and the resulting heat transfer from steam to surface (see for instance Refs. 9 and 10). This latter approach

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is implemented in a recently developed test device, the ETS (3M™, Neuss, Germany), but detailed physical specifications of this instrument are not reported. In this paper, a physical model is presented that describes the behavior of this instrument during a typical steam sterilization process. This model has been validated by experiments in which known fractions of NCGs were introduced in a sterilizer chamber in which an ETS was placed. The paper is organized as follows. In Sec. II, the principle of the ETS will be outlined. The physical modeling of this instrument will be described in Sec. III. The experimental procedure and the results of our measurements will be presented in Sec. IV. In Sec. V, the model predictions will be compared with the data and the model parameters will be evaluated. The paper will be concluded with a discussion in Sec. VI.

II. PRINCIPLE OF THE ETS

The sensing unit of the ETS is schematically depicted in Figure 2. Before the start of a steam penetration test process, the ETS is at room temperature. During the process, a mixture of steam and air will enter the LCP tube of the ETS. The heat capacity of the aluminum (Al) blocks that are attached to the outer side of this tube is so large that they warm up very slowly. Hence the wall of the tube will have a lower temperature than the gas mixture entering the tube. Consequently, the steam will condense on the wall of the LCP tube. The condensate runs off the wall towards the bottom of the tube, where it leaves the ETS. This condensation will establish a continuous flow of steam and air into the LCP tube during the process until the end of the sterilization phase (Figure 1, phases I and II). Air flowing with the steam into the tube will not condense and can only leave via the open end of the tube by diffusion. However, diffusion is a very slow process compared to the flow initiated by the condensing steam on the wall. Therefore, in the tube air will accumulate and the concentration of steam will substantially decrease. This effect will be more pronounced towards the closed end (top) of the tube, where so much air can be present that the steam will not be able to penetrate until this end.

Because the aluminum blocks are well isolated from the outer part of the ETS, the temperature increase of these blocks is directly related to the energy transfer from the gas mixture to the wall of the LCP tube. This energy transfer is dominated by the latent heat that is released during condensation of the steam. The presence of even small amounts of air will significantly reduce the heat transfer. Therefore, it is expected that the temperature increase of an aluminum block closer to the top of the LCP tube (T3, Figure 2) is smaller than that of a lower block (T5). This effect will be larger when more air is present in the sterilizer chamber. If the ETS is used for standard steam sterilization processes, only the temperatures T3 and T5 are read out, but in other applications it may be useful to use different or a larger number of block temperatures. We will return to this point in the discussion.

All temperature sensors are Pt100s with a resolution of 0.01°C and an accuracy of 0.2°C. The resolution and accuracy of the pressure measurements are 0.1 and <0.3 kPa, respectively. After a test, the data measured by the ETS is downloaded in a computer for further processing and the ETS is placed outside the sterilizer chamber to cool down to room temperature before it is used in a subsequent test.
Therefore the pressure is related to the partial densities $\rho_v$ and $\rho_g$ used in our computations. The inner LCP tube is open at one end and the temperature $T$ of the tube is assumed to be thermally shunted in the axial direction by the original Al blocks, which increases the effective axial heat conductivity of the LCP tube in that region by a factor of 10. All dimensions are taken from an ETS identical to the one used in our experiments.

### III. PHYSICAL MODEL

Figure 3 schematically illustrates the model of the ETS used in our computations. The inner LCP tube is open at one end ($z = 0$) and closed at the other end ($z = L_{tube}$). The radius of the tube is $r$. Only the open end of the LCP tube is in contact with the sterilizer chamber. The process in the sterilizer chamber is assumed to be ideal: instantaneous mixing of the water vapor and air occurs, resulting in a homogeneous gas distribution throughout the sterilizer chamber.

We will now successively consider the flow of the gas mixture in the tube, the condensation process and the temperature variation of the aluminum blocks. Since the flow of the gas mixture is modeled in a way largely similar to that presented in Ref. 7, some details will be omitted here.

At the conditions that occur during a steam sterilization process the gas (air) and water vapor act as ideal gases.\(^{11,12}\) Therefore the pressure is related to the partial densities $\rho_v$ and $\rho_g$ and the temperature $T$ by

$$ p = p_v + p_g = \rho_v R_v T + \rho_g R_g T, \quad (1) $$

where $R_i$ is the specific gas constant of component $i$ and the subscripts $v$ and $g$ represent water vapor and gas, respectively. The total density $\rho$ equals $\rho_v + \rho_g$.

Since the Reynolds number of the flow in the tube ($Re = 2\rho V a/\eta$, with $\rho$ the density (kg m\(^{-3}\)), $V$ the velocity (m s\(^{-1}\)), $a$ the radius of the tube and $\eta$ the viscosity (Pa s)) is smaller than 100 and the relative pressure difference over the length of the tube ($\Delta p_{ref}$) is much smaller than unity, the flow inside the tube is locally described by the Poiseuille equation:\(^{13}\)

$$ \pi = -\frac{r^2}{8\eta} \frac{\partial p}{\partial z}, \quad (2) $$

with $\pi$ the velocity (m/s) averaged over the cross-section of the tube and $p$ the pressure (Pa). The conservation of mass in the tube is given by\(^{13}\)

$$ \frac{\partial \rho_i}{\partial t} + \frac{\partial (\rho_i \bar{u})}{\partial z} = 0. \quad (3) $$

Using Fick’s law,\(^{13}\) the equation for the conservation of mass of component $i$ can be written as

$$ \frac{\partial \rho_i}{\partial t} + \frac{\partial (\rho_i \bar{u})}{\partial z} = \frac{\partial}{\partial z} \left( \rho_i D* \frac{\partial \rho_i}{\partial z} \right). \quad (4) $$

$D*$ is a modified diffusion coefficient, which appears because of the radial dependence of the velocity (Taylor dispersion). For the range of parameter values appropriate to the present model it can be approximated by\(^{14,15}\)

$$ D* = D + \frac{a^2 \overline{u}^2}{48 D}. \quad (5) $$

The diffusion coefficient $D$ depends on the temperature and pressure\(^{16}\)

$$ D(p, T) = D_{ref} \left( \frac{T}{T_{ref}} \right)^{\frac{3}{2}} \frac{p_{ref}}{p}, \quad (6) $$

where $D_{ref} = 2.4 \times 10^{-5}$ m\(^2\)/s denotes the reference value of $D$ at $T_{ref} = 313$ K and $p_{ref} = 100$ kPa.\(^{11}\)

The chamber of the sterilizer used in our experiments has a volume of 0.340 m\(^3\), whereas the inner channel of the ETS has a volume of about 3 m\(^3\). The disturbance of the gas mixture at the open end ($z = 0$) of LCP tube of the ETS is therefore neglected and the boundary conditions at $z \rightarrow 0$ are

$$ p(0, t) = p(t), \quad \rho_i(0, t) = \rho_i(t), \quad (7) $$

where $p(t)$ and $\rho_i(t)$ refer to the quantities in the sterilizer chamber, and $p(z, t)$ and $\rho_i(z, t)$ to those within the tube.

Measurements with a thermocouple inside the ETS showed that the gas temperature during the sterilization phase (Figure 1) decreases from 407 K at $z = 0$ to about 380 K at $z = L_{tube}$. During the evacuation of the sterilizer chamber during phase I, the gas temperature decreases to about 323 K. To keep the model manageable, however, we have described the flow of the gas mixture within the tube by an isothermal process, at a temperature corresponding to that at the open end of the tube during the sterilization phase. This approximation was also motivated by preliminary calculations, based on the experimentally observed temperature increase of the Al blocks resulting from condensation of water vapor inside the LCP tube of the ETS. These calculations revealed that the latent heat of the condensing water vapor dominates all other terms in the energy balance of the gas-liquid system within this tube. Of course, one might argue that since the relative pressure difference over the length of the tube is very small, a temperature decrease towards the end of the tube would imply an increase of the density of vapor and air. As such this is correct, but this increase (of about 10%) will hardly affect the condensation rate, which depends primarily on the wall.

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**FIG. 3.** Schematic representation of the model used in the computations. The inner channel of the ETS is modeled as a cylindrical tube with a constant radius $\overline{r}$ and length $L_{tube}$. The 12 aluminum (Al) blocks are approximated by a second tube with length $L_{block}$ and an effective heat capacity $C_{block}$, in which the distance between the original Al blocks is taken into account. The heat conductivities of this tube in the axial and radial direction are taken zero and infinitely large, respectively. The heat conductivities of the LCP tube in the radial and axial direction are used as model parameters. The part of the LCP tube in contact with the outer tube is assumed to be thermally shunted in the axial direction by the original Al blocks, which increases the effective axial heat conductivity of the LCP tube in that region by a factor of 10. All dimensions are taken from an ETS identical to the one used in our experiments.
temperature of the tube, the partial vapor pressure and the air fraction.

Next, we consider the condensation process of the water vapor. We will model this process as film condensation and use standard Nusselt boundary layer theory. This theory predicts a film of condensate running down along the inner wall of the tube with a thickness $\delta(x)$ given by

$$\delta(x) = \left[\frac{4k(T_{\text{sat}} - T_w)\mu x}{\rho_f(\rho_f - \rho_v)gh_{fg}}\right]^{1/4}. \quad (8)$$

In this equation, $k$ is the thermal conductivity of liquid water (0.68 Wm$^{-1}$K$^{-1}$ in the temperature range of interest), $T_{\text{sat}}$ the saturation temperature of the vapor, $T_w$ the wall temperature, $\rho_f$ and $\rho_v$ the density of the fluid and vapor, respectively, $\mu$ the dynamic viscosity of liquid water (0.28 $\times$ 10$^{-3}$ Pa s), and $g$ the gravity constant (9.8 m s$^{-2}$). The parameter $h_{fg}$ is the latent heat of water (2260 kJ kg$^{-1}$). The distance from the top of the film is denoted by $x = L_{\text{tube}} - z$. The density of water is taken as 940 kg m$^{-3}$, where the variation of about 3% in the temperature range of interest is neglected. The vapor density $\rho_v$ is less than 1% of $\rho_f$, so it is neglected in the calculation of $\delta$.

The local heat flux $q(x)$ to the wall of the tube is given by

$$q(x) = \frac{k(T_{\text{sat}} - T_w)}{\delta} \left[\frac{k^2 \rho_f(\rho_f - \rho_v)gh_{fg}}{4\mu}\right]^{1/4} \times (T_{\text{sat}} - T_w)^{3/4} x^{-1/4}. \quad (9)$$

Inserting the numerical parameter values and the radius of the LCP tube of the ETS (3 mm) in this equation, we obtain the following expression for the local heat transfer per meter through the wall of the tube:

$$q_m(z) = P_m(T_{\text{sat}} - T_w)^{3/4}(z - L_{\text{tube}})^{-1/4}, \quad (10)$$

where for an ideal film $P_m = 163$ W m$^{-3/4}$ K$^{-3/4}$. Note that both $T_{\text{sat}}$ and $T_w$ depend on $z$.

The presence of NCGs has been shown to reduce the heat transfer by condensation dramatically. Basically this is caused by the development of a layer with a large fraction of NCGs near the surface of the condensate, through which vapor molecules have to diffuse before they can condense. As an example, we have plotted the relevant data calculated by Minkowycz and Sparrow$^9$ in Figure 4. The other cited studies yielded results which generally agree with these data within about 20%. For computational purposes, we have approximated the data by an empirical curve. We chose an expression of the form

$$q/q_{Nu} = \exp(-C_f(z/\delta)^{1/3}), \quad (11)$$

with $f_{air} = \rho_{air}/(\rho_{air} + \rho_{vapor})$ and $C_f = 5$, since this simple expression nicely describes the very steep behavior for small air fractions. Various alternatives were found to be satisfactory as well, although computationally more expensive. The value of $q_m$ obtained by combining Eqs. (10) and (11) is divided by the latent heat of condensation of water vapor and added as a sink term for $\rho_v$ in Eq. (4).

Finally, we consider the temperature of the tubes depicted in Figure 3. The time constant for heat transfer through the wall of the LCP tube to heat up its outer surface or the Al blocks can be estimated from

$$T_w = w^2 c_p \rho_w / \lambda, \quad (12)$$

where $w$ [m] is the wall thickness, $c_p$ [J K$^{-1}$ kg$^{-1}$] the heat capacity of the wall material, $\rho_w$ [kg m$^{-3}$] the density, and $\lambda$ [W m$^{-1}$ K$^{-1}$] the heat conductivity of this material. Using typical values for the heat conductivity and heat capacity, the time constant for heat transfer through the wall appears to be in the order of 2 s. Since the relevant times in a sterilization process are two orders of magnitude larger, we simplified the problem by assuming a lumped circuit, where the heat capacity is concentrated on the outer side of the LCP tube. The temperature $T_f$ of this outer side can be described by

$$C_t \frac{\partial T_r}{\partial t} = k_{LCP,x} A_{LCP,x} \frac{\partial^2 T_r}{\partial x^2} + q_m. \quad (13)$$

The term $k_{LCP,x}$ denotes the heat conductivity of the LCP tube in the axial direction and $A_{LCP,x}$ is its cross sectional area. $C_t$ represents the heat capacity per meter of the system (inner tube only for $z < (L_{\text{tube}} - L_{\text{block}})$ or both tubes for larger values of $z$), whereas the term $q_m$ is the heat of condensation discussed above. The heat capacity of Al, which varies about 7% in the temperature region of interest, was fitted by a second order polynomial.

The local value of the temperature $T_{\text{sat}}$ was calculated from the partial vapor pressure $p_v = p \times (\rho_v R_v)/(\rho_f R_f + \rho_v R_v)$ using the conversion equations given in Ref. 12. The temperature of the inner wall $T_w$ was eliminated from the calculations by combining Eq. (10) with the equation describing the heat transfer through the wall:

$$q_m = h_{LCP,r}(T_w - T_r), \quad (14)$$

where $h_{LCP,r}$ represents the heat transfer coefficient per meter in the radial direction.
Summarizing, the set of Eqs. (1)–(6) together with the boundary conditions (Eq. (7)) and the sink term for $\rho_v$ describes the flow of the vapor-air mixture in the ETS. The condensation is modeled by Eqs. (10), (11), and (14). The temperature variation of the LCP tube and the Al blocks is modeled by Eq. (13), where the initial temperature is estimated from the experimental data. The resulting set of 3 second order PDEs is solved numerically using the standard pdepe routine of Matlab®. This routine solves initial-boundary value problems for parabolic-elliptic PDEs in one dimension. The numerical solution was obtained at 150 equally spaced grid points along the LCP tube and for every 0.5 s up to 650 s.

IV. EXPERIMENTAL

For the experiments, a Lautenschläger type 3119/4STE test sterilizer (Cologne, Germany) with a volume of 340 L is used. Details of this sterilizer and its steam supply have been described by Benoit et al. Figure 5 shows a typical result for a process during which hardly any Non-Condensible Gases (NCGs) were present in the sterilization chamber. The data presented in this figure indicate that the temperature of the aluminum blocks in the ETS gradually rises from about 25 °C at the start of the final inlet of steam ($t \approx 210$ s) to about 80 °C at the end of the sterilization phase ($t \approx 530$ s). The temperature increase of T3 (near the closed end of the LCP tube) is slightly smaller than that of T5 (more distant from the closed end).

The same process was repeated in the other experiments, but now a known volume of air was injected in the sterilizer chamber just before the final inlet of steam. This volume is calibrated as cm$^3$ at room temperature (295 K). For each air volume, the experiment was repeated three times. From the reproducibility of these experiments, the error is estimated to be about 0.01 °C s$^{-1}$. The observed values of $\frac{dT}{dt}$ for T3 and T5 are plotted in Figure 6 versus the injected volume of air. Inspection of this figure shows that when no air is injected, the temperature of T3 and T5 increases at about the same rate at the start of the sterilization phase. When air is injected, the temperature increase of T3 drops significantly below that of T5, corroborating the entrapment of air near the closed end of the LCP tube of the ETS.

We like to note that that the highest amount of injected air (about 900 cm$^3$) corresponds to a concentration of only about 0.25 % in the sterilizer chamber. Inspection of the data presented in Figure 6 shows that in principle air fractions down

![Figure 5](image1.png)

**FIG. 5.** Example of data measured by an ETS during the experiments. In the upper plot, the solid curve denotes the pressure and the dotted curve denotes the temperature, both measured in the sterilizer chamber. The dashed curve denotes the so-called theoretical temperature, the temperature calculated from the pressure.$^{11}$ The start and end of the sterilization phase are denoted by vertical dotted lines. The lower plot shows the measured temperature of T3 and T5 (see Figure 2), denoted by the dotted and dashed curve, respectively. From these temperatures, an inverse time constant $\tau$ equal to $(dT/dt)/(T_{chamber} - T_i)$. The behavior of these $\tau$-values is denoted by the dashed-dotted and solid curve for T3 and T5, respectively.

![Figure 6](image2.png)

**FIG. 6.** Temperature change per unit of time $(dT/dt)$ at the positions of T3 and T5 of the ETS (see Figure 2) as a function of the volume of air injected just before the final inlet of steam. The derivatives $dT/dt$ are calculated at the start of the sterilization phase. The open squares denote the experimental data, the dashed and solid curves reflect the corresponding model predictions assuming ideal and non-ideal film condensation, respectively.
to about 0.05 \%, present during a sterilization process, can be clearly detected by an ETS.

V. RESULTS

Because the model outlined in Sec. III describes a situation where uninterrupted condensation occurs at the inner wall of the LCP tube, it is used to calculate the behavior of the ETS during the period between the final steam inlet and the end of the sterilization period. During phase I of the process, the sterilizer chamber is successively evacuated and pressurized with steam. We assume that the injected steam contains no residual air and that no condensation occurs in the sterilizer chamber. In that case the air fraction \( f_a \) just before the final inlet of steam is given by \( f_a = (p_a/p_s)(p_v/p_s)^2 \) if no additional air is injected (the pressures \( p_a \), \( p_s \), and \( p_v \) are defined in Figure 1). The total air density, together with the value of \( p_s \) and the temperature \( T_w \) at this moment, specify the initial situation.

The data presented in Figure 5 show that without injection of air the aluminum blocks in the ETS are heated at a rate of on average 0.16 °C s\(^{-1}\) between the start of the final steam inlet and the end of the sterilization phase. From the volume of the 12 Al blocks (4.5 cm\(^3\) per block) a heat capacity of about 130 J K\(^{-1}\) is found, and hence the heat supplied by the condensation of vapor is roughly 21 W over a length \( L_{\text{block}} = 0.093 \) m. If we integrate Eq. (10) over the tube, we obtain a value \( T_{\text{sat}} - T_w \approx 0.6 \) °C and a heat transfer coefficient of the LCP tube \( h_{\text{LCP}, z} \approx 2 \) W K\(^{-1}\)m\(^{-1}\). This would imply that the difference between the wall and gas temperature is very small compared to the temperature drop across the wall of the LCP tube. In that case, the heat transfer to the Al blocks would almost fully be determined by the heat conduction of the inner tube and changes in the condensation process resulting from NCGs would be largely suppressed. The results for this situation are represented by the dashed curves in Figure 6. Inspection of this figure shows that the qualitative characteristics of the experiments are described correctly, i.e., the decrease of \( dT/dt \) at increasing air fractions, which is much more pronounced for T3 than T5. It is obvious, however, that the experimentally observed decrease of \( dT/dt \) at small values of \( V_{\text{air}} \) is much larger than predicted by the model.

The LCP tube of the ETS can fairly accurately be described as a cylindrical channel with a diameter of 6 mm, centered in a square rod with transverse dimensions 8.2 \( \times \) 8.2 mm\(^2\). If we take the heat conductivity of a typical polymer, which is in the order of 0.5 W m\(^{-1}\)K\(^{-1}\), we obtain an estimate \( h_{\text{LCP}} \approx 9 \) W K\(^{-1}\)m\(^{-1}\), which is more than a factor of 4 higher than the value inferred above. Since the heat conductivity of LCP may be even higher, the description of the condensation process by ideal film condensation at the entire inner surface of the LCP tube is probably too much simplified. Actually, inspection of the inner wall of the tube revealed 12 equidistant milled ridges in the axial direction with a height of about 0.3 mm, which may give rise to preferential condensation on only a part of the surface. For this reason, \( h_{\text{LCP}} \) was chosen such that the best agreement with the experimental data was obtained, whereas \( P_m \) was adjusted such that for \( V_{\text{air}} = 0 \) the value of \( dT/dt \) was equal to that observed in our experiments. It appeared that the value for \( h_{\text{LCP}} \) is not very critical; the above estimate of 9 W K\(^{-1}\)m\(^{-1}\) resulting in a value \( P_m = 7.2 \) W m\(^{-3/4}\)K\(^{-3/4}\) was satisfactory. This value of \( P_m \) suggests that the actual condensation process is about a factor of 20 less effective than ideal film condensation.

The heat conductivity \( k_{\text{LCP}, z} \) of the LCP tube in the axial direction, defined in Eq. (13), can be used to optimize the difference in the behavior of T3 and T5. For larger values of \( k_{\text{LCP}, z} \), the behavior of T3 and T5 becomes more similar, for smaller values the difference is largest. The best agreement with the experimental data was obtained for \( k_{\text{LCP}, z} \approx 1.5 \) W m\(^{-1}\)K\(^{-1}\). This is somewhat larger than expected for a typical polymer, although not unrealistic for LCP. Apart from this, the heat conductivity in the axial direction may be enhanced by various contributions that are not included in the model. In this respect, we like to note that the heat transfer via the air between adjacent Al blocks in the ETS (Figure 2) already adds 50 \% to the value calculated for the tube alone. The resulting behavior of \( dT/dt \) for T3 and T5 is represented by the solid curves in Figure 6. Inspection of this figure shows that our model describes the data for both temperature sensors simultaneously within experimental inaccuracy, except for the data for T3 at high values of \( V_{\text{air}} \). We like to note, however, that by small readjustments of the two independent fitting parameters the deviations in this region can be reduced, at the cost of introducing a small error elsewhere.

VI. DISCUSSION

We have demonstrated experimentally that an instrument which has been proposed to assess the steam penetration during a steam sterilization process is very sensitive to even small concentrations of NCGs. Using a rather simple and straightforward physical model, the response of this instrument to additional volumes of air injected in the sterilizer chamber at the start of the final steam inlet can be described very well.

As already discussed above, we have included the effect of the presence of NCGs on the condensation heat transfer by fitting an empirical curve to a set of numerical results reported in the literature (see Figure 4). Inspection of this figure shows that the reduction of the heat transfer depends on both \( T_{\text{sat}} \) and \( T_w \), the reduction being larger as \( T_{\text{sat}} \) or \( T_w \) decreases. In the temperature region of interest, these variations appear to have only a minor effect on the results of our model, in the sense that they can be largely compensated by small adjustments of the model parameters \( h_{\text{LCP}} \) and \( k_{\text{LCP}} \).

One of the simplifications of our model is the implicit assumption of a more or less stationary condensate film on the inner wall of the LCP tube in the ETS. This implies that condensation will occur as soon as \( T_{\text{sat}} > T_{\text{wall}} \), which condition is already satisfied at a pressure of about 10 kPa during the final steam inlet, corresponding to about 2 s after the start. At that moment, however, no sufficient vapor has already entered the tube to create a condensate film as described by Eq. (8). Using the parameter values given in Secs. II–V and assuming that all vapor flowing into the tube will condense, the time to create such a film can be estimated to range between 15 and 40 s. Since it is not known how the initial film formation in an LCP tube with its particular geometry occurs, we included
this effect in first order by assuming that the parameter $P_m$ in Eq. (10) grows linearly with time until the film has formed.

To illustrate the effect of this refinement, we will now consider not only the situation at the start of the sterilization phase, but use our model to calculate the behavior within a larger time interval, covering the region of Figure 5 from the start of the final steam inlet to the end of the sterilization phase. The variation of the temperature of T3 and T5 can be calculated straightforwardly. For the calculation of $\tau_i^{-1} = (dT_i/dt)/(T_{chamber} - T_i)$ for each block i, we took the chamber temperature derived from the partial vapor pressure (Eq. (1)) using the conversion equations given in Ref. 12. This temperature is also referred to as theoretical temperature,\(^{11}\) which is given by the dashed curve in the upper part of Figure 5. These calculations revealed that the behavior of $\tau_i^{-1}$ during the sterilization phase can be predicted reasonably well by the original model, but the predicted behavior just after the start of the final steam inlet is too high by a factor of two.

If we include a film formation time of 20 s, we obtain the temperature variation of T3 and T5 and the behavior of $\tau_i^{-1}$ plotted in Figure 7. These results rather nicely agree with the data presented in Figure 5. We like to note that the experimental data also reveal a time delay of about 20–30 s between the start of the final inlet of steam and the inverse relaxation times $\tau_i^{-1}$ for each block i as denoted by the vertical dotted lines. The temperature variations of T3 and T5 are denoted by the dashed and dotted curves, respectively. The behavior of $\tau_i^{-1}$ is denoted by the solid curve for T3 and T5, respectively. The time $t^*$ denotes the time with respect to the start of the sterilization phase.

The high sensitivity of an ETS to the presence of NCGs in the sterilizer chamber results from the accumulation of air at the end of the LCP tube. This is illustrated very nicely by the calculated results plotted in Figure 8. This figure also reveals that the accumulation of air continues during the sterilization phase. In our model, it is assumed that the fraction of air in the sterilizer chamber does not change from the start of the final steam inlet to the end sterilization phase, since the volume of the sterilizer chamber is about five orders of magnitude larger than that of the inner channel of the LCP tube.

In principle this is correct, but the amount of air injected in the sterilizer chamber is limited and the ETS extracts much more air from the chamber than estimated from the volume of the LCP tube alone. From the data presented in the figure and the radius of the LCP tube it can be inferred that for an injected air volume of 200 cm$^3$ (at 100 kPa) roughly 1.4 cm$^3$ has been accumulated in the tube at the end of the sterilization phase, which will only have a minor effect on the model. From the figure it can also be deduced that changes of the positions of T3 and T5 have a large effect on the measured temperatures. In this respect one should note that our model assumes that the heat transfer by condensation is measured locally, whereas in reality it is averaged over the thickness of an individual Al block (7.1 mm). Possible errors resulting from this approximation are most likely compensated by the choice of the fitting parameters.

Both our experiments and model calculations reveal that the ETS is very sensitive to residual NCGs that are present in the sterilizer chamber before the final inlet of steam. If the values of $\tau_i^{-1}$ of T3 and T5 are significantly lower than their values in the absence of NCGs and $\tau_i^{-1}$ is substantially lower than $\tau_5^{-1}$, the steam penetration in long hollow channels may be impaired significantly, as has been shown previously.\(^{7}\) Our model reveals that the basic principle of the ETS, measuring the effect of entrapped air on the heat transfer by condensation on a cooled surface, permits a very sensitive detection of NCGs in harsh environments like water vapor at high temperatures and pressures.

FIG. 7. Calculated behavior of the temperature variation of T3 and T5 and the inverse relaxation times $\tau_i^{-1} = (dT_i/dt)/(T_{chamber} - T_i)$ for each block i for the situation corresponding to the experimental data presented in Figure 5. The start and end of the sterilization phase are denoted by vertical dotted lines. The temperature variations of T3 and T5 are denoted by the dotted and dashed curves, respectively. The behavior of $\tau_i^{-1}$ is denoted by the dashed-dotted and solid curve for T3 and T5, respectively. The time $t^*$ denotes the time with respect to the start of the sterilization phase.

FIG. 8. Partial air pressures normalized to $p_{ref} = 100$ kPa as a function of position in the inner channel of the LCP tube of the ETS. The pressures at the start of the sterilization phase are given by the solid curves, which are labeled according to the volume of additional air $V_{air}$ injected in the sterilizer chamber. The dashed curve denotes the partial air pressure for $V_{air} = 200$ cm$^3$ at the end of the sterilization phase. The gray areas represent the position of the Al blocks T3 and T5. Note that only the upper part of the inner channel is depicted.
Using this model, adapted versions of this instrument might be designed for applications outside the field of steam sterilization, such as heat exchangers based on steam condensation, where also the efficiency is impaired by the presence of non-condensable gases. Depending on the desired sensitivity for NCGs, one might choose to monitor the temperature of an Al block close to the end of the LCP tube (high sensitivity), close to the open end (lower sensitivity), or a series of blocks (wide sensitivity range). This type of applications may require continuous monitoring of the amount of NCGs, which prohibits cooling down the ETS between successive measurements or tests. In such cases, the ETS itself can be located outside the actual device and the open end of the (vertically oriented) LCP tube of the instrument can be connected to, for instance, a steam supply line. By implementing known heat leaks between the Al blocks and the environment (e.g., room temperature), measurements can be performed in more or less stationary situations, in which the transport of NCGs by diffusion towards the open end of the LCP tube equals the amount of NCGs entering the tube along with the steam. In such a configuration, the “amplifying” action of the ETS will be preserved and an adapted version of the model presented in this paper might be used as a design tool.