CREATING A DIFFUSION CHAMBER WITH A TIME-PERIODIC TEMPERATURE CONDITION AT THE WALL AND SUPERSATURATION UNIFORM OVER THE WHOLE VOLUME

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A simple heat-transfer arrangement enabling the thermal conditions at the walls of a diffusion chamber to be made periodic in time is described and examined analytically. Extensive practical uses for the heat-transfer scheme in question are foreseen.

The idea of creating supersaturation inside a chamber with wet walls by subjecting the latter to a periodic temperature variation was proposed in [1]. An attractive feature of this technique lay in the possibility of achieving the supersaturated state of a vapor–gas mixture, combining constancy in time with uniformity in space. Until recently, however, no success had been achieved in making any apparatus based on this method. The main reason for failure lay in the complexity of the practical problems involved as well as in a lack of knowledge regarding the thermophysical processes taking place at the chamber walls.

Let us first of all consider the heat transfer between an infinite plane wall H thick and the surrounding medium. Let us assume that the heat transfer from the left-hand side h_1 is more efficient that from the right h_2. This corresponds in particular to the case in which one side of the wall is in contact with liquid and the other with air. Under ordinary conditions the heat-transfer coefficient for water is much greater than for air ($h_1 \gg h_2$). Let us assume that heat sources are uniformly distributed over the whole volume of the wall, the intensity of these varying with time in accordance with an arbitrary law q(t). Then the equation describing the change in wall temperature with time takes the form

$$\frac{du}{dt}+\frac{h}{HC\rho}u=\frac{q(t)}{C\rho}+\frac{h}{HC\rho}\theta, \tag{1}$$

where θ is the temperature of the cooling medium; q(t) is a function describing the intensity of the volume heat sources as a function of time.

Let us consider the solution of Eq. (1) for the case in which heat sources act periodically over the whole volume of the wall in the form of rectangular pulses with a period T and an occupation factor of γ. Physically this corresponds to the case in which an electric current is passed through the wall in pulses and Joule heat is released periodically in the volume of the wall. We use an analytical solution for q(t) in the form of a Fourier series:

$$q(t)=q_0\gamma+\frac{2q_0}{\pi}\sum_{1}^{\infty}\frac{\sin\pi n\gamma}{n}\cos\frac{2\pi n}{T}t. \tag{2}$$

After integration of Eq. (1) we shall have, for the steady-state case,

$$u=\theta+\frac{Q\gamma}{Sh}+\frac{2Q}{\pi Sh}\sum_{1}^{\infty}\frac{\sin\pi n\gamma}{n}\cdot\frac{1}{\sqrt{\dfrac{(\omega_n)^2}{\left(\dfrac{h}{HC\rho}\right)^2}+1}}\cos(\omega_n t-\varphi_n), \tag{3}$$

where Q is the electrical power liberated; S is the area of the wall. Here q(t) = Q(t)/SH.

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We see from (3) that the temperature of the wall at the instant of time under consideration is determined by the temperature of the cooling liquid θ, the temperature displacement (constant in time) characterized by the second term of Eq. (3), and the periodically varying third term. It is clearly desirable to achieve conditions under which the amplitude of the third term of the equation may be a maximum. The second term, on the other hand, should be reduced to a minimum, since its presence makes it more difficult to choose the average wall temperature by specifying the temperature θ.

It follows from Eq. (3) that, in order to obtain a large amplitude of the temperature fluctuations, it is essential to maximize $h/HC\rho$. This can only be done by reducing the wall thickness H. It is inappropriate to increase h, as this would reduce the amplitude of the fluctuations in temperature. Hence in designing the chamber it is important to make H as small as technically possible.

We see from Eq. (3) that the amplitude of the temperature fluctuations at the wall depends (inter alia) upon the frequency ω. As we shall later show, the frequency ω is the decisive factor in obtaining supersaturation uniform over the whole volume. It is this quantity which controls the rapidity of damping of the temperature and diffusion waves propagating from the wall. Let us therefore consider all possible cases of relative ω values. According to Eq. (3) we may have the following cases: $\omega \gg h/HC\rho$; $\omega \ll h/HC\rho$; $\omega \sim h/HC\rho$.

For $\omega \gg h/HC\rho$ Eq. (3) takes the form

$$u = \theta + \frac{Q\gamma}{Sh} + \frac{2}{\pi} \cdot \frac{Q}{SHC\rho} \sum_{1}^{\infty} \frac{\sin \pi n \gamma}{n} \cdot \frac{1}{\omega_n} \cos(\omega_n t - \varphi_n). \quad (4)$$

In this case the amplitude of the temperature fluctuations does not depend on h (see (4)) and varies in inverse proportion to ω. The second term of the equation depends on h, but in every case it will be much greater than the amplitude of the temperature fluctuations.

Thus in the case under consideration we have a large temperature displacement with a relatively small amplitude of the temperature fluctuations, i.e., the relation between these quantities is unsuitable for working purposes. However, there is a certain amount of interest in this case, since it enables us to vary the temperature displacement of bias (and hence the average temperature in the chamber) by varying h for a constant amplitude of the temperature fluctuations. There is then no need to use a thermostat; smooth regulation of the temperature in the chamber may be achieved by a smooth variation of h within the limits of the case under consideration ($h \ll \omega HC\rho$).

For $\omega \ll h/HC\rho$ Eq. (3) takes the form

$$u = \theta + \frac{Q\gamma}{Sh} + \frac{2}{\pi} \cdot \frac{Q}{Sh} \sum_{1}^{\infty} \frac{\sin \pi n \gamma}{n} \cos(\omega_n t - \varphi_n). \quad (5)$$

In Eq. (5) the amplitude does not depend on ω and is determined, like the temperature displacement, by the heat-transfer coefficient h. The higher the value of h, the lower is the amplitude of the temperature fluctuations. Hence a relatively high value of $h/HC\rho$ should be obtained by reducing H, avoiding excessive values of the coefficient h. However, the thickness of the wall H is limited by strength considerations. It is practically impossible to use foil thinner than 20 μ. Hence the values of h corresponding to the case in question are quite high, and this reduces the amplitude of the temperature fluctuations in the wall. This case is nevertheless also of great interest, since the amplitude of the temperature fluctuations does not depend on ω, which is extremely convenient for practical research work. It should be noted that, in the case in question, the condition of maximum amplitude of the fluctuations with a minimum temperature displacement may be achieved by suitably choosing the occupation factor γ. We see from Eq. (5) that the temperature u depends on the value of γ. The second term in (5) depends on γ in accordance with the law $\sin \pi n \gamma$, which enables us to use the coefficient γ in order to obtain the maximum amplitude of the temperature fluctuations at the wall with a minimum displacement (relative to the temperature θ). In order to determine the γ for which this condition is satisfied, we must examine the extrema of the difference $([2/\pi] \sin \pi\gamma - \gamma)$, to which the difference between the amplitude of the first harmonic of the third term and the second term of the equation effectively reduces. The resultant value is $\gamma_0 = 0.33$.

Thus for $\gamma_0 = 0.33$ we obtain the maximum possible amplitude of the fluctuations with a minimum temperature displacement, i.e , the relationship most suitable for the work in hand. The second term then makes up only 33% of the maximum possible value, whereas the amplitude of the temperature fluctuations amounts to 86.6% of this.

For $\omega \sim h/HC\rho$ Eq. (3) retains its original form. The temperature condition is affected by both ω and h. However, the optimum working condition may be secured in this case also by once again looking for an extremum of the difference between the amplitude of the first harmonic of the change in temperature (in the third term) and the displacement (the second term of the equation). Let us denote this difference by Δ:

$$\Delta = \frac{Q}{S}\left[\frac{2}{\pi}\,\frac{\sin\pi\gamma}{\sqrt{(\omega HC\rho)^2 + h^2}} - \frac{\gamma}{h}\right]. \tag{6}$$

The quantity Δ depends on many other quantities. However, from the point of view of the investigation of present interest we simply retain γ and h as independent variables, regarding all the other quantities as parameters. Differentiating $\Delta(\gamma h)$ with respect to γ and h, respectively, and equating the resultant expressions to zero, we obtain a system of equations determining the values of γ_0 and h_0:

$$\begin{aligned} &\frac{2\cos\pi\gamma}{\sqrt{(\omega HC\rho)^2 + h^2}} - \frac{1}{h} = 0, \\ &\frac{2\sin\pi\gamma}{\left(\sqrt{(\omega HC\rho)^2 + h^2}\right)^3} + \frac{\gamma}{h^3} = 0. \end{aligned} \tag{7}$$

As a solution to the system of transcendental equations (7) corresponding to the maximum, we have the values $\gamma_0 = 0.3$ and $h_0 = 1.5\,\omega HC\rho$. For these values we obtain the maximum possible amplitude of the temperature fluctuations with a minimum temperature displacement. The heat-transfer coefficient h_0 depends on the frequency ω, the wall material ($C\rho$), and the wall thickness H. However, as already indicated, high values of h are undesirable, since they reduce the amplitude of the temperature fluctuations. Hence H should be made as small as possible and $\gamma = 0.3$ not too high. The resultant relationships are naturally essential when designing the chamber.

Thus we see that the relative value of ω (with respect to $h/HC\rho$) determines both the temperature condition at the wall and also its singularities. On the other hand, the quantity ω determines the manner in which the temperature and diffusion waves propagating from the wall are attenuated and hence the degree of uniformity of the temperature and of the supersaturation in the volume of the chamber. Hence the value of ω should be decided on the basis of those conditions which lead to uniformity of temperature and supersaturation.

In order to determine ω we must study the propagation of the temperature and diffusion waves in the inner region (in the chamber). Here we shall confine ourselves to setting out the final results of an analysis of the propagation of the temperature waves, and shall not specifically analyze the diffusion waves, since the latter are described by completely analogous equations. According to this analysis, the attenuation of the temperature waves close to the wall, i.e., for $r \sim r_0$ (in the case of a cylindrical chamber) and subject to the condition $\sqrt{(w/2a^2)}r \gg 1$, is described by the equation

$$u = A\sqrt{\frac{r}{r_0}}\exp\left\{\sqrt{\frac{\omega}{2a^2}}(r - r_0)\right\}\cos\left[\omega t - \sqrt{\frac{\omega}{2a^2}}(r - r_0)\right]. \tag{8}$$

We see from (8) that the temperature waves propagating from the wall of the chamber suffer rapid attenuation. According to (8) in every case it is essential to make $\sqrt{(\omega/2a^2)}r_0$ as great as possible. This may be achieved both by increasing the frequency ω and by increasing the chamber dimensions r_0. In the case of prespecified chamber dimensions Eq. (8) enables us to determine the value of ω necessary to ensure the desired attenuation of the waves in the zone next to the wall. The quantity ω in turn enables us to find all the thermophysical characteristics needed to ensure a periodic temperature condition at the wall, corresponding to one of the three cases considered ($\omega \ll h/HC\rho$; $\omega \gg h/HC\rho$; $\omega \sim h/HC\rho$).

Let us estimate the period of the temperature fluctuations required in order to obtain a specified degree of uniformity of the temperature in the main volume of the chamber. Let us assume that the chamber wall corresponds to a cylinder 4.5 cm in diameter, $l = 25$ cm long, made of copper foil $H = 20\ \mu$ thick.

TABLE 1. Values of the Coefficient γ_0 and Computing Equations for Determining h_0, δ, A, Q for Three Conditions at the Wall

ω	γ_0	h_0	δ	A	Q
$\omega \sim \frac{h}{HC\rho}$	0,3	$1,5\,\omega HC\rho$	$0,0715\frac{1}{h}\cdot\frac{Q}{S}$	$46,5\frac{1}{\omega}\cdot\frac{Q}{S}$	$\frac{A\omega S}{46,5}$
$\omega \gg \frac{h}{HC\rho}$	0,4	$0,1\,\omega HC\rho$	$0,0953\frac{1}{h}\cdot\frac{Q}{S}$	$88\frac{1}{\omega}\cdot\frac{Q}{S}$	$\frac{A\omega S}{88}$
$\omega \ll \frac{h}{HC\rho}$	0,33	$10\,\omega HC\rho$	$0,0793\frac{1}{h}\cdot\frac{Q}{S}$	$0,1315\frac{1}{h}\cdot\frac{Q}{S}$	$\frac{AhS}{0.1315}$

We see from Eq. (8) that the manner in which the temperature waves attenuate close to the wall is determined by the factor $\sqrt{(\omega/2a^2)}r_0$. Let us specify the value of this factor and require that the temperature fluctuations at a distance of 0.5 cm from the wall (i.e., $r - r_0 = -0.5$) should in amplitude not exceed 1% of the amplitude of the temperature fluctuations at the wall of the chamber. Then in the case of air ($a^2 \approx 0.18$) $\omega = 30.5$ and hence the period T = 0.2 sec. If we require the same damping at a distance of 1 cm from the wall we have $\omega = 7.6$ and the period T = 0.8 sec.

Let us now calculate the corresponding values of the heat-transfer coefficient h and the occupation factor γ for these two examples, and also the temperature displacement, the amplitude of the temperature fluctuations, and the power required in order to obtain the corresponding temperature conditions at the wall of the chamber. For both examples we shall consider three possible conditions at the wall, relating to the three conditions just mentioned.

Table 1 shows the coefficients h_0 and γ_0 derived for these conditions and also the calculating relationships for determining the temperature displacement δ, the amplitude of the temperature fluctuations A, and the power required Q for the copper-foil wall H = 20 μ thick. Using these relationships, we may calculate the effective values for the two examples under consideration. The results of the calculations for all possible wall conditions are presented in Table 2. For practical convenience the values of δ and A are expressed in terms of the ratio Q/S and may be used for various chamber sizes.

For greater clarity, Table 3 gives the results of calculations relating to the examples under consideration for all possible wall conditions with S = 350 cm^2, using a copper-foil wall H = 20 μ thick. The amplitude of the temperature fluctuations is taken as 5°C for every case.

We see from Table 3 that, if greater spatial uniformity is required ($\omega = 30.5$), i.e., if the temperature fluctuations are required to attentuate more rapidly, not only must we have higher values of the frequency ω but we must also have a higher value of the heat-transfer coefficient h_0. Higher values of h_0 in turn lead to an increase in the power required and impose their own limitations on the construction of the system circulating the cooling liquid. According to Table 3, in order to achieve a periodic temperature variation with an amplitude of 5°C at the wall at a frequency of $\omega = 30.5$ a considerably greater power is required for every one of the working conditions that is required in the case of a temperature fluctuation at a frequency of $\omega = 7.6$ with the same amplitude. For each condition a fourfold change in power corresponds to a fourfold change in frequency. We should therefore not use excessive values of ω and present overrigorous demands for spatial uniformity.

We see from the values given in Table 3 that for $\omega \gg h/HC\rho$ there is a considerable temperature displacement $\delta = 33$°C for both examples, in accordance with the foregoing analysis of this case. There is also an unfavorable working relationship between the amplitude of the temperature fluctuations (A = 5°C) and the temperature displacement ($\delta = 33$°C). However, we see from Table 3 that the case under consideration is energetically the most favorable, and as already indicated enables us to vary the temperature displacement δ and hence the mean temperature in the chamber by smoothly varying h for one particular amplitude of the temperature fluctuations (within the range $h \ll \omega HC\rho$). There is accordingly no need to use a thermostat. These characteristics of the case in question naturally make it desirable to create apparatus based on the corresponding principles.

For $\omega \ll h/HC\rho$ the power required is a maximum in both cases. As shown earlier and indicated in Table 1, the amplitude of the temperature fluctuations is independent of the frequency ω. The property is valuable for a number of investigations. However, unless specifically required, this case is inconvenient from the practical point of view, since it is energetically unfavorable, as indicated by the results presented in Table 3.

TABLE 2. Values of the Coefficients γ_0, h_0 and Computing Equations for Determining the Quantities δ, A, Q in the Two Examples in Question (ω = 30.5 and ω = 7.6) for Three Conditions at the Wall with H = 20 μ

ω	ω=30,5					ω=7,6				
	γ_0	h_0, cal/cm² · sec · °C	δ	A	Q	γ_0	h_0, cal/cm² · sec · °C	δ	A	Q
$\omega \sim \frac{h}{HC\rho}$	0,3	$7{,}5 \cdot 10^{-2}$	$0{,}955 \frac{Q}{S}$	$1{,}523 \frac{Q}{S}$	$0{,}657\, AS$	0,3	$1{,}89 \cdot 10^{-2}$	$3{,}78 \frac{Q}{S}$	$6{,}12 \frac{Q}{S}$	$0{,}1635\, AS$
$\omega \gg \frac{h}{HC\rho}$	0,4	$5 \cdot 10^{-3}$	$19 \frac{Q}{S}$	$2{,}88 \frac{Q}{S}$	$0{,}346\, AS$	0,4	$1{,}26 \cdot 10^{-3}$	$75{,}7 \frac{Q}{S}$	$11{,}6 \frac{Q}{S}$	$0{,}0865\, AS$
$\omega \ll \frac{h}{HC\rho}$	0,33	$5 \cdot 10^{-1}$	$0{,}158 \frac{Q}{S}$	$0{,}262 \frac{Q}{S}$	$3{,}8\, AS$	0,33	$1{,}26 \cdot 10^{-1}$	$0{,}63 \frac{Q}{S}$	$1{,}045 \frac{Q}{S}$	$0{,}96\, AS$

TABLE 3. Values of the Coefficients γ_0, h_0 and the Quantities δ, A, Q in the Two Examples in Question (ω = 30.5 and ω = 7.6) for Three Conditions at the Wall with H = 20 μ

ω	ω=30,5					ω=7,6				
	γ_0	h_0, cal/cm² · sec · °C	δ, °C	A, °C	Q, W	γ_0	h_0, cal/cm² · sec · °C	δ, °C	A, °C	Q, W
$\omega \sim \frac{h}{HC\rho}$	0,3	$2{,}7 \cdot 10^{3}$	3	5	1150	0,3	$6{,}8 \cdot 10^{2}$	3	5	286
$\omega \gg \frac{h}{HC\rho}$	0,4	$1{,}8 \cdot 10^{2}$	33	5	605	0,4	$4{,}53 \cdot 10^{1}$	33	5	151
$\omega \ll \frac{h}{HC\rho}$	0,33	$1{,}8 \cdot 10^{4}$	3	5	6650	0,33	$4{,}53 \cdot 10^{3}$	3	5	1680

For $\omega \sim h/HC\rho$ (as indicated by Table 3) the temperature displacement is small (δ = 3°C for an amplitude of A = 5°C). However, the temperature displacement δ cannot be smaller than the amplitude of the temperature fluctuations A, and the low values of δ shown in Table 3 arise from the fact of discarding all the succeeding harmonics (apart from the first) in the expansion of the temperature fluctuations when analyzing Eq. (3). Actually in this case the temperature displacement δ will be equal to the amplitude of the temperature fluctuations, which facilitates the choice and smooth regulation of the temperature of the cooling liquid. We see from Table 3 that this case is energetically acceptable for both examples and is both simple and convenient.

It should be noted that, in view of the close similarity between the mathematical descriptions of the diffusion and heat-conduction processes, if we can satisfy the requirement of adequate spatial uniformity for the temperature we automatically satisfy the corresponding requirements as to the spatial uniformity of the vapor pressure, and hence also the supersaturation.

NOTATION

u	is the temperature at the instant of time t;
t	is the time;
H	is the thickness of wall;
ρ, C	are the density and specific heat of the wall material;
h	is the heat-transfer coefficient between the wall and the cooling medium;
γ	is the occupation factor;
ω, T	are the frequency and period of the oscillations (fluctuations);
A	is the amplitude of the temperature fluctuations;
δ	is the temperature displacement (bias);
Q	is the power required;
r_0, r	are the radius of the cylindrical wall of the chamber and the current radius;
Δ	is the difference between the amplitude of the first harmonic of the series describing the time-periodic variation in wall temperature (in the third term of (3)) and the temperature displacement δ (second term of (3));
a^2	is the thermal diffusivity.

LITERATURE CITED

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