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The correlation of unsteady-state heat transfer data from a sterilizing oven

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Data, showing the pattern of temperature rise in articles undergoing sterilization in an oven under normal working conditions, have been studied. Simple mathematical models, each expressed in terms of a set of differential equations rather than in an explicit analytical form, have been fitted to these data, and the quality of fit and possible applications are here discussed.

To achieve satisfactory sterilization by dry heat it is necessary to heat all parts of an article up to a required (minimum) temperature and to maintain this temperature for a specified period (for example 160°C for 1 hour: British Pharmacopoeia, 1968). Articles to be sterilized are usually stacked in containers in an oven, which is then brought to a temperature above the required minimum. The temperature of the articles themselves rises more slowly, and it is essential to allow sufficient time for the innermost parts to reach the requisite temperature. The heating time may be long, especially if the maximum oven temperature must be severely restricted, due for example to thermal instability of the materials undergoing treatment.

In principle the heating time may be predicted if one has a knowledge of the heat-transfer coefficients at all surfaces under the prevailing conditions, of the system geometry, and of the thermal conductivity and specific heat of the materials involved. Heat transfer coefficients are classically measured by setting up a steady-state experiment, in which the quantity of heat passing through a surface is determined under a constant temperature difference. The apparatus and technique for these measurements can be complex, and the results are dependent on conditions including the fluid flow pattern prevailing. In a sense, too, these measurements are intrinsically artificial, in that the experimental apparatus may differ substantially from the practical configuration, and the relation between the two may be open to debate.

Given the heat transfer coefficients and other requisite data, the problem of predicting the temperature-time profile for an article may be solved analytically only for relatively simple geometrical shapes and for a simple pattern of change in the environmental temperature. Even with substantial simplifying assumptions analytical solutions for particular cases can be extremely complicated (Jaeger, 1945; Carslaw & Jaeger, 1959), and in most cases a numerical solution is required.

In the work outlined here, an attempt has been made to explore an alternative approach to the unsteady-state heat transfer problem. Temperature-time records from an actual sterilization have been examined and mathematical models fitted to them. The models so fitted contain implicitly the heat transfer coefficients and other system properties, although it may not be possible to separate the various quantities contributing to the overall behaviour of the system.

EXPERIMENTAL

The hot-air oven was an electrically-heated, fan-circulated Barlow-Whitney type E3/232S with internal dimensions 96 cm high, 61 cm wide, and 56 cm deep. This was packed with a typical load of articles for sterilization, as shown in Fig. 1. The lower shelf carried tubes and pipettes; the two upper shelves each carried four tin-plate boxes packed with glass Petri dishes. The temperature recordings used were taken at 1 min intervals from each of three thermocouples, respectively (*a*) midway between the four tins on the top shelf, (*b*) in the air space at the centre of one of these tins, and (*c*) in the middle of a Petri dish in the same tin.

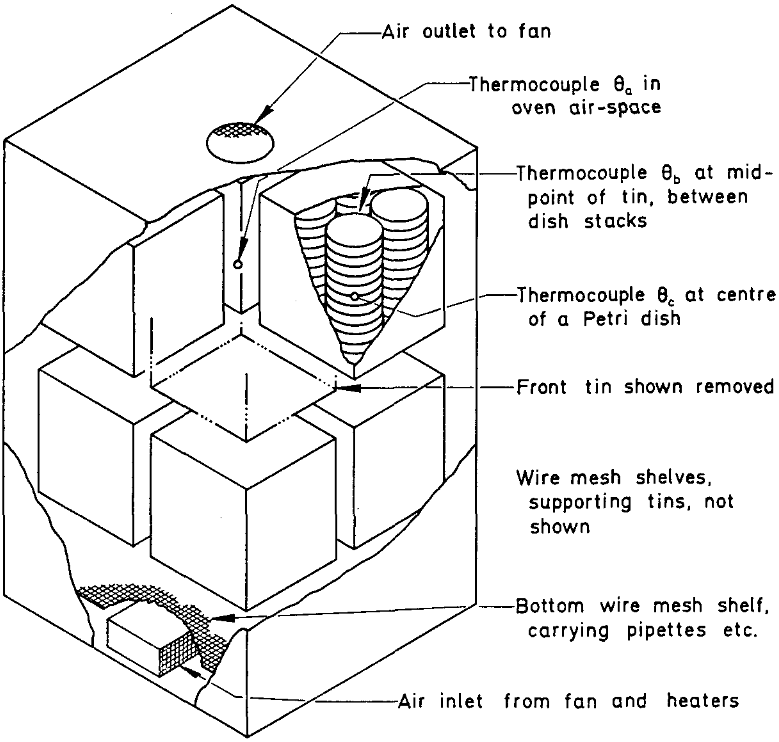

FIG 1. Diagram of the oven and its load, showing the positions of thermocouples.

Mathematical treatment

Let θ_0, θ_1, θ_2 be respectively the temperatures of the air surrounding a tin, of the metal wall of the tin itself, and of the air inside the tin, each assumed to be approximately uniform over the surface. From the definition of a heat transfer coefficient:

$$\text{Heat transfer to tin} = h_{01} A_{01} (\theta_0 - \theta_1)$$

and

$$\text{Heat transfer from tin} = h_{12} A_{12} (\theta_1 - \theta_2)$$

where h_{01}, h_{12} are the local heat transfer coefficients at the inner and outer surfaces and A_{01}, A_{12} are the external and internal surface areas of the tin, respectively.

A net heat gain by the tin will result in a rise in its temperature θ_1, thus:

$$\frac{d\theta_1}{dt} = \frac{1}{C_1}\{h_{01}A_{01}(\theta_0-\theta_1)-h_{12}A_{12}(\theta_1-\theta_2)\} \quad \ldots \quad \ldots \quad (1)$$

or

$$\frac{d\theta_1}{dt} = \frac{1}{C_1}\{p_{01}(\theta_0-\theta_1)-p_{12}(\theta_1-\theta_2)\} \quad \ldots \quad \ldots \quad \ldots \quad (2)$$

where C_1 is the heat capacity of the tin, and

p_{01}, p_{12} are respectively the products $h_{01}A_{01}$,$h_{12}A_{12}$.

The heat transfer to the payload of Petri dishes presents a more complicated problem, since this is thick in the direction of heat flow. A reasonably rigorous treatment would require a stack of dishes to be regarded as a distributed heat-transfer stage, considering it as an infinite set of isothermal, coaxial laminae, each receiving heat from the lamina without, passing heat to that within, and increasing in temperature as a result of the difference; such a treatment would present difficulties due to the relatively complicated shape of the dishes, and would itself disregard asymmetry due to the way the stacks were packed.

A simpler approach is to treat the stack of dishes as a finite number of discrete, notionally-coaxial elements, which behave as the laminae referred to above, and are each supposedly at a uniform temperature at any instant. Such a model has characteristics which approach those of the distributed stage as the number of elements is increased. In the present exercise, only the two simplest forms from this set of possible models have been used.

Model A. This is the grossest possible simplification; the stack of Petri dishes is treated as a single element, at a uniform temperature at any instant. If this has heat capacity C_3 and the temperature is θ_3 an equation analogous to (2) may be obtained

$$\frac{d\theta_3}{dt} = \frac{1}{C_3}p_{23}(\theta_2-\theta_3) \quad \ldots \quad \ldots \quad \ldots \quad \ldots \quad (3)$$

Since the air contained in the tin is of negligible heat capacity the rate at which heat leaves the inner surface of the tin must equal that at which it enters the Petri dishes, thus:

$$p_{12}(\theta_1-\theta_2) = p_{23}(\theta_2-\theta_3) \quad \ldots \quad \ldots \quad \ldots \quad \ldots \quad (4)$$

or

$$\theta_2 = \frac{p_{12}\theta_1 + p_{23}\theta_3}{p_{12} + p_{23}} \quad \ldots \quad \ldots \quad \ldots \quad \ldots \quad (5)$$

Model B. The stack of dishes is treated as composed of two elements, an outer (denoted by subscript 3′) and an inner (subscript 4). The heat transfer equations are:

$$\frac{d\theta_{3'}}{dt} = \frac{1}{C_{3'}}\{p_{23'}(\theta_2-\theta_{3'})-p_{3'4}(\theta_{3'}-\theta_4)\} \quad \ldots \quad \ldots \quad \ldots \quad (6)$$

$$\frac{d\theta_4}{dt} = \frac{1}{C_4}p_{3'4}(\theta_{3'}-\theta_4) \quad \ldots \quad \ldots \quad \ldots \quad \ldots \quad (7)$$

$$\theta_2 = \frac{p_{12}\theta_1 + p_{23'}\theta_{3'}}{p_{12} + p_{23'}} \quad \ldots \quad \ldots \quad \ldots \quad \ldots \quad (8)$$

These two models are represented in block diagram form in Figs 2 and 3.

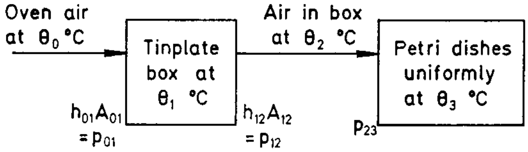

FIG. 2. Block diagram of model A. The temperatures θ_2, θ_3 were compared with the observed values θ_b, θ_c.

Considering the problem in terms of model A, it is possible to solve numerically the set of equations (2), (3) and (5) which represents this model, provided that the initial temperatures and the coefficients **C**,**p** are available and the oven air temperature θ_0 is known as a function of time. The solution may be presented in the form of graphs of θ_1, θ_2, θ_3 against time. The profiles of θ_2, θ_3 will resemble the observed pattern of change of θ_b, θ_c to a greater or lesser degree according to the values of **p** used.

Both model A and model B have been simulated in this way, using smoothed values of the observed air temperatures θ_a as θ_0 and optimizing the vectors **p** to give a best fit, in the least-squares sense, of the models to the observed results. The optimization consisted of minimizing the function

$$\sum\left\{(\theta_2-\theta_b)^2\right\}+\sum\left\{(\theta_3-\theta_c)^2\right\}$$

for model A, or the corresponding function with θ_3 replaced by θ_4 in the case of model B.

The optimization algorithm used was that due to Coggan (1967), based on work by Davidon (1959) and by Fletcher & Powell (1963). The differential equations were solved by Gill's modification of the Runge-Kutta method. The choice of these methods was made primarily on grounds of availability; a general discussion of the treatment of problems of this type has been presented by Rosenbrock and Storey (1966).

In the fitting of model A the parameters p_{01}, p_{12}, and p_{23} were optimized without constraint. To simplify the fitting of model B, the ratio p_{12}/p_{01} was arbitrarily fixed at 1·6 (the ratio optimal for model A); the payload of Petri dishes was treated as 70% outer element, 30% inner element, corresponding roughly to the outer flanged portions and the inner flat portions respectively (Fig. 4). Thus $C_{3'} = 0{\cdot}7\ C_3$ and $C_4 = 0{\cdot}3\ C_3$. With these arbitrary simplifications p_{01}, $p_{23'}$ and $p_{3'4}$ were optimized without constraint.

RESULTS AND DISCUSSION

The time: temperature profiles for the two fitted models A and B are shown in Figs 5 and 6. The observed temperatures θ_b and θ_c are shown for comparison, represented by broken lines.

The simpler model (A) represents the observed behaviour within about 5°C over

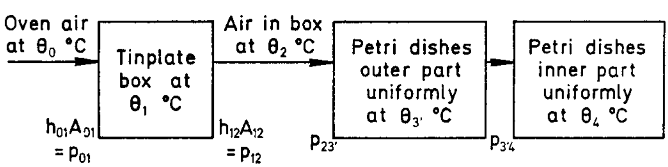

FIG. 3. Block diagram of model B. The temperatures θ_2, θ_4 were compared with the observed values θ_b, θ_c.

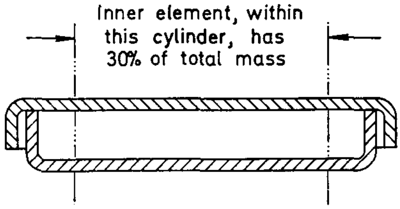

FIG. 4. The two-element model B visualized in terms of a partitioned Petri dish.

the entire range. The observed innermost temperature θ_c rises later and more rapidly than the fitted model; this is the difference to be expected from an attempt to represent a distributed stage by a single exponential transfer stage. The two-element model (B) represents the observed behaviour much more closely—within about 2°C over the entire range. The systematic differences between observations and model still take the same form but are less marked.

The principal numerical data and results are presented in Table 1. It may be noted that the heat capacity of the tinplate container is very much less than that of

Table 1. *Numerical data and results*

Tinplate box: dimensions, cm.	22·0 × 23·3 × 25·4 high	
surface area, cm^2	3220	
weight, g	923	
estimated heat capacity C_1 cal °C^{-1}	102	
Petri dishes: dimensions, cm.	10·2 dia. × 1·8 high	
weight, g	153	
number in box	48	
estimated heat capacity per box, C_3 cal °C^{-1}	1490	
	Model A	Model B
Heat capacity coefficients for elements of dish stacks:		
outer, C_3' cal °C^{-1}	—	1043
inner, C_4 cal °C^{-1}	—	447
Values of fitted heat transfer parameters, cal min^{-1} °C^{-1}		
p_{01}	31·5	32·5
p_{12}	50·5	52·0
p_{23} or p_{23}'	71·1	137·0
$p_{3'4}$	—	33·7
Heat transfer coefficients at surface of box, cal min^{-1} cm^{-2} °C^{-1}		
outside h_{01}	0·0098	(0·010)
inside h_{12}	0·016	(0·016)
$\left\{\frac{1}{h_{01}}+\frac{1}{h_{12}}\right\}^{-1}$	0·0060	0·0062
Overall coefficient, Chu h^{-1} ft^{-2} °C^{-1}	7·4	7·6
Residual variance of inner air temperature about fitted θ_2, °C^2	0·40	1·81
Corresponding standard deviation, °C	0·63	1·34
Residual variance of dish centre temperature about fitted θ_3 or θ_4, °C^2	9·91	2·14
Corresponding standard deviation, °C	3·15	1·46

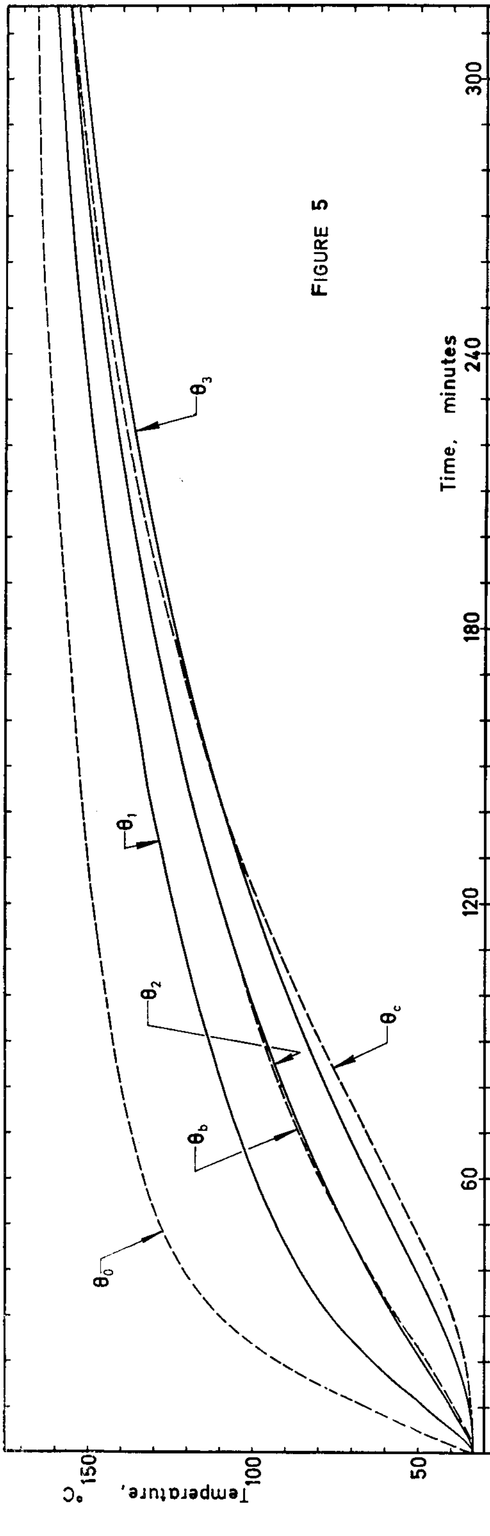

FIGURE 5

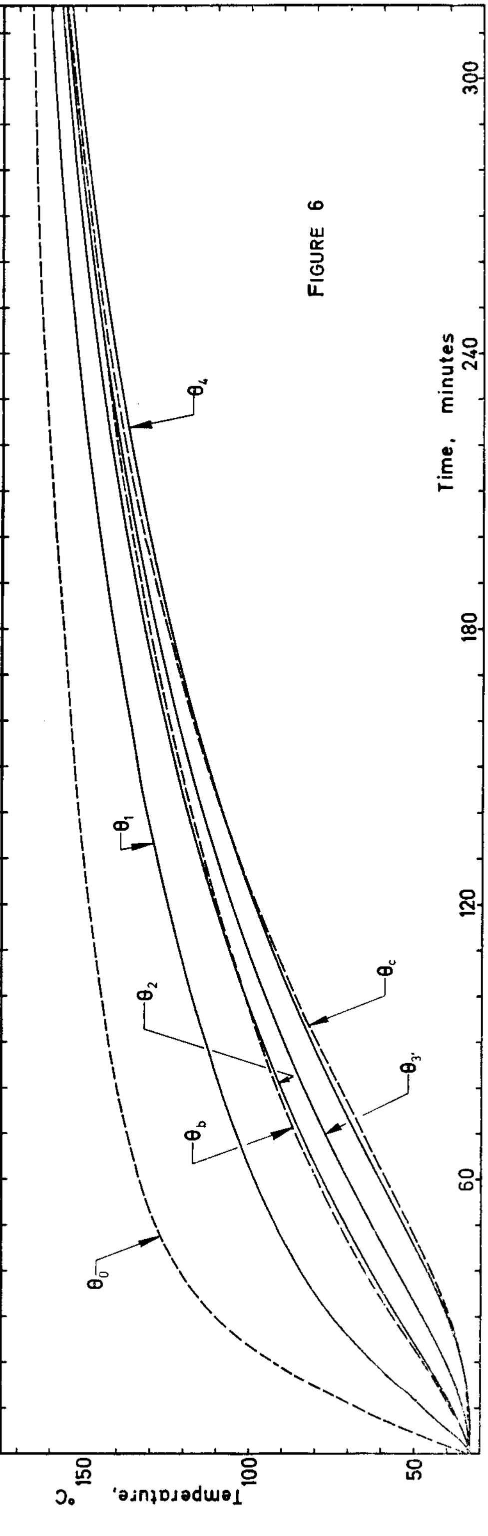

FIGURE 6

the payload of Petri dishes inside it, and a consequence of this was apparent in the process of fitting model A. The values of the individual parameters p_{01}, p_{12}, containing the heat transfer coefficients h_{01}, h_{12} for the outer and inner surfaces of the tin, were clearly much less important than the overall resistance to heat transfer from the outside to the inside of the tin, represented by the overall coefficient in the table. This observation led to the arbitrary decision to fix the ratio p_{01}/p_{12} when fitting model B.

A surprising and as yet unexplained result is that the inner air temperature θ_b was less well represented by model B than by model A. This may be the result of an imperfection of the fitting process rather than of more fundamental origin. As was noted earlier the fitting process used was selected for availability and convenience rather than for fundamental suitability for this purpose. The minimization procedure used had proved highly reliable in previous applications and converged well in initial tests with artificial data. Model A was fitted without excessive difficulty, but with model B convergence was less satisfactory and some attempts with models of this type failed. For this reason more complicated models have not yet been tried.

Conclusions

The practicability has been demonstrated of fitting relatively simple mathematical models to experimental data for a system undergoing sterilization under practical conditions. The models obtained, expressed as a set of simultaneous linear differential equations which contain the heat transfer coefficients in their parameters, yield estimates of these heat-transfer coefficients as a by-product.

The applicability of this approach to design is a matter of conjecture, but it seems probable that correlations obtained in this way, from data for various combinations of sterilizing oven, packing or payload, could be used to predict the behaviour of further, different combinations. Such a treatment might offer practical advantages over either more conventional methods of prediction or a purely empirical approach.

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FIG. 5. Time-temperature profiles for model A.

Broken lines:

θ_0 Oven air temperatures—smoothed observed values from thermocouple θ_a.

θ_b Observed inner air temperature.

θ_c Observed dish-centre temperature.

Continuous lines:

θ_1 Temperature of tinplate container, from model.

θ_2 Inner air temperature, from model (comparable with θ_b).

θ_3 Dish temperature from model (comparable with θ_c).

FIG. 6. Time-temperature profiles for model B.

Continuous lines:

θ_3' Temperature of dish outer element, from model.

θ_4 Temperature of dish inner element, from model (comparable with θ_c).

All other details are as for FIG. 5, above.

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