Modelling optimum processing conditions for the sterilization of prepackaged foods

C. L. M. Silva^{*†}, F. A. R. Oliveira[†] and M. Hendrickx[‡]

Mathematical methods available to optimize heat sterilization of prepackaged foods are critically reviewed. Heat transfer models, with special relevance to those considering external resistance to heat transfer, are discussed. Different optimization methods and constraints are analysed and their applicability is evaluated. Several objective functions suggested in the literature to minimize quality degradation, energy consumption or economic costs are presented along with their impact on optimum conditions. The use of variable heating medium time temperature profile in order to improve sterilization conditions, both in terms of mass average and surface quality retention, is also extensively reviewed. The review suggests that sufficient experimental evidence is lacking. Most models do not provide experimental validation and it is thought that this impairs the potential applications that would arise.

Keywords: sterilization; numerical modelling; optimization; quality

INTRODUCTION

Heat sterilization of foods is a preservative technique that aims to obtain a safe product with a long shelf life and is based on the application of suitable time-temperature conditions to thermally inactivate micro-organisms, its spores and enzymes (Maesmans *et al.*, 1990).

Quality factors, such as colour, flavour, texture or nutrients, are also heat labile. However, the thermal degradation kinetics of these indices is much less temperature-sensitive, allowing the optimization of the process in terms of quality (Teixeira *et al.*, 1969a; Lund, 1977; Holdsworth, 1985). Average quality is important for nutrient retention and other quality

attributes which are important for the consumer but not easily detected sensorially, while optimum surface quality is necessary for quality attributes such as appearance and aroma.

The term 'optimizing thermal processing' can actually refer to different aspects, each of which would, on its own, lead to different results: minimization of toxic compounds (Lund, 1982), minimization of energy consumption and economic costs (Barreiro *et al.* 1984) or maximization of product quality attributes (Teixeira *et al.*, 1969b). It is this last criterion that relates directly to consumer preference and therefore market value.

Although the heat sterilization method has been known since the beginning of the 19th century (Ball and Olson, 1957), the idea of modelling and optimizing processing conditions is relatively recent (Teixeira *et al.*, 1969a; Lund, 1982). The mathematical methods available to optimize the sterilization of prepackaged foods are critically reviewed below and important areas are identified where there is a need for further work.

^{*}Escola Superior de Biotecnologia-UCP, Rua Dr António Bernardino de Almeida, 4200 Porto, Portugal. [‡]Katholieke Universiteit Leuven, Laboratory of Food Technology, Kardinaal Mercierlaan 92, 3001 Heverlee, Belgium. ^{*}To whom correspondence should be addressed

MATHEMATICAL MODELLING OF THE HEAT STERILIZATION OF PREPACKAGED FOODS

The modelling of the heat sterilization of foods requires a mathematical description of the heat flow into the product and the inactivation (degradation) kinetics of microorganisms, enzymes and quality factors (Hallstrom *et al.*, 1988). The energy consumed by the thermal destruction of cells and food quality indices is negligible and therefore the above-mentioned phenomena can be solved independently (Clark, 1978).

Mathematical modelling of heat transfer

Several procedures have been suggested in the literature to solve the temperature distribution during the sterilization of prepackaged foods. These can be divided into three main groups; methods used to model heat transfer to solids, liquids and mixed systems (particulate foods).

Heat transfer into a solid food occurs by conduction and can be described by Fourier's laws. The governing differential equation, for unsteady-state heat transfer (Fourier's second law) into a three-dimensional object, can be written as,

$$\rho C_p \left(\frac{\partial T}{\partial t} \right) = \frac{\partial}{\partial x} \left(k_{xx} \frac{\partial T}{\partial x} \right) + \frac{\partial}{\partial y} \left(k_{yy} \frac{\partial T}{\partial y} \right) + \frac{\partial}{\partial z} \left(k_{zz} \frac{\partial T}{\partial z} \right) \quad (1)$$

using rectangular coordinates (Carslaw and Jaeger, 1959).

Analytical solutions of this equation are only possible for simple geometries, such as slabs, cylinders and spheres, and for relatively simple initial and boundary conditions (e.g. uniform initial temperature distribution and constant surface temperature, i.e. step function of the heating medium temperature). However, with the hardware and computation capacity currently available, numerical methods are a feasible alternative. Two types of numerical solutions are normally used: finite difference and finite element. Both methods can handle problems with initial temperature distributions, non-linear and non-isotropic thermal properties, non-homogeneous systems and time-dependent boundary conditions. The finite difference method is preferred to model regularly shaped objects, while irregular shapes are better modelled by the more complex finite element procedure (Hallstrom *et al.*, 1988; Tucker, 1991).

Since 1969 (Teixeira *et al.*, 1969a, 1969b), many simulation procedures to predict the time-temperature distribution within in-container sterilized conduction heating foods have been published in the literature. Different geometries, types of solution and boundary conditions were considered (Table 1). Most work was developed for simple shapes or boundary conditions. With the exception of the work of Perez-Martin's *et al.* (1988), who modelled the heat transfer into cans with tuna fish and oil using the finite element method, food was always considered homogeneous, linear and isotropic. The head space effect was analysed only by Naveh *et al.* (1983b) and Tucker and Holdsworth (1990, 1991). Naveh *et al.* (1983b) modelled the heat transfer

into a conduction heating product packaged in a glass jar and considered the air heating by conduction. They concluded that products of higher thermal diffusivity indicated a slight shift in the location of the slowest heating zone towards the head space. Tucker and Holdsworth (1990, 1991), to describe the heat transfer into a rectangular container mathematically, used an equivalent heat transfer coefficient at the top to take into account the head space effect and validated the accuracy of the model experimentally. For Biot numbers lower than approximately 10, it has been shown that the global heat transfer coefficient between the heating medium and the food surface lowers the product heating rate (Silva *et al.*, 1992a).

Only few authors have experimentally validated the time-temperature history at the coldest point in the container (Table 1) and a very theoretical approach is normally adopted. However, to assess the usefulness of any mathematical work it is essential to verify the mathematical predictions experimentally.

There is a need to develop heat transfer simulation procedures for non-homogeneous, non-linear or non-isotropic foods, that can include the head space effects and take into consideration non-regular container shapes. Further experimental validation of the theoretical models is also required.

The sterilization modelling of prepackaged liquid foods (convection heating foods) involves the simultaneous solution of the three fundamental transport equations: continuity, motion and energy (Bird *et al.*, 1960).

$$\nabla \cdot \bar{v} = 0 \quad (2)$$

$$\rho \frac{D\bar{v}}{Dt} = \nabla p + \eta \nabla^2 \bar{v} + \rho g \quad (3)$$

$$\rho C_p \frac{DT}{Dt} = k \nabla^2 T \quad (4)$$

for pure fluids with constant density, viscosity and thermal conductivity, and neglecting the viscous dissipation of energy within the fluid. Since these studies are much more complex, they have not received so much attention (Rao and Anantheswaran, 1988). The modelling of prepackaged liquid foods is usually divided into two groups: natural convection heating foods and forced convection heating foods.

A restricted amount of simulation work is available for prepackaged foods heating by natural convection and was reviewed by Rao and Anantheswaran (1988). The pasteurization of a Newtonian liquid food, packed in a glass jar, was first modelled in 1983 (Engelman and Sani, 1983) using the finite element software package FIDAP. Simulated results for velocity distributions agreed well with experimental visual data published by Brandon *et al.* (1981). The temperature and velocity distribution within a Newtonian liquid food packed in a finite cylindrical can was modelled by Datta and Teixeira (1987, 1988). An alternating direction implicit finite difference method was used to solve the governing Navier-Stokes equations in axisymmetric cylindrical coordinates and the results were compared with previously published data. The computer program took up to 10 h of central processing unit time to run in a CDC Cyber 170/730. In 1990, Kumar *et al.* (1990) used

Table 1 Modelling heat transfer into prepackaged conduction heating foods

Reference	Geometry	Products	Type of solution	Boundary conditions	Surface heat transfer coefficient	Experimental validation
Teixeira <i>et al.</i> (1969a, 1969b, 1975b)	Finite cylindrical can	—	Finite-difference 2D	Step-function	h_{∞} (x/k negligible for can wall)	Yes
Manson <i>et al.</i> (1970)	Rectangular container	—	Finite-difference 3D	Variable retort temperature	h_{∞} (x/k negligible for container wall)	No
Singh and Segerlind (1974) De Baerdemaeker <i>et al.</i> (1974)	Finite cylindrical can	—	Finite-element	Step-function	h_{∞} (x/k negligible for can wall)	No
Teixeira <i>et al.</i> (1975a)	Finite cylindrical can	—	Finite-difference 2D	Step-function Sinusoidal function Combination of ramps Single square wave Sequence of steps	h_{∞} (x/k negligible for can wall)	No
Thijssen <i>et al.</i> (1978)	Finite cylinder Sphere Rectangular body	—	Analytical 1D 2D	Step-function	h_{∞} and h_{finite}	No
Saguy and Karel (1979)	Finite cylindrical can	—	Finite-difference 2D	Variable surface temperature	h_{∞} (x/k negligible for can wall)	No
Ohlsson (1980a)	Infinite slab	—	Finite difference 1D	Step-function with linear come-up-time	h_{∞} (x/k negligible)	No
Ohlsson (1980b, 1980c)	Finite cylinder	Fish stew	Finite-difference 2D	Step-function with linear come-up-time	h_{∞} (x/k negligible for container wall)	Yes
Castillo <i>et al.</i> (1980)	Retortable pouch The real shape is not considered	Mixture of corn starch with carboxymethyl-cellulose	Analytical solution 3D	Step-function	h_{∞} (x/k negligible for container wall)	Yes
Tijssen and Kochen (1980)	Finite cylinder Sphere Rectangular body	—	Finite-difference 2D	Variable surface temperature	h_{∞} and h_{finite}	No
Martens (1980)	Infinite slab	—	Analytical 1D	Step-function Variable surface temperature	h_{∞} (x/k negligible for container wall)	Yes
Naveh <i>et al.</i> (1983a)	Finite cylindrical can	Cream style corn Cream style corn with 1% xanthan and 1% avicel Lean pork meat Pumpkin puree	Finite-element 2D	Variable retort temperature	h_{finite}	Yes
Naveh <i>et al.</i> (1983b)	Finite cylindrical can Glass jar	—	Finite-element 2D non-homogeneous	Variable retort temperature	h_{finite}	No
Nadkarni and Hatton (1985)	Finite cylindrical can	—	Numerical solution 2D	Variable surface temperature	h_{∞} (x/k negligible for can wall)	No
Tandon and Bhowmik (1986)	Retortable pouch The real shape is considered	Bentonite solution (10%)	Finite-difference 2D Finite-element 2D	Variable retort temperature	h_{∞} (x/k negligible for container wall)	Yes
Bhowmik and Tandon (1987)	Retortable pouch The real shape is considered	Bentonite solution (10%) Pea puree	Finite-difference 2D	Variable retort temperature	h_{finite}	Yes
Peralta-Rodriguez <i>et al.</i> (1988)	Finite cylindrical can	Tomato pulp	Finite-difference 2D	Variable retort temperature	h_{∞} (x/k negligible for container wall)	Yes
Perez-Martin <i>et al.</i> (1988)	Finite cylindrical can	Albacore	Finite-difference 2D, homogeneous and anisotropic Finite-element 2D, non-homogeneous and anisotropic	Step-function with a linear come-up-time	h_{∞} (x/k negligible for can wall)	Yes

continued

Table 1 continued

Reference	Geometry	Products	Type of solution	Boundary conditions	Surface heat transfer coefficient	Experimental validation
Simpson <i>et al.</i> (1989)	Oval-shaped containers	Green bean puree	Finite-difference 2D	Step-function	h_{∞} (x/k negligible for container wall)	Yes
Hendrickx <i>et al.</i> (1990)	Infinite cylinder Finite cylinder Sphere	—	Analytical 1D 2D	Step-function	h_{∞} (x/k negligible for container wall)	No
Shin and Bhowmik (1990) Bhowmik and Shin (1991)	Finite cylindrical plastic container	Bentonite solution (8%)	Finite-difference 2D, non-homogeneous isotropic	Variable retort temperature	Thermal diffusivity of the wall considered	Yes
Tucker and Clark (1990)	Finite cylindrical plastic can Rectangular polypropylene blocks	Sylgard 180 elastomer	Finite-difference 2D and 3D	Variable retort temperature	h_{finite} (x/k negligible for container wall)	Yes
Tucker and Holdsworth (1990, 1991)	Rectangular polypropylene blocks	Sylgard 170 elastomer	Finite-difference 3D	Variable retort temperature	h_{finite} (x/k considered to estimate an overall heat transfer coefficient) Head space effects considered	Yes
Hendrickx <i>et al.</i> (1993)	Infinite slab Infinite cylinder Sphere	—	Finite-difference 1D	Step-function with a linear come-up-time	h_{∞} (x/k negligible for container wall)	No
Silva <i>et al.</i> (1992a)	Infinite slab Infinite cylinder Sphere	—	Finite-difference 1D	Step-function with a linear come-up-time	h_{finite}	No
Banga <i>et al.</i> (1991)	Finite cylindrical can	—	Finite-difference 2D	Step-function Variable retort temperature	h_{∞} (x/k negligible for can wall)	No
Hendrickx <i>et al.</i> (1992a, 1992b)	Finite cylindrical can	—	Finite-element 2D	Step-function with a linear come-up-time	h_{∞} and h_{finite}	No

1D, 2D, 3D: one-, two- and three-dimensional

the finite element software package FIDAP to simulate the sterilization of a thick viscous Newtonian liquid food, with a temperature-dependent viscosity, packed in a cylindrical container. The results were compared with simulated data for a conduction heating food and it was verified theoretically that natural convection moved the slowest heating point to the bottom centre and caused the bottom of the can to heat up slower. This agrees with experimental results reported by Zechman and Pflug (1989).

There is no research work concerning the mathematical modelling of prepackaged forced convection heating liquid foods. The reason is the complexity of establishing the correct boundary conditions for this situation and the lack of information of physical and thermal properties as a function of temperature and time (Rao and Anantheswaran, 1988). Another obstacle is a strong requirement for CPU time.

There is also a lack of research concerning the modelling of heat flow and experimental studies for the sterilization of prepackaged convection heating foods. The heat transfer into convection heating foods is normally studied experimentally in terms of heat penetration parameters or in terms of dimensionless groups (Rao and Anantheswaran, 1988; Rao *et al.*, 1985; Soulé and Merson, 1985; Rotstein *et al.*, 1988; Anantheswaran and Rao, 1985a, 1985b). When the heating is modelled mathematically several assumptions are usually made, such as: uniform initial temper-

ature and the liquid behaving as a lumped heat capacity system, constant physical and thermal properties and constant heat transfer coefficients. These assumptions are often inappropriate.

The modelling of heat transfer into containers with liquid and particulates (mixed systems) is even more complex. It involves the solution of both the temperature and velocity distribution within the liquid and the temperature profile inside the particles. It is a common procedure to assume a uniform fluid temperature, constant heat transfer coefficients (heat transfer coefficient between the heating medium and container wall, between container wall and liquid food, and between liquid food and food particle), uniform initial temperatures for both particles and fluid, particles with equal sizes and uniformly distributed, and constant physical and thermal properties for both the fluid and particles (Stoforos and Merson, 1990a). These assumptions lead to an approximate but easier solution. Several authors used this model to determine liquid-particle heat transfer coefficients (Lenz and Lund, 1979; Deniston *et al.*, 1987; Stoforos and Merson, 1990b). Rumsey (1984) developed a computer program, using a finite difference method, to model the heat transfer into cans containing liquid with spherical particles based on the above-mentioned assumptions. He compared his results with experimental data and concluded that the model did not accurately predict the temperature at the surface of the spheres. The difference might be due to

the accuracy of the heat transfer coefficient values introduced in the model. No research work has been carried out considering more complex conditions, such as: the particle geometries (cubes, cylinders), particle size distribution, mix of particles with different characteristics (geometries, physical properties).

Kinetics of thermal inactivation (degradation)

Mathematical models for thermal inactivation (degradation) of microorganisms, enzymes or quality factors, are indispensable to quantify thermal treatment effects and to make automatic computational process control possible (Cohen and Saguy, 1985; Holdsworth, 1990). First-order reaction kinetics are considered by most authors to be an acceptable empirical model for chemical and microbiological reactions involved in heat sterilization (Hayakawa, 1978):

$$\frac{dX}{dt} = -k_T X. \quad (5)$$

Within a given temperature range, the temperature dependence of k_T can be expressed according to the Arrhenius equation:

$$k = A \exp(-E_a/RT\theta). \quad (6)$$

Using thermal processing nomenclature the empirical parameters D and z are more frequently used than the $k-E_a$ model:

$$D = \frac{\ln(10)}{k} \quad (7)$$

$$D = D_{T_{ref}} \cdot 10^{(T_{ref}-T)/z}. \quad (8)$$

The relationship between the z value and the activation energy (E_a) (Ohlsson, 1988) can be expressed as

$$z = \frac{\ln(10) \cdot R \cdot T \cdot T_{ref}}{E_a}. \quad (9)$$

Although the empirical $D-z$ model and the $k-E_a$ kinetic model are mathematically different, both produce reliable and consistent results within the range of temperatures normally used in thermal processing (Jonsson *et al.*, 1977; Holdsworth, 1985, 1990).

Several authors reviewed the kinetic data available, both for microbial inactivation and for nutrient thermal degradation (Lund, 1975, 1977; Pflug and Odlaug, 1978; Ohlsson, 1988; Holdsworth, 1990). Nutrient and quality degradation showed significantly less temperature sensitivity than the inactivation of vegetative cells and spores allowing the optimization of the process in terms of quality (Lund, 1977).

Table 2 Optimizing sterilization processes

Reference	Constraint	Objective function	Type of solution	Boundary conditions	Experimental validation
Teixeira <i>et al.</i> (1969)	ASM	$(N/N_O)_{ave}$	Finite-difference 2D; h_{in}	Step-function	No
Teixeira <i>et al.</i> (1975a)	ASM	$(N/N_O)_{ave}$	Finite-difference 2D; h_{in}	Step-function Sinusoidal function Combination of ramps Single square wave Sequence of steps	No
Thijssen <i>et al.</i> (1978)	ASM	$(N/N_O)_{ave}$	Analytical 1D and 2D; h_{in} and h_{finite}	Step-function	No
Saguy and Karel (1979)	ASM	$(N/N_O)_{ave}$	Finite-difference 2D; h_{in}	Variable surface temperature	No
Ohlsson (1980a)	F_C	C_{ave} and C_{surf}	Finite-difference 1D; h_{in}	Step-function with linear come-up-time	No
Ohlsson (1980b, 1980c)	F_C	C_{ave} and C_{surf}	Finite-difference 2D; h_{in}	Step-function with linear come-up-time	No
Thijssen and Kocken (1980)	ASM	$(N/N_O)_{ave}$	Finite-difference 1D and 2D; h_{in} and h_{finite}	Variable surface temperature	No
Martens (1980)	ASM	$(N/N_O)_{ave}$	Analytical 1D; h_{in}	Step-function Variable surface temperature	No
Nadkarni and Hatton (1985)	ASM	$(N/N_O)_{ave}$	Numerical solution 2D; h_{in}	Variable surface temperature	No
Hendrickx <i>et al.</i> (1990)	F_C	$(N/N_O)_{surf}$	Analytical 1D and 2D; h_{in}	Step-function	No
Hendrickx <i>et al.</i> (1993)	F_C	C_{surf}	Finite-difference 1D; h_{in}	Step-function with linear come-up-time	No
Tucker and Holdsworth (1990, 1991)	F_C	C_{ave}	Finite-difference 3D; h_{finite}	Step-function	No
Silva <i>et al.</i> (1992a)	F_C	C_{surf}	Finite-difference 1D; h_{finite}	Step-function with linear come-up-time	No
Banga <i>et al.</i> (1991)	ASM and F_C	$(N/N_O)_{ave}$ and $(N/N_O)_{surf}$	Finite-difference 2D; h_{in}	Constant and variable surface temperature	No
Hendrickx <i>et al.</i> (1992a, 1992b)	F_C	C_{surf}	Finite element 2D; h_{in} and h_{finite}	Step-function with linear come-up-time	No

CRITERIA FOR HEAT STERILIZATION OF PREPACKAGED FOODS

In-container heat sterilization is based on the application of suitable time-temperature profiles, within the food, in order to obtain a commercially sterile product (Richardson *et al.*, 1988).

Two processes are equivalent in terms of the thermal destruction of a given organism if they have the same impact on it. In general, there are two approaches to assess the impact of a process (Table 2): (i) a sterility value at the coldest point (F_c) or (ii) an integrated sterility value (F_s) representing the volume average survival of microorganisms (Stumbo, 1973):

$$F_c = \int_0^{t_p} 10^{(T_c - T_{refm})/z_m} dt \quad (10)$$

$$(X/X_0)_c = 10^{-F_c/D_{refm}} \quad (11)$$

or

$$ASM = \frac{1}{V_T} \int_0^{V_T} 10^{-(F(V)/D_{refm})} dV \quad (12)$$

$$F_s = D_{refm} \log (ASM) \quad (13)$$

The sterility value at the coldest point assures a minimum sterility in all points of the food, therefore this is the most adequate criterion.

The target microorganism normally used to determine food safety is *Clostridium botulinum* (Richardson *et al.*, 1988), for which $z_m = 10^\circ\text{C}$ (Pflug *et al.*, 1978).

It is impossible experimentally to detect levels of microorganisms as low as 1 in 10^9 units (criterion for final level of microorganisms to avoid public health hazard (Pflug, 1987) and usually the sterility of the processed food product is experimentally evaluated using the time-temperature data at the coldest spot (Equations 10 and 11). This thermal process evaluation method is known as the 'general method' or 'Bigelow method'. A complete review on mathematical procedures to determine sterilization values is presented by Hayakawa (1977, 1978).

Based on years of experience, the sterilization industry has been processing low-acid prepackaged foods with F_0 values calculated in such a way that the endpoint probability of microorganism survival is smaller than a defined safety value. To avoid risks due to unknown conditions, a safety factor (dependent on the heating conditions) is usually multiplied by the sterility value yielding a target F_0 value (Pflug, 1987).

F_c or ASM values have been used as a constraint in studies of optimizing sterilization conditions (Table 2). However, the F_c value is more adequate for evaluating the thermal impact of processing conditions on the final sterility of the product.

OPTIMIZING THE STERILIZATION CONDITIONS

The optimization problem for a sterilization process involves three parts (Norback, 1980): (i) the identification of the design variables that can be controlled, (ii)

the requirements that must be met, and (iii) the definition of the objective function (the mathematical expression of what is to be optimized).

The major constraint for optimizing heat sterilization processes is the target lethality required to obtain a safe product (Holdsworth, 1985).

The heat flow into the food is the lethal agent and its rate can be controlled by the design variables: heating medium time-temperature profile, material properties, food geometry and dimensions (Teixeira *et al.*, 1975a). For a given food product, there is an infinite number of heating medium time-temperature profiles which produce a final product with the required lethality. However, when an objective function is specified, a unique optimum time-temperature profile is obtained (Teixeira *et al.*, 1969a).

Work on the optimization conditions has so far focused on conduction heating foods only and is limited to theoretical considerations. Experimental validation of optimum conditions defined using mathematical procedures has not yet been attempted (Table 2).

Choosing an objective function

There are two main alternatives for an objective function: definition of the conditions for minimizing costs or for maximizing quality, usually quantified in terms of quality parameters (texture, level of nutrients, flavour, etc.). It must be mentioned that both lead to the optimization of the economic efficiency of the process. Lund (1982) referred the importance of minimizing the formation of toxic compounds although information on kinetic data for these type of reactions is very limited (Banga *et al.*, 1991).

Optimizing costs

Barreiro *et al.* (1984) calculated optimum constant sterilization temperatures to minimize the energy requirement of a batch retort and using the sterility value at the coldest point as a constraint. The energy consumption of a batch heat sterilization process was estimated from a thermal energy balance on a stationary retort. However, they did not consider the effect of processing conditions on the final food quality. This can obviously lead to unacceptable products for a competitive market.

Banga *et al.* (1991) considered the minimization of the total processing time in order to improve the productivity of the process. They used the same constraint but added a restriction on the final product surface quality retention, to ensure an adequate quality of the final product. They concluded that variable retort temperature shows significant advantages over constant temperature when the productivity is to be maximized.

Optimizing quality retention

Most research work (Table 2) focuses on the processing conditions leading to a maximum final level of quality factors (Hendrickx *et al.*, 1992b). Quality optimization is possible because the degradation kinetics of quality are much less temperature-sensitive than the kinetics of microorganism destruction (Lund, 1977). The principle

of high-temperature short processing time (HTST) would therefore lead to significant quality improvements and attempts have been made particularly in the development of aseptic processing and rotary sterilization. Unfortunately, the heat transfer rate into conduction heating foods is slow. Taking into consideration the high thermal resistance of some enzymes, it is apparent that the HTST principle finds limited application in this type of foods (Saguy, 1988).

During heat treatment, food quality changes can be divided in two phases. During the first phase, an improvement in quality is observed due to desirable changes induced by the cooking (e.g. softening of texture, development of cooked food aroma). In the second phase (after the optimal cooking is reached) further processing only causes undesirable quality degradation. Only in HTST processing is the cooking level within a reasonable range of the optimum; for other processes food is usually overcooked and the goal is to minimize the thermal degradation (Ohlsson, 1988).

Depending on the quality parameter in question, its optimization can be considered in terms of maximizing surface or volume average quality. The first concept is of interest when optimizing appearance and aroma, while the second one is a more suitable indicator for taste, consistency or nutrient retention (Ohlsson, 1980a; Silva *et al.*, 1992b).

Surface quality optimization

In 1962, Mansfield (1962) proposed the concept of cook (or C) value to compare thermal processes in terms of quality degradation. This parameter is defined similarly to the F_0 value:

$$C_i = \int_0^{t_p} 10^{(T_i - T_{refq})/z_q} dt. \quad (14)$$

When calculated at the surface, it is equivalent to the quality index retention (Silva *et al.*, 1992b):

$$(N/N_0)_{surf} = 10^{-C_{surf}/D_{refq}}. \quad (15)$$

Although optimal constant temperatures had been calculated to optimize the surface quality of prepackaged foods (Ohlsson, 1980a, 1980b, 1980c; Hendrickx *et al.*, 1990, 1992a, 1992b, 1993; Banga *et al.*, 1991; Silva *et al.*, 1992a) (Table 2), only in 1990 was a generalized approach to this calculation suggested (Hendrickx *et al.*, 1990).

Hendrickx *et al.* (1990) correlated optimal temperatures, for infinite surface heat transfer coefficient conditions and one-dimensional containers, with (i) the heat penetration parameter, f_h ; (ii) the target F_c value; (iii) the z_q value; (iv) the initial temperature of the food, T_0 . A multilinear regression equation correlated well with the results. Optimal temperature increases with target F_c value, T_0 and z_q value and decreases with the f_h parameter. Hendrickx *et al.* (1993) extended this equation to consider the effect of the heating medium come-up-time (time required for the heating medium to reach holding temperature) and the cooling sterilization period was taken into consideration in the lethality calculation. It was verified that the initial temperature of the food, the heating medium come-up-time and the cooling period have a small influence

on the optimal temperature. Optimal temperatures decrease linearly with the $\log(f_h)$. This result confirms the advantage of using smaller conduction distance containers (such as trays and pouches compared with conventional cans or glass jars), for which the f_h parameter is smaller and therefore optimal temperatures are higher leading to higher surface quality retention (Silva *et al.*, 1992a).

Even less published work considers the existence of heat transfer resistance at the surface (Table 2). Silva *et al.* (1992a) developed a multi-linear regression equation for finite heat transfer coefficients, similar to the one reported by Hendrickx *et al.* (1993). External resistance was quantified in terms of the Biot number ($Bi = h \cdot l/k$). Optimal temperature was found to increase with $c_1 \cdot \ln(z_q) + c_2/Bi + c_3 \cdot z_q/Bi$, where c_1 , c_2 and c_3 are constants. It was also found that initial product temperature and heating medium come-up-time have a small influence on optimal temperature.

Hendrickx *et al.* (1992a, 1992b) applied and validated the referred generalized regression equations to two-dimensional objects (finite cylindrical cans and retortable pouches). When the Biot number is finite, the temperature at the surface is not uniform and depends on the position. Optimal sterilization temperatures were therefore studied as a function of surface position for different can dimensions (Hendrickx *et al.*, 1992b). Cans where the half-height to radius approaches one, showed the most uniform optimal surface quality distribution.

Experimental validation of the calculated optimal sterilization conditions was not carried out. This validation would require: (i) selection of an adequate quality indicator, (ii) quantification of its kinetics to thermal degradation, (iii) characterization of the heat penetration into the container filled with the product, and (iv) processing the product using different sterilization temperatures in order to find the best conditions. This procedure may be quite laborious, which explains the lack of experimental validation.

Average quality optimization

Two approaches have been used to optimize volume average quality retention (Table 2): (i) maximizing volume average cook-value

$$C_{ave} = \frac{1}{V_T} \int_0^{V_T} \int_0^{t_p} 10^{(T - T_{refq})/z_q} dt dV \quad (16)$$

or maximizing volume average quality retention

$$(N/N_0)_{ave} = \frac{1}{V_T} \int_0^{V_T} 10^{-(C(V)/D_{refq})} dV. \quad (17)$$

They are not however, mathematically equivalent. Volume average cook-value (C_{ave}) does not consider the effect of the D_{refq} value on the optimum processing conditions (Silva *et al.*, 1992b).

Volume average cook-value was used as objective function by Ohlsson (1980a, 1980b, 1980c) and Tucker and Holdsworth (1990, 1991). Although for high D_{refq} values (e.g. for vitamins) the two objective functions (Equations 16 and 17) are equivalent, for low D_{refq} values (e.g. for texture and colour) the use of C_{ave} can result in a significant underestimation of the optimal processing temperature (Silva *et al.*, 1992b). As a

general conclusion, Ohlsson (1980c) stated that optimal temperatures for maximizing overall quality of an infinite slab container are about 7.5 to 10°C higher than the correspondent optimum to maximize surface quality (Table 2). Optimal temperature to maximize overall quality is higher than the one to maximize surface quality because with higher temperatures, higher overprocessing at the surface is compensated by higher overall quality.

All research in this area (Table 2) involves just a few case studies. A generalized approach to the problem is not available and the relation between optimal temperatures and the D_{refq} value has not yet been studied. The linear relationships mentioned in surface quality optimization between optimal temperature and relevant variables are no longer valid for overall quality optimization (Hendrickx *et al.*, 1990).

The influence of a finite surface heat transfer coefficient on optimal temperature has not yet been systematically studied (Thijssen *et al.*, 1978; Thijssen and Kocken, 1980; Tucker and Holdsworth, 1990, 1991) and it is expected that for low Biot numbers the optimal temperature increases. This effect is important for plastic containers and headspace effects. Experimental validation of predicted results is also lacking.

Variable sterilization temperature

The idea of using a variable heating medium temperature to improve product quality has always been considered. However, it was impractical to determine experimentally, by trial and error, the optimum retort time-temperature history. All research in this area deals with conduction heating foods and none considered the effect of a finite surface heat transfer coefficient.

Teixeira *et al.* (1975a) studied four types of boundary conditions (constant function, step functions, ramp functions and sinusoidal functions). It was concluded that the introduction of different surface time-temperature profiles did not offer significant advantages to the final level of overall quality retention, given the limitative step of slow heat transfer rate of conduction heating foods. For the same case study, the nutrient retention resulting from the four profiles were within 5% of each other. However, there were differences of 27% in processing time between constant temperature and sinusoidal function and based on an economic point of view, the fastest process (sinusoidal function) is the best choice. Four different types of retort time-temperature profiles only were tested and it was not possible to ensure that the 'best' profile was one of them.

Saguy and Karel (1979), used the maximum principle theory of Pontryagin to optimize thiamin retention during sterilization of conduction heating canned foods. This mathematical method allows the optimization of a multivariable problem. The authors showed the possibility of finding a unique variable time-temperature profile, but, for the cases studied, an improvement of only about 2% on the total thiamin retention was indicated. It was concluded that constant temperature profile was nearly as good as other methods for optimizing global quality.

Thijssen and Kochen (1980) extended the 'short-cut' method developed by Thijssen *et al.* (1978), which is an

user-friendly tool to calculate optimal sterilization temperatures, in order to apply it to variable temperatures of heating and cooling. However, the effect of different profiles on the quality retention was not studied.

Nadkarni and Hatton (1985) applied Pontryagin's minimum principle of optimal control theory to a distributed parameter model for optimizing overall nutrient retention. It was concluded that bang-bang control of the retort temperature, where the heating and cooling rates should be as fast as possible, is the optimal control strategy. Moreover, single-point switching is best, i.e. there should be only one heating-cooling cycle. It was claimed that this is the optimal control strategy, because it offers no difficulties for practical implementation. In terms of overall nutrient retention, this strategy led to the same values obtained in other work.

All research carried out in the field of variable retort temperature was aimed at obtaining a better final overall quality and it was concluded that variable sterilization temperature did not yield significant improvements. This result can be explained by the slow response of temperatures inside containers to variances of the heating medium temperature (Nicolai and De Baerdemaeker, 1992).

In 1991, Banga *et al.* (1991) studied optimum variable time-temperature profiles for maximizing surface quality. It was concluded that the final retention can be increased by about 20% when compared with the optimum for constant sterilization temperature. However, only one case study was considered and there is a need to investigate more thoroughly the potentialities of a variable sterilization temperature for this type of optimization.

Optimum variable time-temperatures profiles need to be studied in terms of maximizing surface quality and/or economical aspects. The influence of finite surface heat transfer coefficients, different container geometry or size is yet to be investigated.

OPTIMIZATION PROCEDURES

The calculation of the processing time required to reach the sterilization criteria (constraint) for a given time-temperature profile (design variable), by means of an iterative procedure, is common to all work published in this field. Hendrickx *et al.* (1993) suggested the following time increment

$$\Delta t_h = (F_c - F_O)/10(T_h - 121.15^\circ\text{C})/z_m \cdot 60 \text{ (s)} \quad (18)$$

where F_c is the target lethality at the coldest point and F_O is the lethality reached with the current holding time. This equation avoids the target sterility at the coldest point to be passed over.

Most authors (Table 3) determine optimal temperatures manually, i.e. for different constant temperatures or temperature profiles the final quality retention, at the surface or global, is calculated and plotted as a function of the design variable. The optimum is then obtained by graphical observation. In 1985, Holdsworth (1985) reviewed the optimization techniques available. However, few had been used in the field of thermal processing of foods.

Table 3 Optimization method

Reference	Method
Teixeira <i>et al.</i> (1969b)	Manual
Teixeira <i>et al.</i> (1975a)	Manual
Thijssen <i>et al.</i> (1978)	Manual
Saguy and Karel (1979)	Pontryagin's maximum principle
Ohlsson (1980a)	Manual
Ohlsson (1980b, 1980c)	Manual
Thijssen and Kocken (1980)	Manual
Nadkarni and Hatton (1985)	Pontryagin's minimum principle
Hendrickx <i>et al.</i> (1990)	Davis, Swann and Campey
Hendrickx <i>et al.</i> (1993)	Davis, Swann and Campey
Tucker and Holdsworth (1990, 1991)	Manual
Silva <i>et al.</i> (1992a)	Davis, Swann and Campey
Banga <i>et al.</i> (1991)	ICRS/DS
Hendrickx <i>et al.</i> (1992a, 1992b)	Manual

Saguy and Karel (1979) were the first to apply an optimization method (Table 3) in order to calculate optimum sterilization conditions. The maximum principle of Pontryagin was used to determine systemically the best time-temperature profile to minimize overall quality degradation. The minimum principle of Pontryagin was used by Nadkarni and Hatton (1985) to optimize the control strategy of a retort and maximize the final global nutrient retention.

Hendrickx *et al.* (1990, 1993) and Silva *et al.* (1992a) used the Davis, Swann and Campey method (Saguy, 1983) to calculate optimal constant temperatures.

In 1991, Banga *et al.* (1991) proposed a new algorithm, ICRS/DS, for the solution of fixed terminal time optimal control problems. The method is based on a combination of a robust parameterization of the control function and a computationally efficient non-linear programming algorithm. The objective was to calculate optimum variable sterilization temperature in order to maximize surface and overall retention or minimize processing time. However a very significant computation time is required.

The main challenge remaining is the development of optimization methods that are more efficient in terms of accuracy and of reduction of computation time required.

RESEARCH NEEDS

Although research on the modelling of heat transfer into prepackaged conduction heating foods has been published in the literature, there is still a need for mathematical models for non-homogeneous, non-linear or non-isotropic foods, that can include the headspace effects and take into consideration non-regular container shapes. Work such as that presented by Perez-Martin *et al.* (1988), Naveh *et al.* (1983b) and Tucker and Holdsworth (1990, 1991) are an example that should be followed. More experimental work should be carried out to assess proposed models.

In relation to the mathematical description of the heat transfer into convection heating and particulate foods, the lack of research is more noticeable. With the hardware available nowadays it should be possible to develop more sophisticated models that can include more realistic boundary conditions. Such models would find a huge application in determination of parameters,

such as heat transfer coefficients, by minimizing the differences between simulated and experimental data. Rumsey (1984) gives a good example of a model that could be easily improved.

All research carried out in the area of optimizing sterilization conditions of prepackaged foods deal with conduction heating foods. This is a consequence of the poor understanding of the heat transfer process in prepackaged convection heating and particulate foods.

The use of objective functions that take into consideration economical aspects as well as quality, have yet to be defined and the calculated correspondent optimum conditions have to be determined. The work of Banga *et al.* (1991), where the processing time is minimized using as a constraint a limit on the minimum final quality, is an example of how objective functions may be redefined. In relation to quality maximization, an objective function considering both surface quality (that is important in terms of sensory quality) and volume average quality (that is important in terms of nutrient retention) could also be of practical interest, since both may be important for consumer preference.

Optimum overall quality conditions have yet to be systematically studied as a function of all relevant variables. Research work similar to that developed by Hendrickx *et al.* (1990, 1993) and Silva *et al.* (1992a) for surface quality, applied to average quality would be of interest.

For surface quality, the extension of the work of Banga *et al.* (1991) to the calculation of optimum heating medium time-temperature profiles, considering variable surface resistance, is required.

The main challenge remaining in the field of optimization procedures is the development of optimization methods that are more efficient in terms of accuracy and of reduction of computation time required.

Experimental validation is still uncommon and regarding sterilization process optimization it is completely lacking. This validation would require: (i) selection of an adequate quality indicator, (ii) quantification of its kinetics of thermal degradation, (iii) characterization of the heat penetration into the container filled with the product, and (iv) processing the product using different sterilization temperatures in order to find the best conditions. Although such work would be extensive, it is essential to assess the usefulness of any mathematical procedure and should therefore be considered as a priority in this field.

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NOMENCLATURE

A	Pre-exponential factor, frequency factor or Arrhenius factor (s^{-1})
ASM	Mass average survival of microorganisms
Bi	Biot number ($h \cdot l/k$) (dimensionless)

C	Cook-value (min)	T_O	Initial temperature of the food ($^{\circ}\text{C}$)
C_{ave}	Volume average cook-value (min)	T_{ref}	Reference temperature ($^{\circ}\text{C}$)
C_i	Cook-value at position i (min)	T_{refm}	Reference temperature for the microorganism ($^{\circ}\text{C}$)
C_{surf}	Surface cook-value (min)	T_{refq}	Reference temperature for the quality factor ($^{\circ}\text{C}$)
$C(V)$	Cook-value in the volume element dV (min)	$\bar{v}$	Mass average velocity vector (m/s)
C_p	Specific heat (J/(kg K))	V	Volume (m^3)
dV	Volume element (m^3)	V_T	Total volume (m^3)
D	Decimal reduction time (time required for the number or concentration of spores, microorganisms or quality factor to be reduced by a factor of 10 at a given temperature) (min)	X	Level of microorganisms, enzymes or quality index
D_{Tref}	Decimal reduction time at the reference temperature (min)	$(X/X_O)_c$	Survival of microorganisms in the coldest spot
D_{refm}	Decimal reduction time at the reference temperature T_{refm} , for the microorganism (min)	z	z value (number of degrees Celsius required to reduce the D value by a factor of ten) ($^{\circ}\text{C}$)
D_{refq}	Decimal reduction time at the reference temperature, T_{refq} , for the nutrient or quality factor (min)	z_m	z value for the microorganism ($^{\circ}\text{C}$)
D/Dt	$\partial/\partial t + \nabla \cdot \bar{v}$ = substantial derivative	z_q	z value for the nutrient or quality factor ($^{\circ}\text{C}$)
E_a	Activation energy (J/mol)	η	Dynamic viscosity of Newtonian liquid (Pa.s)
f_h	Heat penetration parameter: slope factor of a heating curve (time required for the difference between heating medium temperature and food temperature to decrease by a factor of 10) (min)	θ	Absolute temperature (K)
F_O	Sterility value at reference temperature 121.15 $^{\circ}\text{C}$ and $z_m = 10^{\circ}\text{C}$ (min)	ρ	Density (kg/m^3)
F_c	Sterility value at the coldest spot (min)	∇	del or nabla operator = $\partial/\partial x + \partial/\partial y + \partial/\partial z$
F_S	Integrated sterility value (min)		
$F(V)$	Sterility value in the volume element dV (min)		
g	Acceleration due to gravity (m/s^2)		
h	Equivalent surface heat transfer coefficient (between the heating medium and the food) ($\text{W}/(\text{m}^2\text{K})$)		
h_{finite}	Finite equivalent surface heat transfer coefficient (between the heating medium and the food) ($\text{W}/(\text{m}^2\text{K})$)		
h_{∞}	Infinite equivalent surface heat transfer coefficient (between the heating medium and the food) ($\text{W}/(\text{m}^2\text{K})$)		
k	Thermal conductivity ($\text{W}/(\text{mK})$)		
k_{xx}	Thermal conductivity in the x direction ($\text{W}/(\text{mK})$)		
k_{yy}	Thermal conductivity in the y direction ($\text{W}/(\text{mK})$)		
k_{zz}	Thermal conductivity in the z direction ($\text{W}/(\text{mK})$)		
k_T	Rate constant at temperature T (s^{-1})		
l	Characteristic length (m)		
N	Level of nutrients or quality factor at time t		
N_O	Initial level of nutrients or quality factor		
$(N/N_O)_{ave}$	Volume average quality retention		
$(N/N_O)_{surf}$	Surface quality retention		
p	Fluid pressure (Pa)		
R	Universal gas constant (8.314 J/(mol K))		
t	Time (s)		
t_h	Holding time (s)		
t_p	Thermal processing time (s)		
T	Temperature ($^{\circ}\text{C}$)		
T_c	Temperature in the coldest spot ($^{\circ}\text{C}$)		
T_h	Holding temperature ($^{\circ}\text{C}$)		
T_i	Temperature at position i ($^{\circ}\text{C}$)		

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