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A dynamic object-oriented model for efficient simulation of microbial reduction in dispersed turbulent flow

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Abstract

To account for microbial reduction and food quality changes, first-order chemical reaction was added to a model for axial-dispersed plug flow (ADPF), which was recently developed for efficient¹ computation of time-dependent, large flow systems. It was shown that the extended ADPF model gives accurate results for typical parameter values in liquid-food applications. The analysis was based on Laplace transforms and the models were written in the object-oriented language Modelica, as objects in a library structure which is being developed to simulate complex liquid-food process lines and their control systems. The models were also implemented in such a way as to efficiently account for non-isothermal reactions in heat exchangers. Furthermore, to account for changes in flow rates, a model of dynamically calculating the “true” holding time in a channel was developed. Simulations showed that the model performs better than a straightforward alternative.

Keywords: Chemical reaction, Microbial reduction, Tube reactor, Dispersion, Residence time distribution, Dynamic model, Dynamic simulation, Liquid food, Microorganisms, Heat exchanger.

¹ In the terminology of this paper, efficient is related to computation time for a given level of accuracy.

Article outline

Nomenclature

1. Introduction
 2. Chemical reactions in a proposed approximation of axial-dispersed plug flow
 - 2.1 Laplace transforms and transfer functions
 - 2.2 Modified model
 - 2.3 Implementation of true residence time
 - 2.4 Frequency analysis
 - 2.5 Temperature dependency and the choice of representative temperature
 3. Model implementation and simulation
 - 3.1 Steady state
 - 3.2 Flow transients
 4. Results
 - 4.1 Start-up and steady state
 - 4.2 Flow transients
 5. Summary
 6. Conclusions
- Acknowledgements
Appendix
References

Nomenclature

C	Volumetric or microbial concentration, kg/m^3 or m^{-3}
C_0	Initial volumetric or microbial concentration, kg/m^3 or m^{-3}
\vec{C}	C delayed by transport through the channel, i.e. $\vec{C}(t) = C(t - \tau)$, e.g. $C_{out}(t) = \vec{C}_{in}(t) = C_{in}(t - \tau)$, kg/m^3
c_a	Correction factor defined by Eq. (22), -
c_b	Correction factor defined by Eq. (24), -
D	Dispersion coefficient in axial direction, m^2/s
D_r	Microbial decimal reduction time at the reference temperature T_r , s
E_a	Energy of activation in Arrhenius equation, $k = k_0 e^{\frac{E_a}{R_g T}}$, J/mol
f	Frequency, s^{-1}
\mathbf{G}	Transfer function, i.e. ratio of Laplace transforms for output and input signals
i	Imaginary part of complex numbers, -
k	Chemical reaction constant depending on the temperature, s^{-1}
k^*	Dimensionless chemical reaction constant, $k^* = \tau k$, -
k_0	Chemical reaction constant, s^{-1}
L	Length of flow channel, m
L_0	Length of channel with ideal plug flow, m
N	Number of continuously-stirred tanks in the dispersion model, -
Pe	Péclet number defined by vL/D (the ratio of convective flow rate to dispersive flow rate), -
Q	Volumetric flow rate, m^3/s
R	Radius or hydraulic radius, m
Re	Reynolds number defined by $2\rho Rv/\mu$, -

R_g	Universal gas constant, 8.31451 J/mol K
s	Complex argument of the Laplace transform, s^{-1}
s^*	Dimensionless argument of the Laplace transform, $Js/v = \tau s$, -
T	Absolute temperature, K
T_r	Reference temperature for microbial reduction kinetics, K
\dot{T}	Rate of temperature change, K/s or °C/s
t	Time, s
\bar{t}	t delayed by a virtual transport through the channel, i.e. $\bar{t}(t) = t(t - \tau)$, s
t^*	Dimensionless time, $t^* = \frac{t}{\tau}$, -
V	Total volume of channel, m^3
V_0	Volume of channel with ideal plug flow in the new model (see Fig. 1), m^3
V_N	Volume of one of the N continuously stirred tanks, m^3
v	Mean velocity over a channel cross-sectional area, m/s
x	Axial spatial coordinate (along the fluid channel), m
z_r	The microbial decimal reduction temperature increase, °C or K

Greek letters

ΔX	Change in X
μ	Dynamic viscosity defined by $\mu = \sigma / \dot{\gamma}$, Pa s
ρ	Density, kg/m^3
τ	Average transport time (dwell time or holding time) for a fluid passing through a channel (see Fig. 1), V/Q or L/v , s More generally, to account for dynamic delay, i.e. varying velocities: $\tau = \tau(v(t)) : L = \int_0^{\tau} v(t) dt$
τ_0	Transport time (dwell time or holding time) for a fluid flowing through all the N plug-flow pipes in the proposed model (see Fig. 1), $L_0/v = V_0/Q$, s More generally, to account for dynamic delay, i.e. varying velocities: $\tau_0 = \tau_0(v(t)) : L_0 = \int_0^{\tau_0} v(t) dt$
τ_{0N}	As above for τ_0 for the plug-flow pipe, with the total length L_0/N , in one of the N control volumes in the proposed model
τ_N	Time constant for a fluid flowing through one of the N continuously stirred tanks in the new model (see Fig. 1), V_N/Q , s
ω	Angular frequency defined by $2\pi f$, radians/s
ω^*	Dimensionless angular frequency, $L\omega/v = \tau\omega$, -

General superscripts

A^*	Dimensionless form of variable A (further explained above for each variable)
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General subscripts

$ADPF$	Axial-dispersed plug-flow (ADPF) model
$ADPFC$	Axial-dispersed plug-flow with first-order chemical reaction (ADPFC) model
$PFNCST$	Dispersion model combining plug flow with the N -continuously-stirred-tanks (PFNCST) model

<i>PFNCST^C</i>	New proposed dispersion model with chemical reaction, combining plug flow with the N-continuously-stirred-tanks model and first-order chemical reactions (PFNCST ^C)
<i>PFNCST^{CX}</i>	Dispersion model with chemical reaction combining plug flow with the N-continuously-stirred-tanks model and first-order chemical reactions (PFNCST ^{CX}). This model has no static compensation factor and gives a static error.
<i>i</i>	Continuously stirred tank number
<i>in</i>	Into the system
<i>out</i>	Out of the system

Other general symbols

A	Laplace transform of <i>A</i>
\vec{X}	<i>X</i> delayed by transport through a channel, i.e. $\vec{X}(t) = X(t - \tau)$, e.g. $X_{out}(t) = \vec{X}_{in}(t) = X_{in}(t - \tau)$,

Model acronyms

ADPF	Axial-dispersed plug flow
ADPF ^C	Axial-dispersed plug flow with chemical reaction
N-CST	<i>N</i> continuously stirred tanks
PFNCST	Plug flow combined with <i>N</i> continuously stirred tanks
PFNCST ^C	Plug flow combined with <i>N</i> continuously stirred tanks and chemical reaction of first-order (= proposed model)
PFNCST ^{CX}	Plug flow combined with <i>N</i> continuously stirred tanks and chemical reaction of first-order. This model has no static compensation factor and gives a static error.

1. Introduction

Simulation gives important insight into the behaviour of dynamic systems and is becoming more and more important in the development of complex liquid-food processing equipment and lines. The basis for simulations is dynamic models. We are engaged in developing such models (Skoglund, 2003; Skoglund, Årzén & Dejmek, 2006; Skoglund & Dejmek, 2006; Skoglund & Dejmek, 2007) in the language Modelica (Modelica Association, -; Tiller, 2001). The Modelica language is non-causal, object-oriented, and suitable for physical modelling.

In liquid-food processing plants, e.g. dairies, heat treatment is a standard method of inactivation of microorganisms. The mechanism is generally well described by first-order chemical reactions, e.g. Kessler (2002), p. 137. Heat treatment is often performed by first heating the food in a heat exchanger, then holding the food at the desired temperature by letting it flow through a tube with a specified volume (“holding tube”) corresponding to a desired time at a given volumetric flow rate, and finally cooling the food in another heat exchanger. Sometimes the final stage of heating (the last few degrees of temperature increase) is performed by direct steam injection instead of indirect heating by a heat exchanger. In such cases the initial cooling is performed by flash boiling.

During this heating/cooling process the liquid food is normally flowing turbulently through the system. To calculate the chemical reactions in fluids passing through tubes, i.e. tube reactors, the residence time distribution is commonly used. In food sterilization and pasteurization applications – to be on the safe side – only one residence time is used, and that is the time corresponding to the breakthrough time, i.e. the time taken for the fastest particle in the fluid to flow from the inlet to the channel exit. Unless the flow is of plug-flow type, the breakthrough time is shorter than the mean residence time due to dispersion (diffusion, turbulence and velocity profile). The breakthrough time divided by the mean residence time is called the efficiency factor, and can be calculated according to Kessler (2002), p. 15. The effective holding tube length is the actual length multiplied by the efficiency factor. Furthermore, since the flow in cases like this, i.e. turbulent flow with high Reynolds numbers, is well described by the axial-dispersed plug flow (ADPF) model, e.g. Levenspiel & Smith (1957) and Serpemen & Deckwer (1974), it is proposed here that calculations of chemical reactions be based on ADPF with dispersion coefficients, as an alternative to residence times and efficiency factors. Since the proposed model accounts for the total effect, not just the breakthrough time, it should give a more exact calculation of the chemical reaction.

Chemical reactions in heat exchangers have been studied previously. Most studies deal with steady-state conditions, e.g. Hawthorn, Ackerman & Nixon (1968) and Phillips, Lauschke & Peerhossaini (1997). Regarding dynamic simulation, Galle, Agar & Watzenberger (2001) worked with gas flows and Haugwitz (2005) with liquids. In all of these studies the thermal balance was affected by the enthalpy of chemical reactions. However, in the present model the thermal effect of the chemical reaction is ignored as in liquid-food applications it is many orders of magnitude smaller than the typical temperature measurement accuracy.

In the present work an extension of the previously reported models for heat exchangers (Skoglund, Årzén & Dejmek, 2006) and ADPF (Skoglund & Dejmek, 2007) was developed to account for first-order chemical reactions, responsible for both the reduction of microorganisms and the loss of some nutrients in food, e.g. proteins (Kessler, 2002, p. 165). The heating, holding and cooling in a typical food process were simulated.

Furthermore, since the production capacity of heat treatment of liquid food is often varied, it is of interest to be able to simulate the effect of flow variations on chemical reactions. Therefore, the dynamic effect on chemical reactions due to variations in the holding time caused by flow changes was modelled and simulated in a novel way.

2. Chemical reactions in a proposed approximation of axial-dispersed plug flow

For turbulent flow in pipes, the ADPF model can be employed (Levenspiel & Smith, 1957; Serpemen & Deckwer, 1974; Taylor, 1954). With a sink term corresponding to first-order chemical reaction the mass balance is given by Eq. (1).

$$\frac{\partial C}{\partial t} + v \frac{\partial C}{\partial x} - D \frac{\partial^2 C}{\partial x^2} + kC = 0 \quad (1)$$

The first-order chemical reaction accounts for both reduction of microorganisms and loss of some nutrients in food, e.g. proteins (Kessler, 2002, p. 165).

To understand the dynamics, it is appropriate to carry out further analysis in the frequency domain by Laplace transformation.

2.1 Laplace transforms and transfer functions

Laplace transform of Eq. (1) is given by:

$$(k + s)\mathbf{C} + v \frac{\partial \mathbf{C}}{\partial x} - D \frac{\partial^2 \mathbf{C}}{\partial x^2} = 0 \quad (2)$$

For a semi-infinite pipe where the exit concentration is studied at $x = L$, this equation represents a system with the following transfer function. (See Appendix.)

$$\mathbf{G}_{ADPF^C}(s) = e^{\frac{vL}{2D}} e^{-\frac{vL}{2D} \sqrt{1 + \frac{4D}{v^2}(k+s)}} \quad (3)$$

which, expressed with the dimensionless variables $s^* = \tau s$, $k^* = \tau k$ and $Pe = vL/D$, becomes:

$$\mathbf{G}_{ADPF^C}(s^*) = e^{\frac{Pe}{2}} e^{-\frac{Pe}{2} \sqrt{1 + \frac{4}{Pe}(k^* + s^*)}} = e^{-\frac{Pe}{2} \left(\sqrt{1 + \frac{4}{Pe}(k^* + s^*)} - 1 \right)} \quad (4)$$

Skoglund & Dejmek (2007) showed how the ADPF model can be approximately translated into a series of N identical control volumes, each consisting of a pipe with plug flow (transport delay τ_{0N}), in series with one continuously stirred tank (the PFNCST model). This is illustrated in Fig. 1. The agreement between this model and the ADPF model was demonstrated with frequency analysis. Furthermore, it was shown that the PFNCST model was computationally more efficient than the N -continuously-stirred-tanks (N-CST) model.

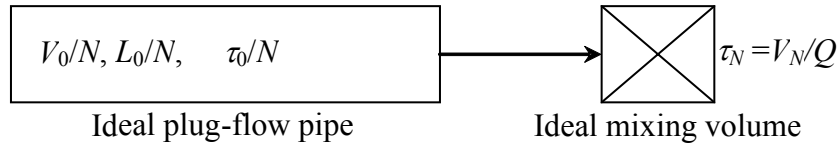


Fig. 1. One of N identical “control volumes” of the PFNCST model.

In the following we will expand the PFNCST model to include first-order chemical reactions and analyse its accuracy relative to the ADPF model. Hence, we formulate the mass balance of the PFNCST model with chemical reaction according to Fig. 1. After rearranging the control volumes to obtain one ideal plug-flow pipe and N continuously stirred tanks, we arrive at Eq. (5) for each of the N continuously stirred tanks.

$$\text{For } \forall i \in [1, N]: V_N \frac{\partial C_i}{\partial t} + QC_i - QC_{i-1} + kV_N C_i = 0 \quad (5)$$

where

$$C_0(t) = C_{in}(t - \tau_0)e^{-k\tau_0} \quad (6)$$

Eq. (6) states that the inlet concentration to the first tank is equal to the inlet concentration to the studied pipe delayed by the transport-delay time τ_0 and changed concentration by chemical reaction during the same time, i.e. solution to Eq. (18) below. Thus it represents the delay and chemical reactions in all (N) ideal plug-flow pipes together. The combined transfer function of Eqs. (5) and (6) is

$$\mathbf{G}_{PFNCST^{CX}}(s) = \frac{e^{-\tau_0(k+s)}}{(1 + \tau_N(k+s))^N} \quad (7)$$

where the time constant $\tau_N = V_N/Q$ has been introduced. This transfer function corresponds to an arbitrary input concentration, i.e. boundary condition for $x = 0$ of Dirichlet type. It also corresponds to the output concentration at $x = L$ for a semi-infinite pipe causing no boundary effects at $x = L$.

Volume conservation and the average transport time $\tau = V/Q$ gives:

$$V = V_0 + NV_N \quad (8a)$$

or in terms of residence times

$$\tau = \tau_0 + N\tau_N \quad (8b)$$

The condition for the PFNCST model to be a good approximation of the ADPF model presented by Skoglund & Dejmek (2007) was:

$$V_N = V \sqrt{\frac{2}{NPe}} \quad (9a)$$

or in terms of residence times

$$\tau_N = \tau \sqrt{\frac{2}{NPe}} \quad (9b)$$

Thus the relationship between the volume of the plug flow pipe (V_0) and the perfect mixing cells (V_N) is determined by the Péclet number (Pe) and the number of mixing cells (N). It was demonstrated that the accuracy is good for small N and increases as N increases to the limit of $Pe/2$.

In present case the limiting conditions¹ (from Eq. (13) in Skoglund & Dejmek (2007)) include k and will be valid for sufficiently low values of k . In food applications, that is normally the case and the validity of the assumption will be confirmed in the frequency analysis below. Inserting Eqs. (8b) and (9b) into Eq. (7) and using the dimensionless variables $k^* = \tau k$ and $s^* = \tau s$ yields:

$$\mathbf{G}_{PFNCST^{CX}}(s^*) = \frac{e^{-\left(1 - \sqrt{\frac{2N}{Pe}}\right)(k^* + s^*)}}{\left(1 + \sqrt{\frac{2}{NPe}}(k^* + s^*)\right)^N} \quad (10)$$

This is identical to replacing s^* with $(k^* + s^*)$ in Skoglund & Dejmek (2007), Eq. (21). Comparing Eqs. (4) and (10) shows that the models have different static responses.

$$\mathbf{G}_{ADPF^C}(0) = e^{-\frac{Pe}{2}\left(\sqrt{1 + \frac{4k^*}{Pe}} - 1\right)} \quad (11)$$

$$\mathbf{G}_{PFNCST^{CX}}(0) = \frac{e^{-k^*\left(1 - \sqrt{\frac{2N}{Pe}}\right)}}{\left(1 + \sqrt{\frac{2}{NPe}}k^*\right)^N} \quad (12)$$

The error is shown in Fig. 2, clearly indicating that it is small, though not zero, for typical parameter values.

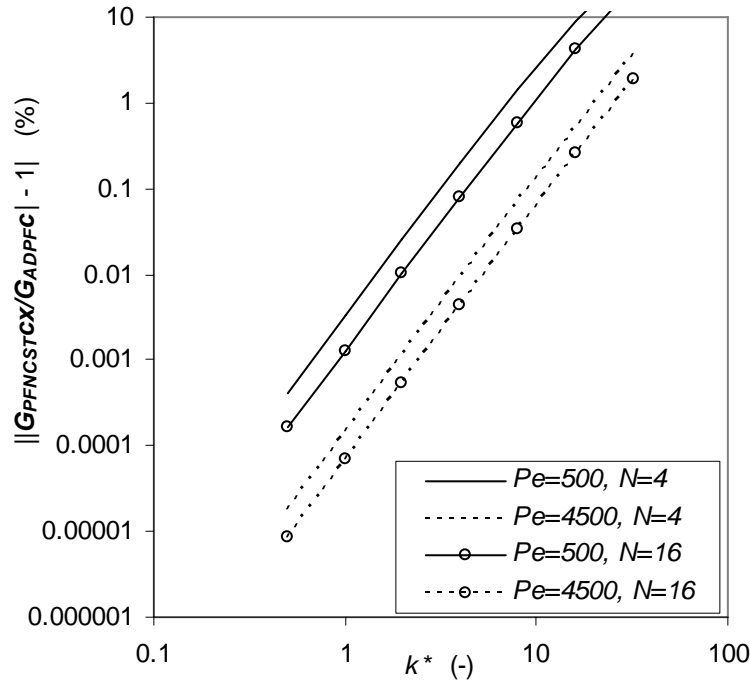


Fig. 2. Static error (i.e. for $\omega = 0$) of the $PFNCST^{CX}$ model, compared with the $ADPF^C$ model, for two values of the Péclet number, each for two values of N .

¹ $\left|\frac{4D}{v^2}(k+s)\right| \ll 1$ and $|N\tau_N(k+s)| \ll 1$

2.2 Modified model

While accurate dynamic modelling is the major thrust of our effort, food industry process lines operate most of the time at steady state or quasi-steady state conditions, and thus static error of the model is of paramount importance. Therefore we suggest a modified model, which fulfils this requirement and includes the no chemical reaction condition ($k = 0$) as a limiting case, i.e. a model that converges to the PFNCST as $k \rightarrow 0$. The modified model uses the PFNCST model to account for the dispersion and dynamics (Eq. (10) with $k^* = 0$) and superimposes upon it the static gain from the ADPF^C model (Eq. (11)) accounting for static reaction kinetics. This essentially ad hoc choice will be justified in the frequency analysis in section 2.4, which shows the accuracy of the model. The modified model (PFNCST^C) leads to the following transfer function,

$$\mathbf{G}_{PFNCST^C}(s^*) = \mathbf{G}_{ADPF^C}(0) \cdot \mathbf{G}_{PFNCST}(s^*) = e^{-\frac{Pe}{2} \left(\sqrt{1 + \frac{4k^*}{Pe}} - 1 \right)} \frac{e^{-\left(1 - \sqrt{\frac{2N}{Pe}}\right)s^*}}{\left(1 + \sqrt{\frac{2}{NPe}}s^*\right)^N} \quad (13)$$

which expressed in terms of τ_0 (instead of τ in $k^* = \tau k$), by utilizing Eqs. (8b) and (9b), is

$$\mathbf{G}_{PFNCST^C}(s^*) = e^{-\frac{Pe}{2} \left(\sqrt{1 + \frac{4\tau_0 k}{Pe}} - 1 \right)} \frac{e^{-\left(1 - \sqrt{\frac{2N}{Pe}}\right)s^*}}{\left(1 + \sqrt{\frac{2}{NPe}}s^*\right)^N} \quad (14)$$

This applies to the whole channel. When discretized, each control volume (Fig. 1) has the transfer function:

$$\left[\mathbf{G}_{PFNCST^C}(s^*)\right]^{1/N} = e^{-\frac{Pe}{2N} \left(\sqrt{1 + \frac{2\tau_{0N}k}{Pe}} - 1 \right)} \frac{e^{-\left(1 - \sqrt{\frac{2N}{Pe}}\right)\frac{s^*}{N}}}{1 + \sqrt{\frac{2}{NPe}}s^*} \quad (15)$$

Where $\tau_{0N} = \tau_0/N$ was introduced for the transport delay in each plug-flow part of the N control volumes. This is the proposed dynamic model for taking ADPF into account simultaneously with first-order chemical reaction.

2.3 Implementation of true residence time

It is immediately obvious that in real 3D systems there is a difference between the actual residence time of a particle and the average residence time, due to the spatial variation of transport velocity. However, due to the temporal variation of transport velocity, even in one dimensional approximation there is a difference between the instantaneous residence time, and the actual residence time of a particle, the transport delay. The used software platform Modelica/Dymola can account for true transport delay according to Eq. (16b) below of any variable including time. The working principle is like a conveyor belt with varying speed, which means that any variable that should be delayed by transportation is loaded into a vector that is shifted with the transportation speed (flow velocity). The length of the vector corresponds to the length of the conveyor belt (pipe). Thus a true match between a volume passing through a channel element and its corresponding transport time according to Eq. (16a) will be available if time is stored into the vector.

$$\tau_{0N}(t) = t - \bar{t}_{in} \quad (16a)$$

This is a way to calculate the dynamic delay, τ_{0N} , according to its general definition:

$$\tau_{0N}(t) = \tau_{0N}(v(t)) : \frac{L_0}{N} = \int_0^{\tau_{0N}} v(t) dt \quad (16b)$$

Eq. (16a) gives the actual residence time, τ_{0N} , for the fluid that exits the channel as the difference between the actual time, t , and the time when the fluid entered the channel, \bar{t}_{in} , which corresponds to the concentration of the fluid when it entered the channel, \bar{C}_{in} . This novel method was used to achieve a good dynamic model of the chemical reaction since the true transport time according to Eq. (16a), rather than the instantaneous transport time $\tau = L/v$, is the time the fluid “experiences” on its path along the channel. The performance of this method is demonstrated later in this paper.

2.4 Frequency analysis

To proceed, we had to assess the accuracy of the assumption in section 2.2, i.e. that Eq. (13) is a good approximation of Eq. (3). Thus we determined the error in the model regarding amplitude and phase. Eqs. (4) and (13) were used to plot these errors. Figs. 3a, 3b, 3c and 3d show curves for the relative error in amplitude, $\|\mathbf{G}_{PFNCSTC}(i\omega^*) / \mathbf{G}_{ADPFC}(i\omega^*) - 1\|$, and the error in phase shift, $|\arg(\mathbf{G}_{PFNCSTC}(i\omega^*)) - \arg(\mathbf{G}_{ADPFC}(i\omega^*))|$. It can be seen that both errors are small over a wide range of ω^* , Pe and k^* relevant for normal liquid food applications with turbulent flow, even for moderate values of N .

The relevance of the range of Pe is exemplified by applying the typical values of geometry (L and R) flow (v), fluid (ρ and μ) and reaction parameters (k , i.e. k_0 , E_a and T , see section 2.5 and 3.1). For example if $v = 1$ m/s, $L = 20$ m, $2R = 0.0486$ (a standard tube), $\rho = 997$ kg/m³ and $\mu = 0.0013$ Pa s gives $Re = 3.7 \cdot 10^4$ and $Pe = 1100$ (by using Eq. (25)).

For the reaction constant the extreme value of $k^* = 60$ in Figs. 3c and 3d corresponds to the very high reaction rate for *Clostridium botulinum* spores at 140 °C, and a holding time (τ) of 4.4 s, or the lower rate at 130 °C but for $\tau = 40$ s.

The range of ω^* is justified by the fact that concentration variations normally can be considered small. The highest frequency is determined by the fastest changes, and in liquid food applications, e.g. dairies, the most dramatic variation concerns changeover of valves, which seldom takes place in a shorter time than 2 second for one stroke, and twice that time for a full period. In the frequency domain it corresponds to 0.25 Hz. Our estimation is therefore that most variations can be considered as frequencies well below 0.25 Hz. With a long holding time, τ , of 30 s it means that maximum of $\omega^* \approx 50$. The validity of the frequency range is also supported by the previously performed analysis of axially-dispersed plug-flow (Skoglund & Dejmek, 2007)). In that analysis a pulse response (with frequency content on the higher side of the range) was compared with the exact solution.

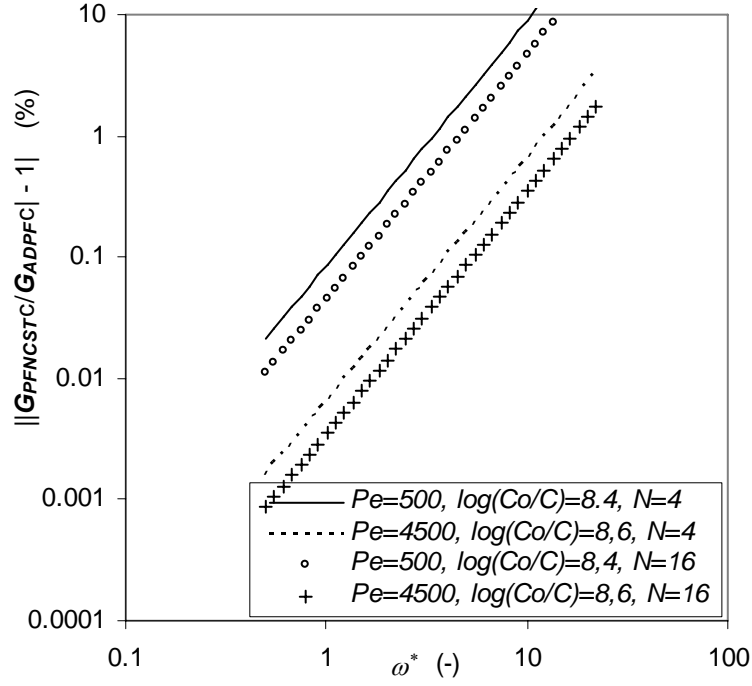
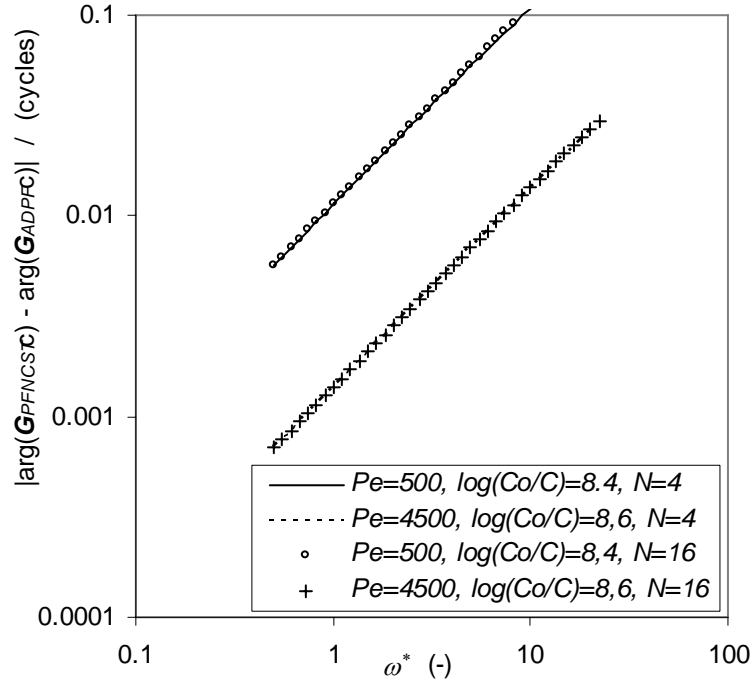
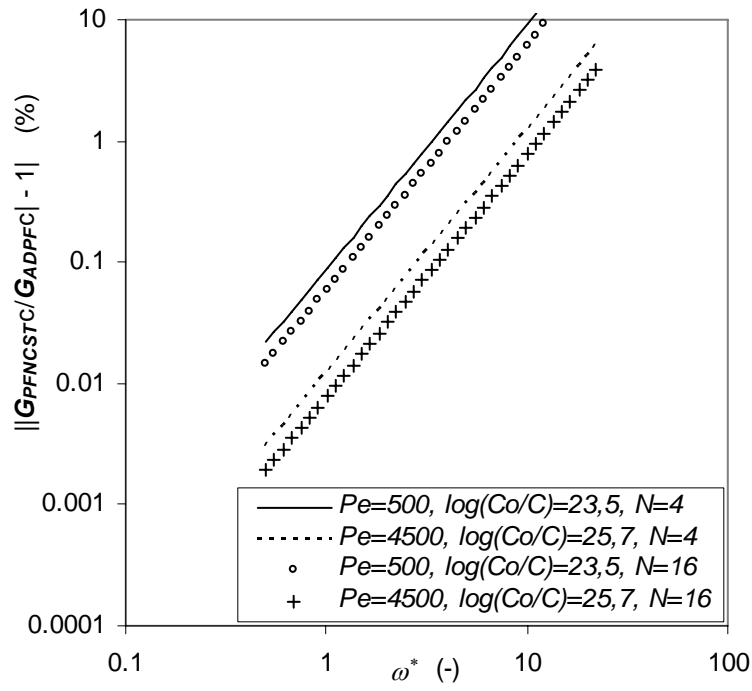


Fig. 3. Accuracy of the proposed model for different degrees of dispersion and chemical reaction rates and at different discretization, N . The plot is made for two values of the Péclet number, each for two values of N . The given values of $\log(C_0/C)$ are valid for stationary conditions, i.e. $\omega = 0$, according to Eq. (11).

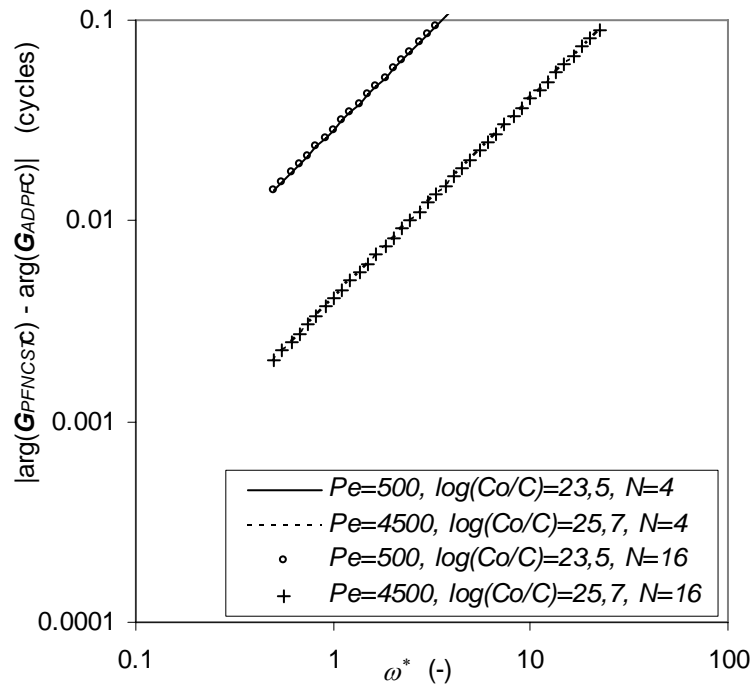
a. The accuracy is displayed as the error in terms of the relative amplitude difference $\|\mathbf{G}_{PFNCSTC}(i\omega^*) / \mathbf{G}_{ADPFC}(i\omega^*) - 1\|$. The plot is made for $k^*=20$.



b. The accuracy is displayed as the error in terms of the phase-shift difference $|\arg(\mathbf{G}_{PFNCSTC}(i\omega^*)) - \arg(\mathbf{G}_{ADPFC}(i\omega^*))|$. Notice that the difference between the results for $N = 4$ and 16 is not resolved in this diagram. The plot is made for $k^*=20$. (Note that the phase-shift difference is expressed in cycles instead of radians.)



c. As **a.** but $k^* = 60$.



d. As **b.** but $k^* = 60$.

2.5 Temperature dependency and the choice of representative temperature

The heat treatment of liquid food is often accomplished by heating the food in a heat exchanger, holding the food at this temperature for a given time and cooling the food in another heat exchanger. In the first and last of these steps the temperature of the fluid changes as it flows along the channel.

The chemical reaction constant k depends on the temperature according to the well-known Arrhenius equation.

$$k(T) = k_0 e^{-\frac{E_a}{R_g T}} \quad (17)$$

See Eqs. (26) and (27) below for the conversion of the commonly used parameters for decimal reduction time, D_r , and decimal reduction temperature increase, z_r , to k_0 and E_a .

When calculating the effect of the chemical reactions, the most straight-forward method of increasing the accuracy to an appropriate level would be to increase the degree of discretization in the numerical computation, and use the mean value of the inlet and outlet temperatures in each control volume for the calculation of $k(T)$. To decrease the error for a given degree of discretization we approximated the temperature change through a control volume with a linear function. For a sample exposed to a time-dependent temperature, the change in concentration due to a first-order reaction follows the relationship in Eq. (18).

$$\frac{dC}{dt} = -Ck = -Ck_0 e^{-\frac{E_a}{R_g T(t)}} \quad (18)$$

Here Eq. (17) was used. A linear temperature change can be expressed as follows.

$$T(t) = T_{in} + \dot{T}t = T_{in} \left(1 + \frac{\dot{T}t}{T_{in}} \right) \quad (19)$$

Inserting this into Eq. (18) and evaluating yields Eq (20).

$$\ln \left(\frac{C_{out}}{C_{in}} \right) = \int_0^\tau -k_0 e^{-\frac{E_a}{R_g T_{in} \left(1 + \frac{\dot{T}t}{T_{in}} \right)}} dt \approx \int_0^\tau -k_0 e^{-\frac{E_a}{R_g T_{in}} \left(1 - \frac{\dot{T}t}{T_{in}} \right)} dt \quad (20)$$

The approximation on the right-hand side is motivated by the fact that

$|\dot{T}| \leq |\dot{T}\tau| = |\Delta T| \ll T_{in}$ in normal cases with sufficient discretization. Evaluating the right-hand side gives Eq. (21),

$$\ln \left(\frac{C_{out}}{C_{in}} \right) \approx -\tau k_0 e^{-\frac{E_a}{R_g T_{in}}} \frac{e^{\frac{E_a \dot{T} \tau}{R_g T_{in}^2}} - 1}{\frac{E_a \dot{T} \tau}{R_g T_{in}^2}} = -\tau k_0 e^{-\frac{E_a}{R_g T_{in}}} \frac{e^{\frac{E_a \Delta T}{R_g T_{in}^2}} - 1}{\frac{E_a \Delta T}{R_g T_{in}^2}} \quad (21)$$

where ΔT is the temperature change through the channel element. Here, the first part is identified as $\tau k(T_{in})$, and we can identify the factor

$$c_a = \frac{e^{\frac{E_a \Delta T}{R_g T_{in}^2}} - 1}{\frac{E_a \Delta T}{R_g T_{in}^2}} \quad (22)$$

as the correction factor for the linear change from the temperature T_{in} to the temperature $T_{in} + \Delta T$. It should be noted that the error in the approximation in Eq. (20)

is less, the greater the value of T_{in} . This means that the integration should be performed from the high temperature to the low temperature. Thus for $\dot{T} < 0$, Eq. (21) with the corresponding correction factor in Eq. (22) should be used and for $\dot{T} > 0$, the following approximation is better.

$$\ln\left(\frac{C_{out}}{C_{in}}\right) \approx -\tau k_0 e^{-\frac{E_a}{R_g T_{out}}} e^{\frac{E_a \dot{T} \tau}{R_g T_{out}^2} - 1} = -\tau k_0 e^{-\frac{E_a}{R_g T_{out}}} e^{\frac{E_a \Delta T}{R_g T_{out}^2} - 1} \quad (23)$$

Here, the first part is identified as $\tau k(T_{out})$ and we can identify the corresponding correction factor

$$c_b = \frac{e^{\frac{E_a \Delta T}{R_g T_{out}^2} - 1}}{e^{\frac{E_a \Delta T}{R_g T_{out}^2}}} \quad (24)$$

Note that in both cases the exponents and denominators are all negative (ΔT is negative in Eq. (22) and positive in Eq. (24)). The conclusion is that the general expression for the model is

$$\ln\left(\frac{C_{out}}{C_{in}}\right) = -\tau k(\max\{T_{out}, T_{in}\}) \frac{1 - e^{-\frac{E_a |\Delta T|}{R_g \max\{T_{out}, T_{in}\}^2}}}{\frac{E_a |\Delta T|}{R_g \max\{T_{out}, T_{in}\}^2}} \quad (24b)$$

where $\max\{T_{out}, T_{in}\}$ means the maximum value of T_{out} and T_{in} , and $k(\max\{T_{out}, T_{in}\})$ refers to Eq. (17).

Summarizing, to model efficiently chemical reaction in a one dimensional model, taking into account variable flow rate, variable material properties, variable temperature and dispersion, we propose discretization into N identical elements each consisting of a transport delay and an ideal mixer, which proportions are given by Eqs. 9a and 9b, the true transport delay evaluated according to Eqs. 16a and 16b, and the non-isothermal effect on the concentration change calculated with a correction given by Eq. 24b.

3. Model implementation and simulation

The new dispersion model for turbulent flow with first-order chemical reactions was implemented in the modelling language Modelica¹. Chemical reactions were added to the dispersion model for pipes developed by Skoglund & Dejmek (2007). Both models (dispersion and chemical reaction) were also implemented as extensions to the heat-exchanger model developed by Skoglund et al. (2006). Since microbial deactivation in liquid-food applications deals with very small quantities, the thermal effect of the chemical reactions (endothermic or exothermic) on the system is negligible, and was therefore ignored in the models. Two systems were simulated: (i) a system describing the steady state of the final heater, holding and first cooling in a typical milk sterilizer (see description in section 3.1 below), and (ii) a system describing the transient response of a chemical reaction in an isothermal holding tube (tube reactor) exposed to a step flow change.

¹ Modelica was the program language. The tool was Dymola supplied by Dynasim AB.

In the simulations, the dispersion coefficient, D , was calculated according to Wen & Fan (1975), p. 146:

$$\frac{D}{Lv} = \frac{1}{Pe} = \frac{2R}{L} \left(3.0 \cdot 10^7 Re^{-2.1} + 1.35 Re^{-0.125} \right) \quad (25)$$

3.1 Steady state

In the present study a complete system of component models was set up to simulate a production line with a first-order chemical reaction typical for liquid-food applications. The system is illustrated in Fig. 4a, and can be described as follows.

- A tubular heat exchanger (industrial type Tetra Pak MT 85/12x16C-6 with the heat exchanger area 20,6 m² and the actual overall heat transfer coefficient 2880 W/Km²) for the final heating, where preheated milk enters at 98 °C, and hot water enters in counter-current flow at 139.5 °C.
- A 5.5 m long pipe with an inner diameter of 48.6 mm (standard type with an outer diameter of 51 mm) including 2 bends (90°) used as a holding tube after the final heater.
- A heat exchanger (the same type as for heating but with the heat exchanger area 13,7 m² and overall heat transfer coefficient 2740 W/Km²) for the initial cooling after the holding tube where the cooling water enters in counter-current flow at 89 °C.
- A milk flow rate of 7150 kg/h.
- A water flow rate on the heating- and cooling-medium side of the heat exchangers of 7300 kg/h.

The chemical reaction constants used were those for a typical food-spoiling microorganism. The commonly used parameters are the following.

D_r = the time required to reduce the microbial concentration by one decimal (i.e. a factor 10) at a reference temperature T_r

z_r = the temperature increase required from the reference temperature T_r to increase the reaction rate by a factor of 10.

In this study typical values for the spores of the microorganism *Clostridium botulinum* were used: Walstra, Geurts, Noomen, Jellema & Boekel (1999), Table 6.4, and Burton (1988), Table 2.2: $D_r = 12$ s and $z_r = 10$ °C, both at the reference temperature $T_r = 121.1$ °C. Since the proposed model is based on the chemical reaction constants k_0 and E_a , the following conversion was performed.

$$E_a = \ln(10) R_g T_r^2 \left(\frac{1}{z_r} + \frac{1}{T_r} \right) \quad (26)$$

$$k_0 = \frac{\ln(10) e^{\frac{E_a}{R_g T_r}}}{D_r} \quad (27)$$

With the values given above this yields $E_a = 305.1$ kJ/mol and $k_0 = 5.105 \cdot 10^{39}$ s⁻¹.

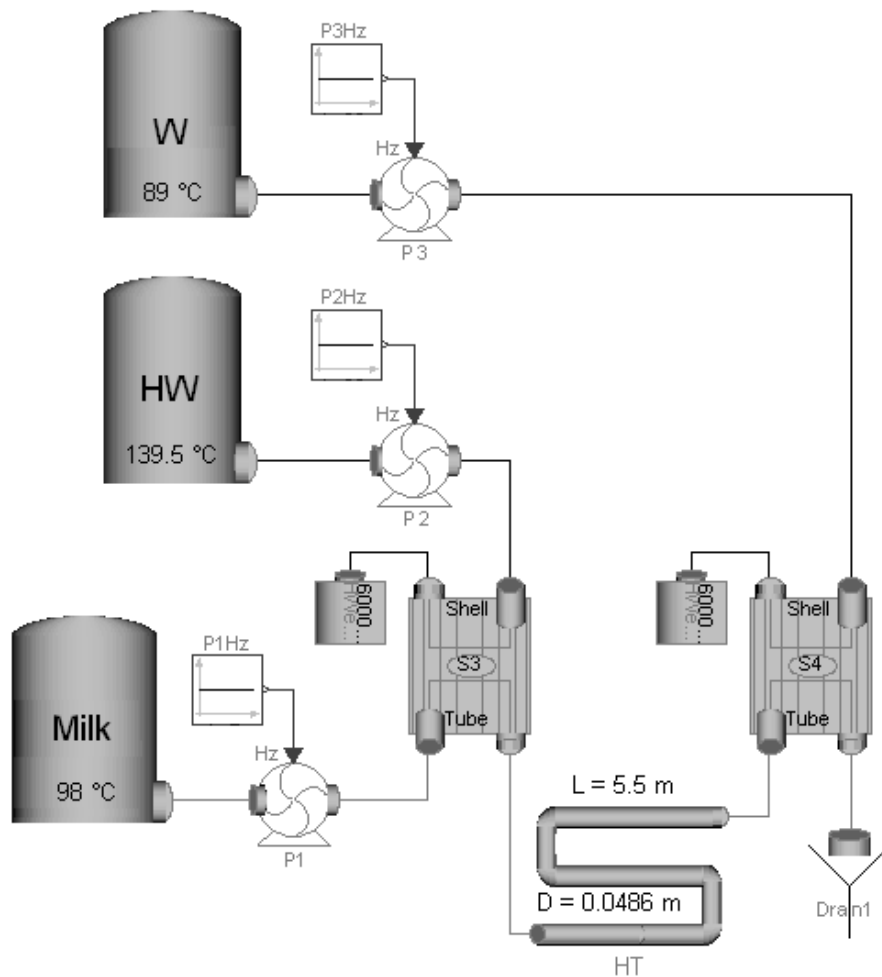


Fig. 4a. The design of a theoretical experiment as a system of dynamic models. The system consists of the final heating (S3), holding (HT) and first cooling (S4) in a typical industrial milk sterilizer.

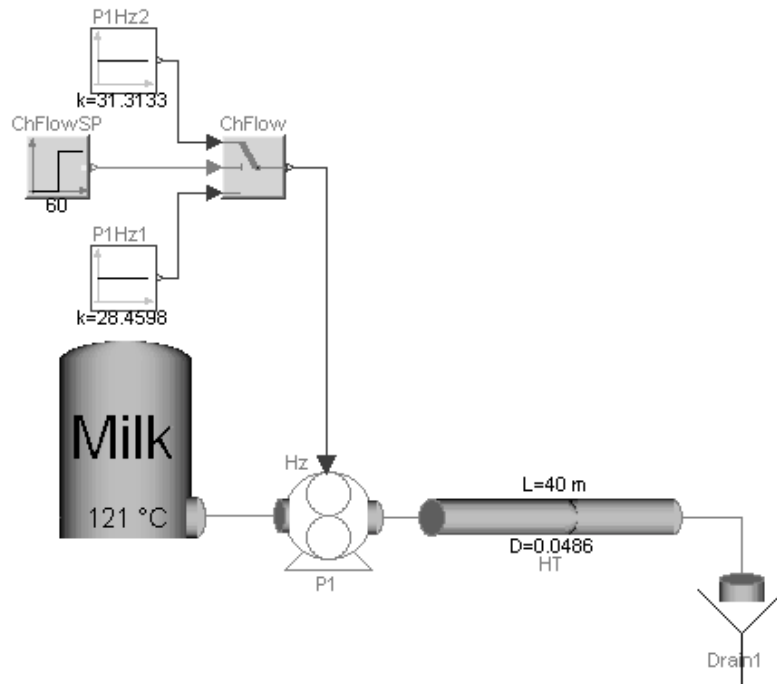


Fig. 4b. The design of a theoretical experiment as a system of dynamic models. The system consists of a holding tube through which the flow is exposed to a step change in volumetric flow rate.

3.2 Flow transients

To demonstrate the ability of the model to account for “true” transport delays according to Eq. (16a) and thus flow variations, the system illustrated by Fig. 4b was set up.

- A source of milk at 121 °C
- A 40 m long pipe with an inner diameter of 48.6 mm (standard type with an outer diameter of 51 mm).
- A milk flow rate of 7150 kg/h that increases by 10 % after 60 s

The same reaction constants (E_a and k_0) were used as in the steady-state case above.

4. Results

Simulations¹ were carried out by numerically solving the system of model equations using the Dassl solver in Dynasim’s Modelica-based program, Dymola, version, 5.3d.

4.1 Start-up and steady state

The following simulations were run.

- 1) The system was simulated according to Fig. 4a with only the heater included. In order to ensure that stationary conditions ($s^* = 0$) had been reached, the simulation was run for 3600 s in the model. The pure plug flow model was used, i.e. Eq. (15) with $D = 0$ ($Pe = \infty \Rightarrow \tau_N = 0$ according to Eq. (9b)) to study just the non-isothermal condition without any dispersion. Thus the transfer function (as $Pe \rightarrow \infty$) is $\left[\mathbf{G}_{PFNCST^c}(0) \right]^{1/N} = e^{-\frac{\tau_0 k}{N}}$ as expected.
 - a) Simulation by straightforward handling of the varying temperature in the heater, using the mean value of the temperatures in the inlet and outlet of each control volume.
 - b) Simulation by the proposed handling of the varying temperature in the heater according to Eq. (24b).
- 2) The whole system according to Fig. 4a was simulated. In order to ensure that stationary conditions had been reached, the simulation was run for 3600 s in the model.
 - a) With plug flow as above and simulation of the varying temperature in the heater according to Eq. (24b) (or Eqs. (21) and (23)). In other words simulation with the same proposed models as in 1b).
 - b) With dispersion according to Eq. (15) and simulation of the varying temperature in the heater according to Eq. (24b). In other words simulation with the same simulation as in 2a) but with dispersion instead of plug flow.

The reduction of microorganisms (*Clostridium botulinum* spores) according to simulations 1a and 1b above (stationary) is shown in Fig. 5. The required computational power, in terms of CPU² time, is also plotted. Fig. 5 clearly shows that the proposed model (simulation 1b) obviously converges faster than the straightforward method of handling the non-constant temperature (explained above about simulation 1a). The combined plot shows that, for a given level of accuracy, the straightforward model requires approximately 10 times more CPU time than the proposed model.

¹ A standard PC was used: Dell Optiplex SX270 with Intel® Pentium® 4 CPU 3.2 GHz, 1.0 GB RAM.

² Central Processing Unit

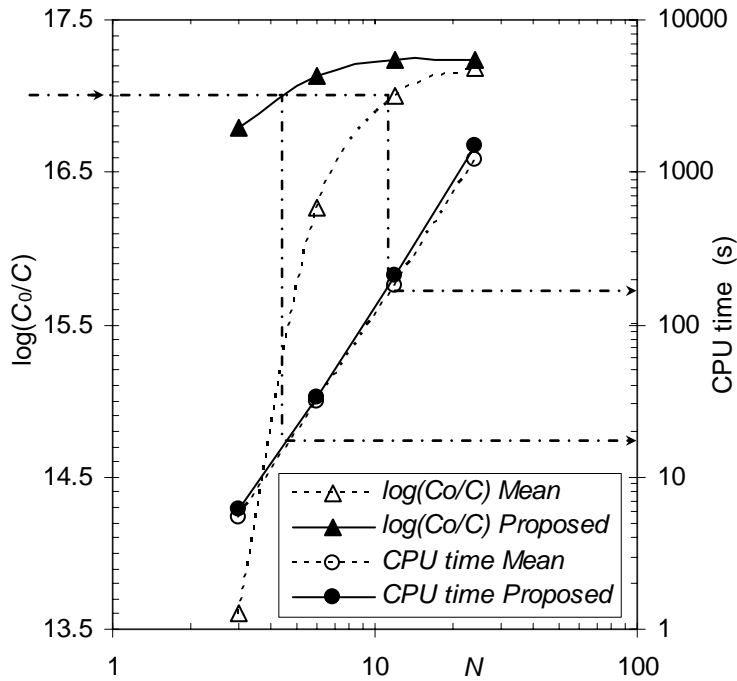


Fig. 5. The calculation accuracy and CPU time as a function of model discretization. The logarithmic reduction of microorganisms, $\log(C_0/C)$ (or other substance) by first-order reaction in a subsystem of the system in Fig. 4a. where only the heater (S3) was included. Note that C is the concentration at the exit of the heater. The plot also shows the required CPU time, i.e. computational power. The proposed way of handling the temperature change is shown as well as the straightforward, i.e. using the mean value of the temperatures in the inlet and outlet of each control volume. It is clearly visible that the proposed model requires less CPU time for the same accuracy than the straightforward model requires. The dash-dotted lines indicates a chosen level of accuracy that requires approx. 190 s of CPU time for the straightforward model but only 19 s of CPU time for the proposed model.

The reduction of microorganisms according to simulations 2a and 2b above are shown in Fig. 6 as a function of the axial coordinate through the system. The temperature is plotted in the same graph. For comparison, the result from a commonly applied industrial procedure for calculating the reduction in the holding tube is included in the graph. In that procedure the holding time for the fastest particle – the effective holding time – is used to calculate the reduction according to Eq. (18), i.e. $\ln(C_{out}/C_{in}) = -\tau_e k$, where τ_e is the effective holding time, i.e. the average holding time τ , multiplied by an efficiency factor given by Kessler (2002), p. 15. The commonly applied industrial procedure obviously underestimates the thermal effect. However, the plug-flow calculation leads to overestimation.

A commonly accepted industrial interpretation of standards and legislation (FAO/WHO Codex Alimentarius; FDA and Centre for Food Safety and Applied Nutrition (2002)) is that the reduction of *Clostridium botulinum* spores should reach at least $\log(C_0/C) = 12$ in food sterilization processes. It is also a well-known fact that the contribution to the deactivation of the microorganisms is great during the heating

and cooling in sterilizers based on indirect heating by means of heat exchangers. Fig. 6 confirms this.

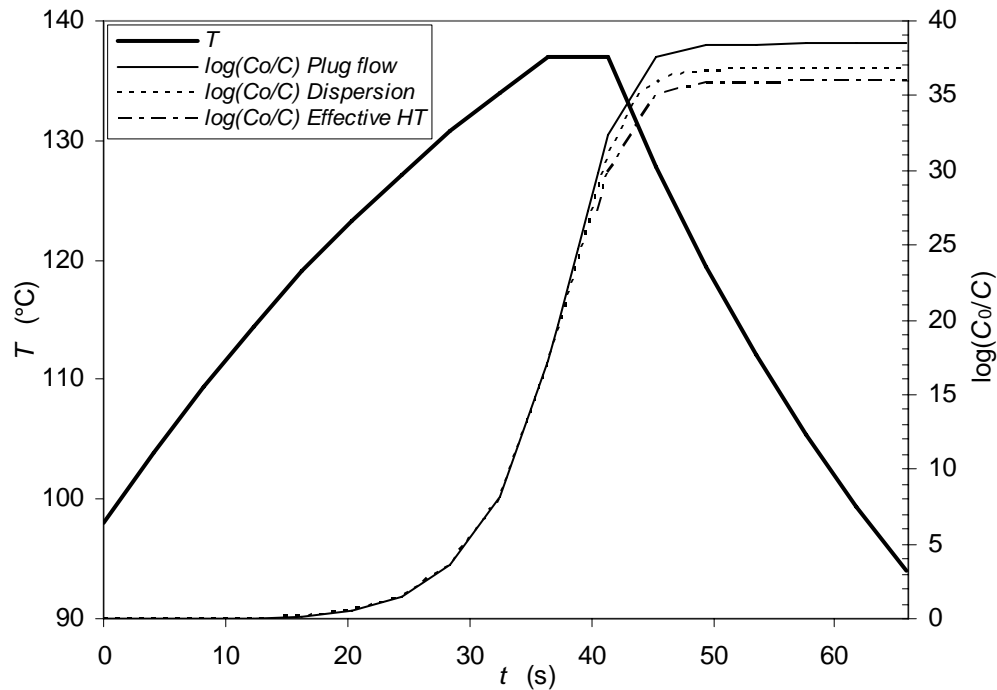


Fig. 6. The reduction of microorganisms (*Clostridium botulinum* spores) along the axial heat-exchanger coordinate expressed in terms of time. The corresponding temperature profile is also shown. The plot shows calculation results based on plug flow (simulation 2a), dispersed flow (simulation 2b) and dispersed flow but with a holding tube with plug flow and effective (reduced) holding time by an efficiency factor given by Kessler (2002), p. 15 (commonly applied industrial calculation procedure). Note that C is the concentration at a position in the system corresponding to the time it takes for the fluid to flow from the inlet to the position.

4.2 Flow transients

The system according to Fig. 4b was simulated for 120 s with both pure plug flow and with dispersion included. In both of these cases simulations were done with

1. The instantaneous holding time, i.e. $\tau = L/v$ (straightforward method).
2. The proposed “true” holding time according to Eqs. (16a) and (16b).

The results of the simulations are shown in Figs. 7a and 7b which clearly show that the instantaneous transport delay model (see point 1 above) performs poorly whereas the proposed “true” transport delay model shows good performance. In fact the “true” transport delay model exhibits the same results with no further discretization, i.e. with $N = 1$, as can be analytically derived, which means a linear change in $\log(C_0/C)$ from the value corresponding to the initial flow rate to the value corresponding to the increased flow rate initiated at 60 s. The performance of the instantaneous transport delay model depends greatly on the degree of discretization, requiring large values of N , i.e. considerable computation (CPU time), for reasonable accuracy. Fig. 7b shows that the instantaneous model requires more CPU time by a factor of over 100 to achieve a result with accuracy close to the result of the proposed model.

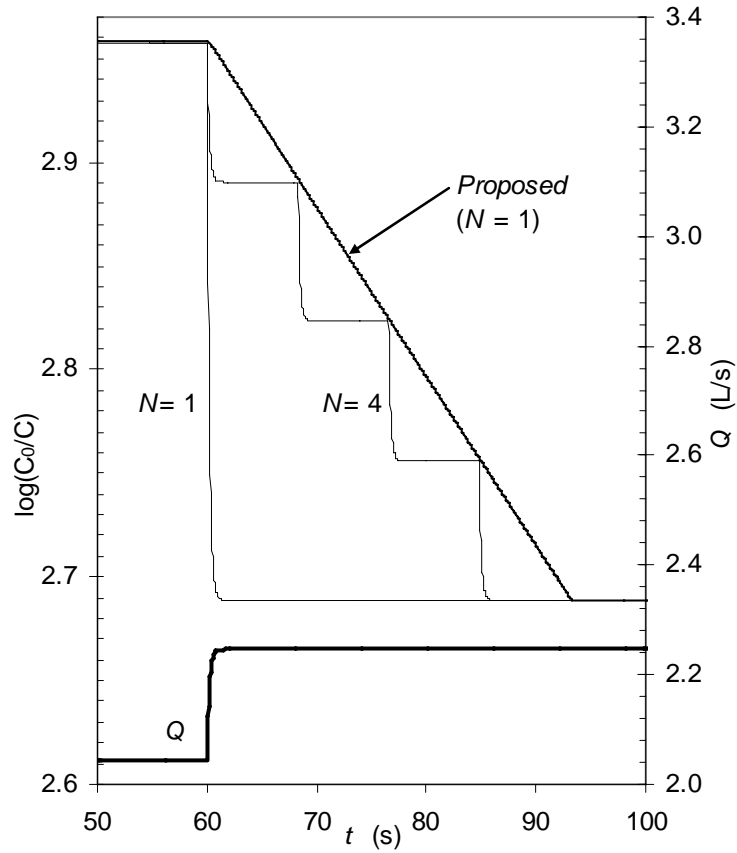


Fig. 7a. Simulation result of the system according to Fig. 4b with *plug flow model* using both the method of calculating the instantaneous holding time, $\tau = L/v$, (just marked with degree of discretization) and the proposed method of calculating the “true” holding time according to Eqs. (16a) and (16b). The “true” holding time yields the same result independently of discretization (N). The 10 % step change of the flow rate is shown at the bottom of the graph. Note that $C = C(L, t)$.

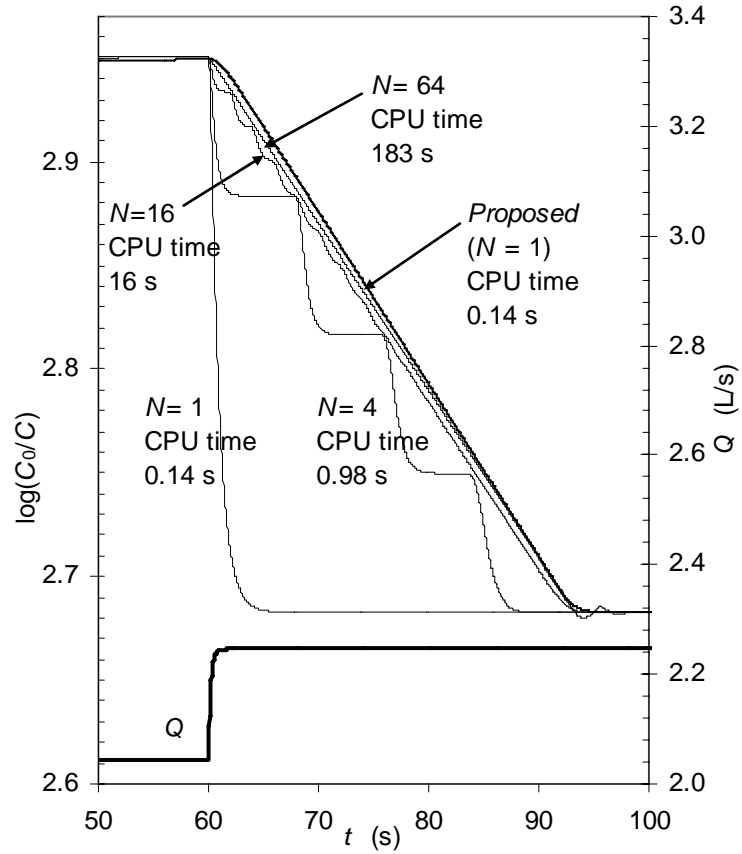


Fig. 7b. As Fig. 7a but with *dispersion model* instead of plug flow model, and two more simulations ($N = 16$ and $N = 64$) for the instantaneous method (see caption of Fig. 7a). During the simulations, the dynamically calculated dispersion coefficient, D , according Eq. (25), was $0.0159 \text{ m}^2/\text{s}$ at the low flow rate and $0.0173 \text{ m}^2/\text{s}$ at the high flow rate. The need for computational power in terms of required processor time (CPU time) is shown in the graph.

5. Summary

In order to account for reduction of microorganisms and nutrients in liquid-food production lines, a novel model was formulated for dynamic simulation of first-order chemical reaction in channels where the flow can be described as axially dispersed (Eqs. (14) and (15)), which is often the case when processing liquid food. The model was based on a previously demonstrated computationally efficient model. The model was implemented in tube models and tubular heat-exchanger models. A method of efficiently accounting for non-isothermal conditions was also implemented in the heat-exchanger models (Eq. (24b)). Simulations showed that the novel method performed better than the straightforward method, based on mean temperatures.

Furthermore, a novel method of including a “true” dynamic transport delay (Eqs. (16a) and (16b)) was included in the models to account for varying flow rates. Simulations showed that the “true” transport delay method performed better as regards computation time than the straightforward method of calculating the instantaneous transport delay $\tau = L/v$.

6. Conclusions

The proposed formulations of the dynamic model of dispersed uniaxial flow with temperature change and first-order chemical reactions cover a large part of the needs for modelling of complex processing lines in the food industry, including the key issues of microorganism growth and elimination, and the loss of nutrients and formation of heat-induced compounds. In comparison to the naïve implementation of the standard calculations, the model provides good accuracy with reduced computational requirements.

Acknowledgements

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Appendix

Derivation of the transfer function, Eq. (3)

The starting point is the following PDE for a semi-infinite pipe.

$$\frac{\partial C(x,t)}{\partial t} + v \frac{\partial C(x,t)}{\partial x} - D \frac{\partial^2 C(x,t)}{\partial x^2} + kC(x,t) = 0 \quad (\text{A1})$$

We introduce the Laplace transform

$$\mathbf{C}(x,s) = \int_0^\infty e^{-st} C(x,t) dt \quad (\text{A2})$$

Then we find the Laplace transformation of Eq. (A1)

$$s\mathbf{C}(x,s) - C(x,0) + v \frac{\partial \mathbf{C}(x,s)}{\partial x} - D \frac{\partial^2 \mathbf{C}(x,s)}{\partial x^2} + k\mathbf{C}(x,s) = 0 \quad (\text{A3})$$

Where

$$\mathbf{C}(0,s) = \mathbf{C}_{in}(s)$$

and $C(x,0)$ is the initial condition, i.e. the concentration distribution in the pipe at time $t = 0$. Now assume that the initial condition is zero, this gives

$$(k+s)\mathbf{C} + v \frac{\partial \mathbf{C}}{\partial x} - D \frac{\partial^2 \mathbf{C}}{\partial x^2} = 0 \quad \text{or} \quad \frac{\partial^2 \mathbf{C}}{\partial x^2} + \frac{v}{D} \frac{\partial \mathbf{C}}{\partial x} - \frac{k+s}{D} \mathbf{C} = 0 \quad (\text{A4})$$

Thus the general solution to the differential equation Eq. (A4) is:

$$\mathbf{C}(x,s) = e^{\frac{vx}{2D}} \left(c_1 e^{\frac{vx}{2D} \sqrt{1 + \frac{4D}{v^2}(k+s)}} + c_2 e^{-\frac{vx}{2D} \sqrt{1 + \frac{4D}{v^2}(k+s)}} \right)$$

The solution must be finite when $x \rightarrow \infty$, which implies that $c_1 = 0$. Implicitly this means that we study a semi-infinite long pipe with no other boundary than $x = 0$. For $x = 0$ we have the inlet concentration (Dirichlet type of boundary condition)

$$\mathbf{C}_{in}(s) = \mathbf{C}(0,s) = c_2$$

$$\text{Thus } \mathbf{C}(x,s) = \mathbf{C}_{in}(s) e^{\frac{vx}{2D}} e^{-\frac{vx}{2D} \sqrt{1 + \frac{4D}{v^2}(k+s)}} \quad (\text{A6})$$

But the outlet concentration is the concentration for $x = L$, which means

$$\mathbf{C}_{out}(s) = \mathbf{C}_{in}(s) e^{\frac{vL}{2D}} e^{-\frac{vL}{2D} \sqrt{1 + \frac{4D}{v^2}(k+s)}} \quad (\text{A7})$$

Thus the transfer function is:

$$\mathbf{G}(s) = \frac{\mathbf{C}_{out}(s)}{\mathbf{C}_{in}(s)} = e^{\frac{vL}{2D}} e^{-\frac{vL}{2D} \sqrt{1 + \frac{4D}{v^2}(k+s)}} = \mathbf{G}_{ADPF^c}(s) \quad (\text{A8})$$

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