OPTIMAL CONTROL OF DISPERSIVE TUBULAR CHEMICAL REACTORS: PART II

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Abstract: While in the first paper of this series of two, optimal jacket fluid temperature control profiles have been derived for a family of dispersive tubular chemical reactors, the performance of these optimal profiles is further assessed in this second part. First, the performance of the optimal bang-bang control is compared with that of a more easily implementable constant jacket fluid temperature and secondly, the transient behaviour is studied, when the steady-state optimal control law is applied from the reactor start-up.

Keywords: chemical industry, distributed-parameter systems, model-based control, optimal control, reactor control, second-order systems

1. INTRODUCTION

As already mentioned in part I of this series (Logist *et al.*, 2005), the problem of optimal control of (nonlinear) chemical processes remains an intriguing and persistent challenge for chemical process control engineers. The reactor under study in this series of papers is a classical dispersive tubular reactor in which an irreversible, exothermic, first-order reaction takes place. A surrounding heating/cooling jacket is present to control the reactor as to maximise the conversion and to minimise the energy cost.

In previous works (Logist *et al.*, 2004; Logist *et al.*, 2005), the optimal jacket fluid temperature profile has been derived analytically by applying Pontryagin's minimum principle, yielding a bang-bang control law. Based on physical knowledge a maximum-minimum (max-min) control has been adopted and the position for switching from the maximum to the minimum jacket fluid temperature has been numerically optimised by means of a weighted shooting-type procedure. The optimisation has been performed for a family of tubular reactors, ranging from reactors with a CSTR-like behaviour to nearly plug flow reactors (PFR).

This second paper first illustrates the improved performance of the max-min control law over a more easily implementable constant jacket fluid temperature control. Secondly, the reactor performance under transient conditions is assessed, i.e., when the optimised steady-state control is applied from the start-up of the empty reactor.

However, due to the distributed nature and the inherent nonlinearities of the transient reactor system, transient simulations require the solution of a set of two coupled partial differential equations (PDEs). Additionally, the dispersion parameters, or equivalently the Peclet numbers, strongly influence the nature of these equations and thereby also the numerical procedures to solve them.

In this contribution, it is illustrated how a combination of numerical methods is required for investigating the whole reactor type range.

The organisation of the paper is as follows. Section 2 introduces the dynamic mathematical model. Section 3 compares the reactor performances under the analytically derived optimal bang-bang control and the more easily implementable constant jacket fluid temperature control law. Section 4 highlights the numerical techniques needed for accurate transient simulations, which are applied in Section 5. In this latter section the transient reactor behaviour is discussed. Finally, Section 6 summarises the main conclusions.

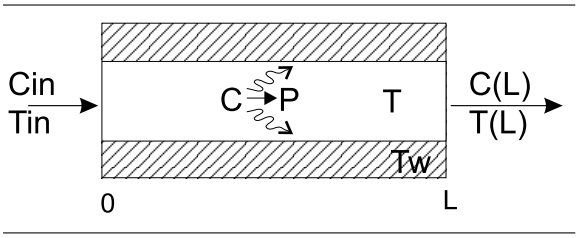

Fig. 1. Tubular reactor with surrounding heating/cooling jacket.

2. THE DYNAMIC AXIAL DISPERSION MODEL

Describing the reactor under transient conditions by a 1D-model with axial mass and heat dispersion results in the following system of two second-order partial differential equations with respect to the spatial coordinate z and the time t,

$$\frac{\partial T}{\partial t} = D_1 \frac{\partial^2 T}{\partial z^2} - v\frac{\partial T}{\partial z} - \frac{\Delta H}{\rho C_p} k_0 C e^{\frac{-E}{RT}} + \frac{4h}{\rho C_p d}(T_w - T) \quad (1)$$

$$\frac{\partial C}{\partial t} = D_2 \frac{\partial^2 C}{\partial z^2} - v\frac{\partial C}{\partial z} - k_0 C e^{\frac{-E}{RT}} \quad (2)$$

subject to four Danckwerts boundary conditions (Danckwerts, 1953),

$$D_1 \frac{\partial T(0,t)}{\partial z} = v(T(0,t) - T_{in}); \ \frac{\partial T(L,t)}{\partial z} = 0$$
$$D_2 \frac{\partial C(0,t)}{\partial z} = v(C(0,t) - C_{in}); \ \frac{\partial C(L,t)}{\partial z} = 0$$

and two initial conditions reflecting the empty state at start-up,

$$T(z,0) = T_0(z) = T_{in} = 340 \text{ K}$$
$$C(z,0) = C_0(z) = 0 \text{ mole/L}$$

where all variables have the same meaning as in the first paper.

3. ASSESSMENT OF OPTIMALITY

In this section, the steady-state reactor performance under two different controls, i.e., the analytically derived optimal bang-bang control, and a more easily implementable constant jacket fluid temperature control, is numerically investigated.

Under steady-state conditions the dynamic partial differential equations (1) and (2) and their respective boundary conditions reduce to the following set of ordinary differential equations,

$$D_1 \frac{d^2 T}{dz^2} - v\frac{dT}{dz} - \frac{\Delta H}{\rho C_p} k_0 C e^{\frac{-E}{RT}} + \frac{4h}{\rho C_p d}(T_w - T) = 0$$
$$D_2 \frac{d^2 C}{dz^2} - v\frac{dC}{dz} - k_0 C e^{\frac{-E}{RT}} = 0$$

subject to the following boundary conditions

$$D_1 \frac{\partial T(0)}{\partial z} = v(T(0) - T_{in}); \ \frac{\partial T(L)}{\partial z} = 0$$
$$D_2 \frac{\partial C(0)}{\partial z} = v(C(0) - C_{in}); \ \frac{\partial C(L)}{\partial z} = 0$$

Once again, dimensionless variables are introduced for numerical convenience (see Part I).

The calculations are performed for tubular reactors ranging from nearly plug flow reactors ($Pe = 10^8$) to almost perfectly mixed continuous stirred tank reactors ($Pe = 0.01$), using the weighted shooting-type procedure described in Part I. It should be noted here, that for these simulations the same fixed parameter values and the same terminal cost criterion (Equation (3)), being a trade-off between conversion and energy consumption, are used as in the previous paper.

$$\mathcal{J}[u] = (1-A)\underbrace{C(L)}_{\mathcal{J}_1[u]} + A\underbrace{\frac{(T(L)-T_{in})^2}{K_1}}_{\mathcal{J}_2[u]} \quad (3)$$

Since a constant jacket fluid temperature is easier to implement than the max-min step profile (only one heat exchanger, operating at a constant, intermediate temperature T_w is needed instead of two), the performance of such a constant profile is compared with that of the optimal one. For 121 equally distributed jacket fluid temperatures T_w ranging from $T_{w,min}$ (280 K) to $T_{w,max}$ (400 K) the exact inlet conditions are calculated and used to compute the value of the cost criterion $\mathcal{J}$ (Equation (3)). Figure 2 illustrates the evolution of the minimal total cost and of the optimal constant jacket fluid temperature as a function of the Peclet number Pe.

The following conclusions can be deduced from this figure.

- For high Peclet numbers the optimal jacket fluid temperature is equal to the plug flow

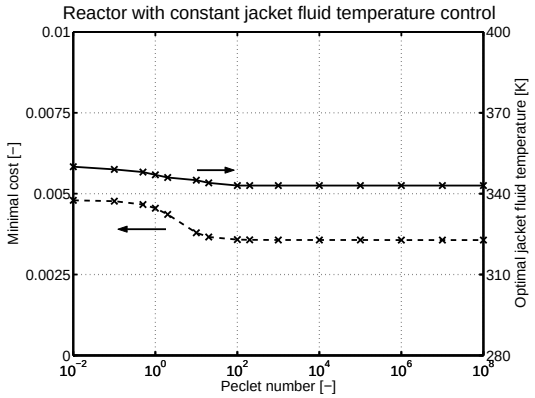

Fig. 2. Minimal cost value (- - -) and optimal constant jacket fluid temperature (—) as a function of the Peclet number Pe.

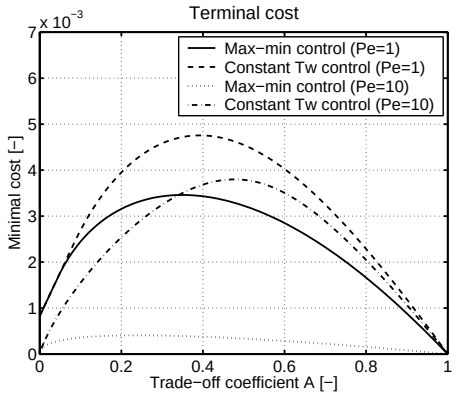

Fig. 3. Minimal cost value as a function of the trade-off coefficient A for $Pe = 1$ and $Pe = 10$.

value of 343 K, while for low Peclet numbers the optimal jacket fluid temperature slightly increases to the CSTR value.

- A similar transition in the minimal cost value as in the first paper is observed here. However, the decrease in cost is not as sharp as before. For all Peclet numbers this more easily implementable constant jacket fluid temperature control results in a higher cost, which illustrates the optimality of the analytically derived control law. However, this effect is less pronounced for highly dispersive tubular reactors. In practice, the control engineer will have to decide whether the improved performance of the max-min control is worth installing a second heat exchanger.

The same effect is also observed in Figure 3, where the optimal cost values of both controls are depicted at intermediate Peclet numbers of 1 and 10, for A varying from 0 to 1. Clearly, for all A values, the analytically derived bang-bang control has a better, or at least an equal performance as the constant jacket fluid temperature control.

4. TRANSIENT SIMULATION TECHNIQUES

In this section, some technical details are given about the employed transient simulation methods.

In literature, an enormous number of numerical methods for various types of partial differential equation problems has been published over the past decades, e.g., finite differences methods, finite elements methods and collocation methods (see, e.g., Hundsdorfer and Verwer (2003)).

Recently, a promising new routine, called *sequencing method*, belonging to the class of operator splitting methods, has been published. The rationale of this method is the separation of the different physical and chemical reactor phenomena (i.e., dispersion, reaction and convection) (Renou *et al.*, 2003).

Another novel evolution is the release of a Matlab-based *method of lines* toolbox MatMOL, which provides the user a variety of easily understood methods and examples which can be employed for the rapid prototyping of new dynamic simulation codes (Vande Wouwer *et al.*, 2004).

Since the nature of the governing partial differential equations is heavily affected by the dispersion parameters, a combination of numerical methods is required for investigating the whole reactor range. In this paper, a variant of the sequencing method is used for reactors with limited dispersive behaviour, while methods from the MatMOL toolbox are exploited for more dispersive reactors.

The *method of lines* solution of PDEs is based on two steps: the spatial derivatives are first approximated by, e.g., finite differences, and then the system of semi-discrete equations is integrated in time. The success of this approach is due to the availability of efficient time integrators for solving the resulting mixed systems of (ordinary) differential and algebraic equations (DAEs) (Vande Wouwer *et al.*, 2004).

In the MatMOL toolbox, various discretisation stencils (uniform/nonuniform, different order and positioning) are available for approximating the spatial derivatives in the PDEs and the boundary conditions. Low-order schemes, e.g., first-order approximations, which lead to the classic *tanks-in-series* reactor model, always yield smooth solutions. However, the resulting profiles are not always accurate, especially for high Peclet values steep gradients are smeared due to the presence of so-called *numerical diffusion*. The application of high-order schemes, e.g., fifth-order approximations, results in more accurate transient profiles up to much higher Peclet values, but for very high Peclet numbers, they give rise to nonphysical oscillations in front of and/or behind steep gradients due to so-called *numerical dispersion*. Observe

Table 1. Optimised results for the max-min profile.

Pe [-]	z_1^* [m]	$\mathcal{J}^*$ [-]
0.01	0.58	$4.7783 \cdot 10^{-3}$
0.1	0.58	$4.6185 \cdot 10^{-3}$
0.5	0.58	$3.9488 \cdot 10^{-3}$
1	0.58	$3.2274 \cdot 10^{-3}$
2	0.57	$2.1795 \cdot 10^{-3}$
10	0.54	$3.3644 \cdot 10^{-4}$
20	0.54	$1.2252 \cdot 10^{-4}$
100	0.54	$1.8565 \cdot 10^{-5}$
200	0.54	$1.1845 \cdot 10^{-5}$
1000	0.54	$7.6388 \cdot 10^{-6}$
10^4	0.54	$6.7872 \cdot 10^{-6}$
10^5	0.54	$6.7638 \cdot 10^{-6}$
10^6	0.54	$6.7566 \cdot 10^{-6}$
10^7	0.54	$6.7555 \cdot 10^{-6}$
10^8	0.54	$6.7555 \cdot 10^{-6}$

that both numerical diffusion and dispersion are computational artefacts and, by consequence, may not be confused with their physical equivalents. Increasing the grid density, decreases these undesired effects, but also increases the computational burden. Hence, an acceptable trade-off between computation time and accuracy has to be found.

For low and intermediate Peclet values ($Pe \leq 10^4$), a *five point biased upwind* and a *five point centred* scheme are selected from the MatMOL toolbox to approximate the first-order and the second-order spatial derivatives, respectively, on a uniform grid with 501 points. The Danckwerts boundary conditions are taken into account at the boundary points. The resulting system of DAEs is then integrated in time with the `Matlab`© (The Mathworks Inc., Natick) routine `ode15s`.

The sequencing method is based on the separation of the different phenomena. At each time step, convection, dispersion and reaction (i.e., the chemical reaction part itself as also the exchange through the reactor wall) are applied successively on the reactor mesh. For weakly dispersive reactors, this method exhibits excellent shock capturing features, while at the same time for highly dispersive reactors also accurate simulations are obtained. The only minor drawback in the latter case is the fact that this method has some problems to capture the inlet gradient correctly. Again this effect can be decreased by increasing the number of grid points. Unfortunately, the computational expense also increases (Renou *et al.*, 2003).

For very high Peclet values ($Pe > 10^4$), an adapted version of the sequencing method over a uniform grid with 500 grid points has been elaborated in `Matlab`©.

5. TRANSIENT SIMULATIONS

In this section, the start-up process is simulated for the reactors with the previously optimised

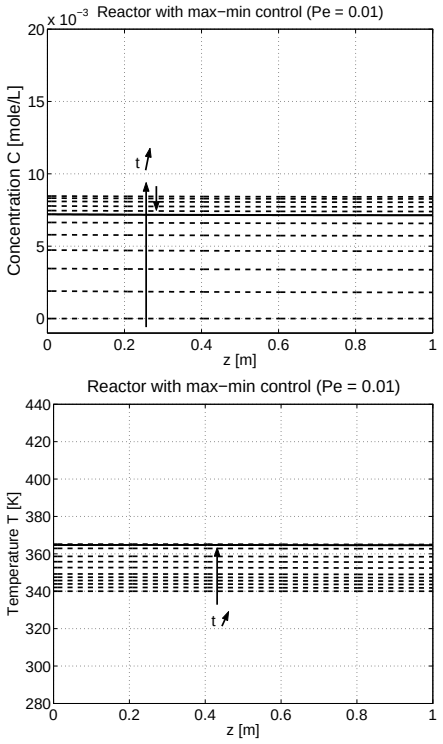

Fig. 4. Concentration (top) and temperature (bottom) profiles for $Pe = 0.01$ under the optimised max-min control ($z_1^* = 0.58$ m): transient evolution (- - -), steady-state from the transient regime (—) and from the weighted shooting-type technique (—).

bang-bang control. The transient evolution of the temperature and the concentration profiles are computed for the same 15 Peclet values as in the first paper (i.e., 10^8, 10^7, 10^6, 10^5, 10^4, 10^3, 200, 100, 20, 10, 2, 1, 0.5, 0.1 and 0.01). The optimised switching positions z_1^* and the minimal cost values $\mathcal{J}^*$ for the equally weighted terminal cost criterion are summarised in Table 1. Due to space limitations the transient results for only four more or less characteristic Peclet values (i.e., 0.01, 1, 10 and 10^8) are displayed in Figures 4, 5, 6 and 7. For each Peclet number the concentration (top) and temperature (bottom) profiles at 0, 1, 2, 3, 4, 5, 7, 9, 11, 15, and 20 s (dashed lines) are shown together with the steady-state profile *(i)* found as a result of the transient regime (i.e., profiles at the final simulated time of 50 s) and *(ii)* obtained with the weighted shooting-type (solid lines).

From Figures 4, 5, 6 and 7 the following conclusions can be drawn about the transient behaviour.

- A clear transition is visible in the four figures. Figure 4 displays very flat reactor profiles, close to the perfectly mixed behaviour of a CSTR. The profiles in Figures 5 and 6 are still smooth, yet, larger gradients in both the concentration and the temperature profile are visible due to the decreasing dispersion degree. And finally, in Figure 7 extremely

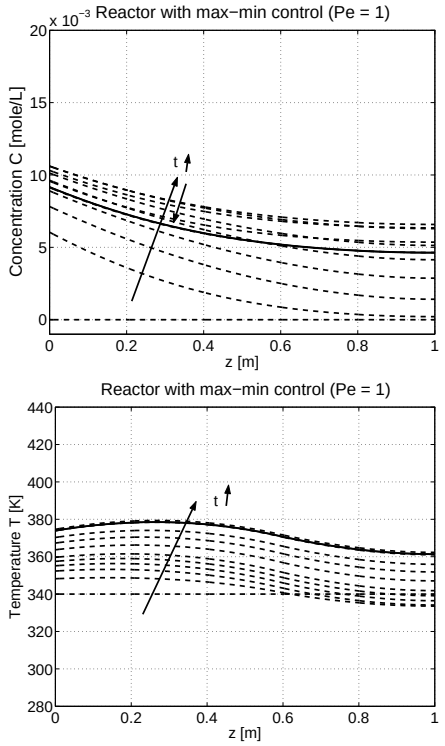

Fig. 5. Concentration (top) and temperature (bottom) profiles for $Pe = 1$ under the optimised max-min control ($z_1^* = 0.58$ m): transient evolution (- - -), steady-state from the transient regime (—) and from the weighted shooting-type technique (—).

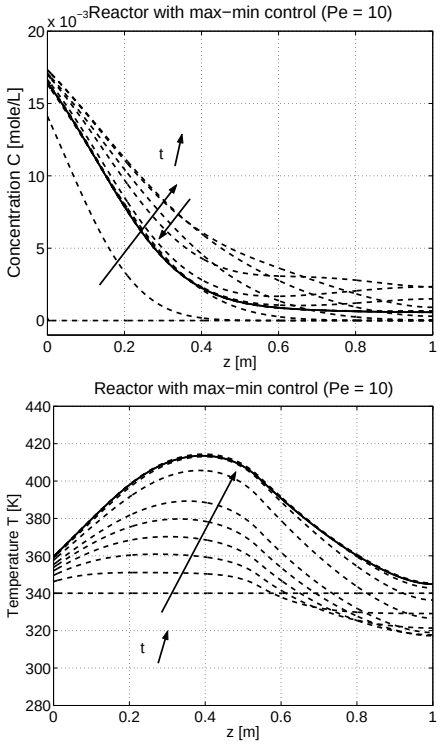

Fig. 6. Concentration (top) and temperature (bottom) profiles for $Pe = 10$ under the optimised max-min control ($z_1^* = 0.54$ m): transient evolution (- - -), steady-state from the transient regime (—) and from the weighted shooting-type technique (—).

sharp fronts are observed, because the fluid flows as a plug through the reactor.

- Convergence towards the steady-state is always achieved. This can be explained because in this study neither of the limit cases (plug flow reactor and CSTR) exhibits multiple solutions: a PFR never yields multiple solutions and the criterion for a CSTR with surrounding cooling jacket and irreversible first-order reaction ensures for this case a unique solution (Morbidelli *et al.*, 1987).
- The accuracy of the transient simulations is satisfactory. The obtained steady-state profile hardly differs from the one found with the weighted shooting-type technique: the two solid lines are undistinguishable.
- During the transient phase, no physical or constructive constraints are violated, i.e., the concentration and the reactor temperature always remain positive and the temperature inside the reactor does not reach unacceptably high levels, which is a very important conclusion from a practical point of view.

Finally two of the methods mentioned in Part I of this series of two papers for solving the steady-state boundary value problem, i.e., the *weighted shooting-type procedure* used in Part I and the *(false) transient method*, which is *de facto* implemented in this paper, can be compared based on their computational efficiency. Although the resulting steady-state profiles are the same, the computational burden differs. In Table 2 the computation times (on a 2.4 GHz PC with 512 Mb RAM) for the steady-state profiles displayed in Figures 4 to 7 are indicated. As can be seen, the former method is generally faster (although the initial guess for the optimisation routine influences the calculation time). However the latter technique has the advantage of yielding an *all-in-one* (i.e., steady-state and transient) solution.

Table 2. Computation time for steady-state profiles (^{a}: MatMOL toolbox, ^{b}: sequencing method).

	Weighted shooting-type	False transient
$Pe = 0.01$	2.9 s	29.4^a s
$Pe = 1$	15.7 s	35.7^a s
$Pe = 10$	1.9 s	38.6^a s
$Pe = 10^8$	25.1 s	391.1^b s

6. CONCLUSIONS

In the context of optimal control of chemical reactors, this series of two papers focusses on the optimal jacket fluid temperature profiles for exothermic tubular chemical reactors with axial mass and heat dispersion.

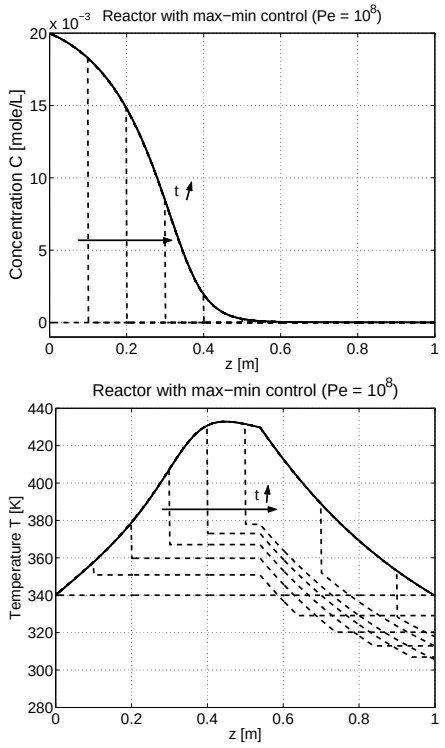

Fig. 7. Concentration (top) and temperature (bottom) profiles for $Pe = 10^8$ under the optimised max-min control ($z_1^* = 0.54$ m): transient evolution (- - -), steady-state from the transient regime (—) and from the weighted shooting-type technique (—).

In the first paper (Part I, see Logist *et al.* (2005)), the optimal steady-state results, which were previously analytically derived by applying Pontryagin's minimum principle, are numerically optimised, using a weighted shooting-type procedure ensuring the Danckwerts boundary conditions are satisfied. In addition, the influence of the trade-off coefficient A in the cost criterion is investigated.

In this second paper (Part II), the improved steady-state performance of the analytically derived bang-bang control over a more easily implementable constant jacket fluid temperature control law is illustrated. Obviously, a trade-off has to be made in practice between the decrease in performance and the extra cost and complexity of installing a second heat exchanger to impose the optimal step profile. Afterwards, the transient reactor behaviour, i.e., when the optimised control is applied from the reactor start-up, is investigated for a whole family of tubular reactors, i.e, from almost perfectly mixed reactors with a CSTR-like behaviour (for low Peclet numbers) to nearly plug flow reactors (for high Peclet numbers). Since the nature of the governing PDEs (and by consequence also the required solution method) strongly depends on the Peclet number, two different numerical schemes are adopted, exploiting their specific strong points. For low and intermediate Peclet numbers a high-order stencil from the Matlab-based MatMOL toolbox, which implements a finite differences method of lines (MOL) approach, is employed, while for high Peclet values an adapted version of a sequencing method is implemented. The reactor simulations show that physical or constructive constraints are never violated during the transient regime and that convergence towards the optimised steady-state results is always achieved. Finally, the computational efficiency of two numerical methods for solving the steady-state problem, i.e., the weighted shooting-type procedure employed in Part I and the (false) transient method used in this paper, is compared.

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