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Derivation of generic optimal reference temperature profiles for steady-state exothermic jacketed tubular reactors

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Abstract

Despite the existence of advanced controllers for jacketed tubular reactors, generic optimal steady-state reference temperature profiles are rather scarce. In this paper optimal (infinite dimensional) temperature profiles are derived by exploiting a combination of analytical and numerical optimal control techniques for two cost criteria which both involve a trade-off between a conversion and an energy cost. Hereby, the following results are obtained: (i) a more complex model which also incorporates heat transfer, yields more valuable references than a simple model which only takes the reaction into account, (ii) the analytical approach allows to extract generic features in the reference profiles, enhancing the robustness against model mismatch, and (iii) the performance loss due to practical implementation hardware, i.e., the availability of only a finite number of isothermal jacket elements, is quantified.

Key words: process control, optimal control theory, jacketed tubular reactor

1 Introduction

Due to market saturation and global competition, today's chemical industry strives for more efficient processes to reduce the production costs. Mathematical process models have proven over the past decades to be extremely valuable tools for optimising the operation and control of chemical processes, resulting in the desired profit improvement [1,2]. Application of these model based techniques to jacketed tubular reactors, still important workhorses in process industry, paves the way to improved operation policies.

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In practice, the jacket of a tubular reactor often consists of a finite number of *isothermal jacket parts*. Several advanced control schemes have been developed for this type of reactors [3–6], which all require optimal temperature reference profiles. Reference temperature profiles can be computed using optimal control theory. However, it is not always clear which variable has to be selected as the independent variable in the resulting optimal control problems, e.g., the reactor temperature itself, [7–10], the reactor temperature gradient [11,12], the heat transfer coefficient distributions [13–16] or the jacket fluid temperature [17–19]. In addition, most of the references are derived merely numerically, i.e., (i) without extracting generic analytical features which remain valid under model mismatch, and (ii) without assessing the performance loss (compared to the theoretically/conceptually best performance) that is associated with the hardware of the practical controller implementation, i.e., the fact that the optimal temperature profile cannot be implemented as such.

Motivated by these observations, this study focusses on the derivation of generic reference temperature profiles for the optimal and safe steady-state operation of an exothermic jacketed tubular reactor by exploiting both analytical and numerical optimal control techniques. Two different cost criteria which both combine a conversion and an energy cost are considered. Hereby, reference temperature profiles are computed from two different optimal control problems. The first reference is based on the reaction model only, while the second relies on a model which also incorporates the heat transfer between the reactor and its jacket. The former formulation yields a simpler optimal control problem with the temperature as independent variable while the latter leads to a more complex optimal control problem with the jacket fluid temperature as independent variable. However, due to incorporation of heat transfer issues, the references based on the latter approach may yield a better performance in practice as the model stands closer to reality. Therefore, the performance of both reference temperature profiles is assessed for a practical reactor configuration with a finite number of isothermal jacket parts and the performance loss due to the hardware is evaluated. In addition, also the robust character of the generic features under model mismatch is illustrated.

The organisation of the paper is as follows. Section 2 introduces the reactor model. Section 3 reviews the optimisation procedure, which is applied in Section 4 to obtain the generic solutions. Section 5 discusses practical issues related to (i) the reactor model on which the reference has to be based for a practical reactor, (ii) the performance loss due to the actual hardware (i.e., a limited number of isothermal jackets), and (iii) the robustness of (the generic features of) the profiles under uncertainty. Section 6 summarises the conclusions.

2 Tubular reactor

2.1 Configuration and mathematical model

Inside the tubular reactor under study an exothermic, irreversible first-order reaction with Arrhenius type kinetics takes place. To remove the reaction heat a jacket surrounds the reactor. Since in practice convective phenomena often dominate over diffusive and dispersive phenomena, a plug flow reactor model is adopted to describe the reactor behaviour. Assuming in addition, perfect radial mixing, a constant volume of the fluid in the reactor, and a constant density and heat capacity of the reacting fluid [20], the model consists of a set of non-linear ordinary differential equations (ODEs).

$$\begin{aligned} \frac{dC}{dz} &= -\frac{k_0}{v} C e^{\frac{-E}{RT}} & C(0) &= C_{in} \\ \frac{dT}{dz} &= -\frac{\Delta H}{v\rho C_p} k_0 C e^{\frac{-E}{RT}} + \frac{4h}{v\rho C_p d} (T_w - T) & T(0) &= T_{in} \end{aligned}$$

where C [mole $\cdot$ L $^{-1}$] and T [K] indicate the reactant concentration and the temperature, v [m $\cdot$ s $^{-1}$] the fluid velocity, ΔH [J $\cdot$ kmole $^{-1}$] the heat of reaction ($\Delta H < 0$ for an exothermic reaction) and ρ [kg $\cdot$ m $^{-3}$], C_p [J $\cdot$ kg $^{-1}$ $\cdot$ K $^{-1}$], k_0 [s $^{-1}$], E [J $\cdot$ mole $^{-1}$], R [J $\cdot$ mole $^{-1}$ $\cdot$ K $^{-1}$], h [W $\cdot$ m $^{-2}$ $\cdot$ K $^{-1}$], d [m] and T_w [K] the fluid density, the specific heat, the kinetic constant, the activation energy, the ideal gas constant, the heat transfer coefficient, the reactor diameter and the jacket fluid temperature, respectively. For the practical configuration with N isothermal jacket parts, the jacket temperature can be written as:

$$T_w = \sum_{i=1}^N [Hs(z - z_i) - Hs(z - z_{i+1})] T_{w,i}$$

with $Hs(z)$ the standard Heavyside function and $T_{w,i}$ the temperature in the i^{th} jacket part.

To compensate for the large differences in order of magnitude of the model variables (possibly resulting in numerical simulation and optimisation inaccuracies), dimensionless variables x_1 , x_2 and x_3 are introduced:

$$x_1 \triangleq \frac{C_{in} - C}{C_{in}}, \quad x_2 \triangleq \frac{T - T_{in}}{T_{in}} \quad \text{and} \quad x_3 \triangleq \frac{T_w - T_{in}}{T_{in}}.$$

When defining the constants α , β , γ and δ as follows:

$$\alpha \triangleq k_0 e^{\frac{-E}{RT_{in}}}, \beta \triangleq \frac{4h}{\rho C_p d}, \gamma \triangleq \frac{E}{RT_{in}}, \text{ and } \delta \triangleq -\frac{\Delta H C_{in}}{\rho C_p T_{in}},$$

the steady-state plug flow equations are:

$$\frac{dx_1}{dz} = \frac{\alpha}{v} (1 - x_1) e^{\frac{\gamma x_2}{1+x_2}} \quad (1)$$

$$\frac{dx_2}{dz} = \frac{\alpha \delta}{v} (1 - x_1) e^{\frac{\gamma x_2}{1+x_2}} + \frac{\beta}{v} (x_3 - x_2) \quad (2)$$

subject to the boundary conditions: $x_1(0) = 0$ and $x_2(0) = 0$.

2.2 Optimisation strategy

As a tubular reactor is a continuous system, it is meant to be operated in steady-state in order to produce large amounts of a product with a high and constant quality. Transient operation phases such as start-up, shut-down or transitions, usually only constitute a minor fraction of the time. Hence, most effort is made to optimise the reactor performance under steady-state conditions as it may not be worthwhile to optimise the transients.

The optimal control problem of finding generic optimal steady-state reference profiles along the reactor length can be tackled from different angles. First, the *temperature profile inside the reactor* $T(z)$ can be selected as the independent variable. This option ensures a very direct influence on the reactor performance, leading to a highly efficient conceptual reactor operation. However, the practical implementation of the resulting temperature profile via the surrounding reactor jacket may be difficult due to possible discontinuities in the reference profile, requiring an infinite heat transfer. When starting from a model that also describes the heat transfer between the reactor and its jacket, a second, alternative optimal control problem with the jacket fluid *jacket fluid temperature profile* $T_w(z)$ as independent variable is obtained. The resulting optimal temperature profiles, which already account for heat transfer restrictions, can also serve as references. However, a performance loss is expected due to a less direct influence on the elements in the performance measure. Furthermore, an additional performance loss is encountered, as the jacket fluid temperature T_w cannot vary continuously in space, but is restricted to a finite number of isothermal parts $T_{w,i}$.

Optimal infinite dimensional spatial profiles, e.g., $T(z)$ and $T_w(z)$, can be obtained using analytical and/or numerical *optimal control* techniques (see [21])

for an excellent overview). Whenever a finite number of degrees of freedom has to be optimised, e.g., the temperatures $T_{w,i}$ in the various isothermal jackets purely numerical techniques are resorted to.

3 Optimal control problem

3.1 Statement of the optimal control problem

The optimal control problems encountered in this study consist of *finding an admissible control along the spatial dimension of the reactor* $u(z) \in U$ *which causes the reactor to follow an admissible state trajectory* $\mathbf{x} \in X$, *while at the same time minimising a cost criterion* J [22,23], or when formulated mathematically:

$$\min_{u(z), z \in [0, L]} J = \underbrace{h[\mathbf{x}(L)]}_{\text{Terminal cost}} + \underbrace{\int_0^L g[\mathbf{x}(z), u(z)] dz}_{\text{Integral cost}} \quad (3)$$

subject to the system equations and path constraints on the states and control:

$$\frac{d\mathbf{x}}{dz} = \mathbf{f}[\mathbf{x}(z), u(z), z] \quad \text{with} \quad \mathbf{x}(0) = \mathbf{x}_0 \quad (4)$$

$$\mathbf{0} \geq \mathbf{C}[\mathbf{x}(z), u(z), z] \quad (5)$$

Remark 1. Since in practice often only upper and/or lower limits are present, this study only considers simple bounds on (i) the control ($u_{min} - u(z) \leq 0$ and $u(z) - u_{max} \leq 0$) and on (ii) the states ($\mathbf{x}_{min} - \mathbf{x}(z) \leq \mathbf{0}$ and $\mathbf{x}(z) - \mathbf{x}_{max} \leq \mathbf{0}$).

Remark 2. The terminal cost part takes reactor outlet values into account, whereas the integral cost part takes account of complete reactor profiles. This classification into terminal and integral cost parts allows an easy interpretation of the cost criterion. However, to facilitate the forthcoming mathematical derivations, the integral term is converted into an additional terminal term $x_{cost}(L)$ by introducing an additional state $x_{cost}(z)$:

$$x_{cost}(z) \triangleq \int_0^z g[\mathbf{x}(z), u(z)] dz$$

When defining the augmented state $\mathbf{x}_a$, the augmented system equations $\mathbf{f}_a$ and the augmented terminal cost $h_a[\mathbf{x}_a(L)]$ as follows:

$$\mathbf{x}_a \triangleq \begin{bmatrix} \mathbf{x}(z) \\ x_{cost}(z) \end{bmatrix},$$

$$\mathbf{f}_a[\mathbf{x}_a(z), u(z), z] \triangleq \begin{bmatrix} \mathbf{f}[\mathbf{x}(z), u(z), z] \\ g[\mathbf{x}(z), u(z), z] \end{bmatrix}$$

with $\mathbf{x}_{a,0} \triangleq \begin{bmatrix} \mathbf{x}_0 \\ 0 \end{bmatrix}$ and $h_a[\mathbf{x}_a(L)] \triangleq h[\mathbf{x}(L)] + x_{cost}(L)$ an equivalent *terminal cost* optimal control problem is obtained:

$$\min_{u(z), z \in [0, L]} J = h_a[\mathbf{x}_a(L)] \quad (6)$$

$$\text{subject to } \frac{d\mathbf{x}_a}{dz} = \mathbf{f}_a[\mathbf{x}_a(z), u(z), z] \quad \text{with } \mathbf{x}_a(0) = \mathbf{x}_{a,0} \quad (7)$$

$$\mathbf{0} \geq \mathbf{C}[\mathbf{x}_a(z), u(z), z] \quad (8)$$

3.2 Pontryagin's Minimum Principle

According to the Minimum Principle of Pontryagin the abovementioned (augmented) optimal control problem can be reformulated as that of minimising the Hamiltonian function H with respect to the control input $u(z)$ [21,23]:

$$\min_{u(z), z \in [0, L]} H = \boldsymbol{\lambda}^T \mathbf{f}_a[\mathbf{x}_a(z), u(z), z] + \boldsymbol{\mu}^T \mathbf{C}[\mathbf{x}_a(z), u(z), z]$$

subject to the state equations (7), the system of costates $\boldsymbol{\lambda}(z)$, and the path constraints:

$$\frac{d\boldsymbol{\lambda}}{dz} = -\frac{\partial H}{\partial \mathbf{x}_a} \quad \text{with } \boldsymbol{\lambda}(L) = \frac{\partial h_a}{\partial \mathbf{x}_a} \bigg|_L$$

$$0 = \boldsymbol{\mu}^T \mathbf{C}$$

with $\boldsymbol{\mu}(z)$ a vector of Lagrange multipliers, which are strictly positive when the corresponding path constraint is active and zero otherwise. To minimise the Hamiltonian the following necessary condition has to be satisfied:

$$\frac{\partial H}{\partial u} \equiv \boldsymbol{\lambda}^T \frac{\partial \mathbf{f}_a}{\partial u} + \boldsymbol{\mu}^T \frac{\partial \mathbf{C}}{\partial u} = 0 \quad (9)$$

Since there are boundary conditions at both the inlet and the outlet, the necessary conditions result in a so-called *two point boundary value problem*.

Additionally, an optimal solution consists of one or more different intervals. Within each interval the control is continuous and differentiable, whereas discontinuities are allowed at the interval borders or *switching points*. Therefore, a global analytical optimal control is in most cases impossible to compute. For terminal cost optimal control problems, however, analytical expressions for the control within each interval can be obtained based on the elimination of the costates according to the following procedure [21].

- *With active constraints* the control is independent of the costates and can be found by repeatedly differentiating the active constraint with respect to the spatial coordinate z and substituting $\mathbf{f}_a[\mathbf{x}_a(z), u(z), z]$ for $\frac{d\mathbf{x}_a}{dz}$ until the control u appears explicitly. For instance in the case of simple bounds on the control input, no differentiation at all is required. The constrained control is simply equal to its lower or upper limit, u_{min} or u_{max} , respectively.
- *Without active constraints* an optimal solution independent of the costates can be found based on the loss of rank of a matrix $\mathbf{M}$. The operator Δ for an arbitrary vector $\mathbf{v}$ is defined as:

$$\Delta \mathbf{v} = \frac{\partial \mathbf{v}}{\partial \mathbf{x}_a} \mathbf{f}_a - \frac{\partial \mathbf{f}_a}{\partial \mathbf{x}_a} \mathbf{v} + \sum_{k=0}^{\infty} \frac{\partial \mathbf{v}}{\partial u^{(k)}} u^{(k+1)}$$

with $u^{(k)}$ the k -th derivative of u with respect to z . The matrix $\mathbf{M}$:

$$\mathbf{M} = \left[\frac{\partial \mathbf{f}_a}{\partial u} : \Delta \frac{\partial \mathbf{f}_a}{\partial u} : \Delta \left(\Delta \frac{\partial \mathbf{f}_a}{\partial u} \right) : \dots \right]$$

is obtained by continuing the differentiation of $\frac{\partial \mathbf{f}_a}{\partial u}$ until the structural rank of $\mathbf{M}$ is equal to ρ , the dimension of the state space that can be reached by manipulating u . If $\rho = n$, with n the number of states, the condition $\det(\mathbf{M}) = 0$ is equivalent to the set of costate equations and yields the optimal control u . The system described by the condition $\det(\mathbf{M}) = 0$ may contain derivatives of u of which the initial values then constitute a set of additional decision variables. If $\rho < n$, a transformation is required to group all states influenced by u . The determinant of the corresponding submatrix of $\mathbf{M}$ has to be employed in this case. For *control affine* Hamiltonians the determinant condition reveals the possible occurrence of *singular arcs*.

The resulting constrained and unconstrained arcs must be pieced together (i.e., the optimal sequence and the optimal switching points have to be determined). When the optimal sequence is known, only the switching points have to be determined by optimisation. Physical insight can help to guess the optimal structure, or alternatively, a suitable purely numerical technique, e.g., control vector parameterisation [24–27] can directly lead to the identification of the optimal sequence. Moreover, an automated structure detection procedure has been developed [28,29].

3.3 Control vector parameterisation

Control vector parameterisation (CVP) is a purely numerical method for solving optimal control problems, which transforms the original infinite dimensional optimisation problem described by Equations (6), (7) and (8) into a finite dimensional Non-Linear Program (NLP). Hereto, the control input is parameterised (often by piecewise constant parts), whereas the state equations remain continuous. The optimal parameter values are obtained through minimisation of the cost criterion by an NLP-solver. The Fortran code used in this study implements a CVP approach based on the following elements and techniques.

- **Parameterisation.** The parameterisation allows (discontinuous) *piecewise constant* (PWC) parts on both fixed and adaptive grids.
- **NLP solvers.** Four routines are available: two *stochastic*, global routines, i.e., Integrated Controlled Random Search [30] and Particle Swarm Optimisation (see, e.g., [31]) and two *deterministic*, local, gradient based Successive Quadratic Programming (SQP) routines from the NAG Fortran library [32], i.e., E04UCF (based on NPSOL [33]) and E04UGF (based on SNOPT [34]).
- **Gradient calculation.** For both SQP routines estimates for the gradient of the cost function are required. In the program, these values can be computed either by *finite difference perturbations* or by integration of the first-order *sensitivities* of the ODE system, which is numerically more reliable [25,35].
- **ODE solver.** The integration of both the system and the sensitivity equations is carried out by D02EJF (from the NAG Fortran library [32]), a stiff initial value problem solver which uses a variable order, variable step method implementing the Backward Differential Formulae (BDF).
- **Constraint handling.** *Control constraints* are implemented as bounds on the optimisation variables. *State constraints* are taken into account in two ways [26], (i) as point constraints at the discretisation points, and (ii) through forcing the integral of the violation below a small tolerance.

4 Steady-state operation

The two cost criteria considered in this study are the same as in Smets et al. [17]. The criteria involve an inherent trade-off between maximising a reactant conversion and minimising an energy cost, and reflect the current trend towards process integration and intensification. The conversion cost is measured by the reactant concentration at the reactor outlet. This terminal cost is extended in the first criterion (10) with a second terminal cost that penalises the deviation of the exit temperature $T(L)$ from the inlet temperature T_{in} in order to minimise downstream processing costs induced by (too) hot streams

and to maximise the recuperation through the jacket of the heat generated by the reaction. To minimise the chance of hot spots occurring, an integral type cost, which penalises temperature deviations from the inlet temperature along the reactor, is added in the second criterion (11).

$$J_{TC} = (1 - A)C(L) + A \frac{1}{K_1} (T(L) - T_{in})^2 \quad (10)$$

$$J_{TIC} = (1 - A)C(L) + A \int_0^L \frac{1}{K_2} (T(z) - T_{in})^2 dz \quad (11)$$

The parameter A [-], which can range from zero to one, enables a trade-off between the two cost parts, while the parameters K_1 [$\text{K}^2 \cdot \text{mole}^{-1} \cdot \text{L}$] and K_2 [$\text{K}^2 \cdot \text{mole}^{-1} \cdot \text{L}$] are scaling factors in order to bring both cost parts in the same order of magnitude. Equivalent formulations (Equations (12) and (13)) are obtained when using the dimensionless variables.

$$J_{TC} = (1 - A)C_{in}(1 - x_1(L)) + A \frac{T_{in}^2}{K_1} x_2^2(L) \quad (12)$$

$$J_{TIC} = (1 - A)C_{in}(1 - x_1(L)) + A \int_0^L \frac{T_{in}^2}{K_2} x_2^2(z) dz \quad (13)$$

For both criteria, the reactor temperature T and jacket fluid temperature T_w are selected as independent variables for the optimal control problem, each time accounting for bounds on the concentration ($0 \text{ mole} \cdot \text{L}^{-1} \equiv C_{min} \leq C(z) \leq C_{max} \equiv C_{in}$), the temperature ($T_{min} \leq T(z) \leq T_{max}$) and the jacket fluid temperature ($T_{w,min} \leq T_w(z) \leq T_{w,max}$).

Optimal profiles for the reactor temperature $T(z)$ and jacket temperature $T_w(z)$ are determined in three steps. First, analytical expressions for all possible arcs are computed based on the abovementioned costate elimination procedure. Second, optimal control profiles are determined numerically through CVP using a fixed uniform PWC parameterisation with 50 pieces. Hereby, the hybrid combination of a stochastic (ICRS) and a deterministic (E04UCF) NLP-solver significantly increases the chances of converging to the global minimum [36]. Gradients are calculated using the first-order sensitivities. This choice for the combination of ICRS and E04UCF with gradients based on sensitivities is given by its best performance in preliminary tests. From the numerically obtained optimal control profile the optimal sequence of arcs is identified. In the third step a new control parameterisation is built using the analytical expressions found in the first step and their optimal sequence obtained in step 2. This analytical parameterisation (AP) approach then leads to an optimisation problem with mainly the switching positions as decision variables.

The same parameter values as in [17], are adopted (Table 1). First, the results for the terminal cost criterion (10) are presented, after which the results for the combined terminal and integral cost criterion (11) are discussed.

4.1 Terminal cost criterion

In this section optimal profiles are derived for the terminal cost criterion.

4.1.1 Reactor temperature $T(z)$

As a first case the reactor temperature T is selected as independent variable for the optimal control problem. Consequently, the mathematical model only consists of Equation (1) with the dimensionless concentration x_1 as the state variable and the dimensionless temperature x_2 as the control input u . The terminal cost criterion for this case does not fit into the general optimal control problem formulation stated by Equations (3), (4) and (5) as the terminal cost part also contains the control variable u , i.e., the reactor temperature T at the reactor outlet $z = L$. Therefore, the cost criterion has to be modified slightly. The second part in the cost criterion only vanishes when the outlet temperature $T(L)$ is set equal to the inlet temperature T_{in} . This situation can be enforced by adding a point constraint $T(L) = T_{in}$. In the formalism of Equation (8), such a point constraint is equivalent to $T(L) \leq T_{in}$ and $T_{in} \leq T(L)$ (or $u(L) \leq u_{point} = 0$ and $0 = u_{point} \leq u(L)$). These adaptations make the bounds for u varying in space, but do not affect the conversion. The terminal cost criterion now reads: $J_{TC} = (1 - A)C(L)$, or:

$$J_{TC} = (1 - A)C_{in}(1 - x_1(L)). \quad (14)$$

A summary of the entire problem can be found in the overview Table 2.

As outlined before, in a first step, the set of all possible arcs is determined analytically through elimination of the costates. Assuming active constraints reveals three possible arcs, i.e., u_{min} , u_{max} and u_{point} . A control taking care of active concentration constraints is not necessary as the reactant concentration of an irreversible first-order reaction decreases strictly monotonically towards zero, which can only be reached after an infinite reaction time. This result is true for all operation levels and, hence, it will not be repeated every time. To check the possibility of an arc inside the feasible region, i.e., when no state or control constraint is active, the system model and its first derivative are required:

$$\mathbf{f}_a = \left[\frac{\alpha}{v}(1 - x_1)e^{\frac{\gamma u}{1+u}} \right]; \quad \frac{\partial \mathbf{f}_a}{\partial u} = \left[\frac{\alpha}{v}(1 - x_1)e^{\frac{\gamma u}{1+u}} \frac{\gamma}{(1 + u)^2} \right]$$

The matrix $\mathbf{M} = \left[\frac{\partial \mathbf{f}_a}{\partial u} \right]$ has a structural rank of one. To satisfy the determinant condition $\det(\mathbf{M}) = 0$, an infinite temperature or an infinite reactor length is required, because complete conversion, i.e., $(1 - x_1(L)) = 0$, is only achieved for an irreversible first-order reaction after an infinite reaction time.

In a second step, CVP is employed to determine the optimal control profiles numerically. The results are depicted in Figure 1 and indicate a sequence of u_{max} inside the reactor and u_{point} at the reactor outlet to be optimal. The interpretation based on the different cost terms, is that high temperatures inside the reactor are favourable for the conversion. Only at the reactor outlet the temperature is equal to T_{in} in order to minimise the terminal heat loss.

In a third step, the AP is optimised with only the switching position from u_{max} to u_{point} as a decision variable. As can be seen from Figure 1 the AP results confirm the previous observations. Note also that the difference between the CVP and the AP results is hardly visible.

The abovementioned profile yields the lowest cost value, nevertheless, the implementation is rather hard as it requires an almost infinitely fast increase from T_{in} to T_{max} at the inlet and an almost infinitely fast decrease from T_{max} to T_{in} at the outlet. The explanation of this optimal profile reflects the inherent trade-off in the cost criterion. Because high temperatures favour conversion, it is important to reach T_{max} as fast as possible and to maintain this temperature as long as possible. However, to reduce the terminal heat loss the temperature has to be lowered (as fast as possible) towards the reactor outlet.

4.1.2 Jacket temperature $T_w(z)$

As a second case the jacket fluid temperature T_w is adopted as independent variable for the optimal control problem. Consequently, the mathematical model consists of Equations (1) and (2) with the dimensionless concentration x_1 and the dimensionless temperature x_2 as state variables and the dimensionless jacket fluid temperature x_3 as the control input u . A summary of the resulting optimal control problem is provided in the overview Table 2.

The set of constrained arcs contains three elements: u_{min} , u_{max} and u_{path} . Differentiating the reactor temperature constraint once with respect to the spatial coordinate z yields the control input which keeps this constraint active:

$$u_{path} = x_2 - \frac{\alpha\delta}{\beta}(1 - x_1)e^{\frac{\gamma x_2}{1+x_2}}$$

The matrix $\mathbf{M} = \left[\frac{\partial \mathbf{f}_a}{\partial u} : \Delta \frac{\partial \mathbf{f}_a}{\partial u} \right]$ is constructed using:

$$\mathbf{f}_a = \begin{bmatrix} \frac{\alpha}{v}(1-x_1)e^{\frac{\gamma x_2}{1+x_2}} \\ \frac{\alpha\delta}{v}(1-x_1)e^{\frac{\gamma x_2}{1+x_2}} + \frac{\beta}{v}(u-x_2) \end{bmatrix}$$

$$\frac{\partial \mathbf{f}_a}{\partial u} = \begin{bmatrix} 0 \\ \frac{\beta}{v} \end{bmatrix}; \quad \Delta \frac{\partial \mathbf{f}_a}{\partial u} = \begin{bmatrix} -\frac{\beta}{v} \frac{\alpha}{v} (1-x_1) e^{\frac{\gamma x_2}{1+x_2}} \frac{\gamma}{(1+x_2)^2} \\ -\frac{\beta}{v} \left(\frac{\alpha\delta}{v} (1-x_1) e^{\frac{\gamma x_2}{1+x_2}} \frac{\gamma}{(1+x_2)^2} - \frac{\beta}{v} \right) \end{bmatrix}$$

Based on the analysis of the matrix $\mathbf{M}$ and its determinant:

$$\det(\mathbf{M}) = \frac{\beta^2}{v^2} \frac{\alpha}{v} (1-x_1) e^{\frac{\gamma x_2}{1+x_2}} \frac{\gamma}{(1+x_2)^2} = 0$$

it is found that an optimal arc inside the feasible region is impossible. Similarly, $\mathbf{M}$ has a structural rank of two and can only lose rank for an infinite reactor length or an infinite temperature. As can be seen in Figure 2, depicting the optimal profiles based on the CVP approach, the optimal sequence is identified as u_{max} , followed by u_{min} , u_{path} and u_{min} . The switching positions of the optimal sequence using the analytical expressions for the different arcs are then optimised in the AP approach, yielding the final results.

The interpretation of the phases is the same as before, i.e., (i) heating-up as fast as possible until T_{max} is reached, (ii) remaining at T_{max} and (iii) finally cooling-down as fast as possible. The first two phases increase the conversion, whereas the last part decreases the terminal energy loss. Note the small u_{min} part in between u_{max} and u_{path} . This part is necessary because u_{path} cannot be entered directly after u_{max} when attaining T_{max} , as the path constrained control input u_{path} requires in that case values lower than the minimum u_{min} . Without a temperature limit, the constrained parts vanish and the optimal control is of the *max-min* type which confirms the analytical results of [17].

4.2 Terminal and integral cost criterion

In this section optimal profiles are derived for the terminal and integral cost criterion. Note that for this case *optimal control theory* has to be applied to the augmented model which takes the integral cost part into account via an additional cost state.

$$\frac{dx_{cost}}{dz} = A \frac{T_{in}^2}{K_2} x_2^2 \quad \text{with} \quad x_{cost}(0) = 0 \quad (15)$$

As a result the integral cost part can be transformed into a terminal cost.

$$J_{TIC} = (1 - A)C_{in}(1 - x_1(L)) + x_{cost}(L) \quad (16)$$

4.2.1 Reactor temperature $T(z)$

Again the reactor temperature T is first selected as the independent variable for the optimal control problem. Consequently, the mathematical model consists of Equation (1) augmented with Equation (15). The dimensionless concentration x_1 and the cost state x_{cost} are the state variables and the dimensionless temperature x_2 acts as the control input u . A summary of the optimal control problem is presented in Table 2.

Since only simple bounds are imposed on the control, only u_{min} and u_{max} are required as constrained arcs. The matrix $\mathbf{M} = \left[\frac{\partial \mathbf{f}_a}{\partial u} : \Delta \frac{\partial \mathbf{f}_a}{\partial u} \right]$ is constructed based on:

$$\mathbf{f}_a = \begin{bmatrix} \frac{\alpha}{v}(1 - x_1)e^{\frac{\gamma u}{1+u}} \\ A \frac{T_{in}^2}{K_2} u^2 \end{bmatrix}; \quad \frac{\partial \mathbf{f}_a}{\partial u} = \begin{bmatrix} \frac{\alpha}{v}(1 - x_1)e^{\frac{\gamma u}{1+u}} \frac{\gamma}{(1+u)^2} \\ 2A \frac{T_{in}^2}{K_2} u \end{bmatrix};$$

$$\Delta \frac{\partial \mathbf{f}_a}{\partial u} = \begin{bmatrix} \frac{\alpha}{v}(1 - x_1)e^{\frac{\gamma u}{1+u}} \frac{\gamma}{(1+u)^2} \left(\frac{\gamma}{(1+u)^2} - \frac{2}{(1+u)} \right) \\ 2A \frac{T_{in}^2}{K_2} \end{bmatrix} \frac{du}{dz}$$

This matrix clearly has a differential nature since the first derivative of the control appears explicitly. The structural rank of the matrix is two and according to the determinant condition:

$$\det(\mathbf{M}) = 2A \frac{T_{in}^2}{K_2} \frac{\alpha}{v} (1 - x_1) e^{\frac{\gamma u}{1+u}} \frac{\gamma}{(1+u)^2} \left[1 - u \left(\frac{\gamma}{(1+u)^2} - \frac{2}{(1+u)} \right) \right] \frac{du}{dz} = 0$$

there are only two possibilities for the matrix to lose rank since only the two last factors can be equal to zero for a reactor of finite length with finite temperatures. The former factor is only equal to zero for a temperature of 367.38 K, while the latter induces a constant reactor temperature when being equal to zero. As the latter requirement is a generalisation of the former, it is clear that the determinant condition is always satisfied when a constant reactor temperature is applied. The exact constant temperature values become additional decision variables in the AP, in addition to the switching points.

Additional information on the optimal solution can be obtained when directly

using Pontryagin's formulation. This leads to the following Hamiltonian H :

$$H = \lambda_1 \frac{\alpha}{v} (1 - x_1) e^{\frac{\gamma u}{1+u}} + \lambda_{cost} A \frac{T_{in}^2}{K_2} u^2 + \mu_1 (u_{min} - u) + \mu_2 (u - u_{max})$$

subject to the dynamic costate equations $\frac{d\lambda_1}{dz} = \lambda_1 \frac{\alpha}{v} e^{\frac{\gamma u}{1+u}}$ and $\frac{d\lambda_{cost}}{dz} = 0$ with as boundary conditions $\lambda_1(L) = -(1 - A)C_{in}$ and $\lambda_{cost}(L) = 1$. The necessary condition (9) now becomes:

$$\frac{\partial H}{\partial u} = \lambda_1 \frac{\alpha}{v} (1 - x_1) e^{\frac{\gamma u}{1+u}} \frac{\gamma}{(1+u)^2} + 2\lambda_{cost} A \frac{T_{in}^2}{K_2} u - \mu_1 + \mu_2 = 0 \quad (17)$$

The following additional conditions from [23] lead to interesting insights.

Condition 1. For problems with control inequality constraints all the costates, the Hamiltonian and its first derivative with respect to u have to be continuous across a switching point.

Condition 2. If the Hamiltonian does not explicitly depend on the independent variable, i.e., the spatial coordinate z , the Hamiltonian is constant when evaluated on an optimal trajectory.

Hence, based on the first additional condition, it is clear that the costate $\lambda_{cost}(z)$ equals one over the entire range. Consequently, it is replaced by this value in all further derivations. Furthermore, since in this case $\frac{d(\lambda_1(1 - x_1))}{dz} = 0$ and both λ_1 and x_1 are continuous, the product $\lambda_1(1 - x_1)$ is continuous and constant over the entire length. As the Hamiltonian must be constant (and continuous across the possible switchings) when evaluated on an optimal trajectory, it is clear that the optimal control consists of one or more constant intervals, which confirms the previous result. Finally, since the product $\lambda_1(1 - x_1)$ and the optimal control are constants, the Lagrange multipliers also have a constant value inside each interval, i.e., zero when the constraints are inactive (i.e., when $u > u_{min}$ for μ_1 and when $u < u_{max}$ for μ_2) and strictly positive otherwise. Since it is not a priori known whether or not the optimal solution comprises bounded arcs, both possibilities have to be checked.

- *Assuming bounded arcs* are part of the optimal control, the additional conditions have to be satisfied across the switching point. Conceptually, three arc types are possible, i.e., at the upper bound u_{max} , at the lower bound u_{min} and inside the feasible region u_{inside} . Combining these parts allows four switching possibilities, i.e., between u_{inside} and u_{max} , between u_{inside} and u_{min} , between u_{min} and u_{max} , and between two different values inside the feasible region $u_{inside,1}$ and $u_{inside,2}$. Using the continuity of the Hamiltonian together with the continuity of $\lambda_1(1 - x_1)$, the following relation is

found for two different optimal controls u_1 and u_2 across a switching point:

$$\frac{\alpha}{v} \lambda_1 (1 - x_1) = A \frac{T_{in}^2}{K_2} \frac{u_1^2 - u_2^2}{e^{\frac{\gamma u_2}{1+u_2}} - e^{\frac{\gamma u_1}{1+u_1}}}$$

This relation is then substituted into the first-order necessary condition (17). Applying the resulting condition to each of the two intervals, i.e., before and after the switching point, yields two additional relations from which two unknown values can be computed. When switching between $u_1 = u_{max}$ and $u_2 = u_{min}$, the two active Lagrange multipliers μ_1 and μ_2 can be computed. When switching between $u_1 = u_{inside}$ and $u_2 = u_{min}$ or between $u_1 = u_{inside}$ and $u_2 = u_{max}$, the active Lagrange multiplier can be calculated from the necessary condition on the constrained part, whereas for the part inside the feasible region, the necessary condition yields the following non-linear relation from which the optimal constant control value u_1 can be computed:

$$A \frac{T_{in}^2}{K_2} \left(\frac{u_1^2 - u_2^2}{e^{\frac{\gamma u_2}{1+u_2}} - e^{\frac{\gamma u_1}{1+u_1}}} e^{\frac{\gamma u_1}{1+u_1}} \frac{\gamma}{(1+u_1)^2} + 2u_1 \right) = 0 \quad (18)$$

Solution of the non-linear equation for u_2 equal to u_{min} and u_{max} yields for u_1 optimal temperature levels (in dimensions) of 444.17 K and 355.27 K, respectively. Switching between two different values inside the feasible region requires the simultaneous solution of the necessary conditions for each of the u_{inside} values. However, the only solution to these equations requires $u_{inside,1} = u_{inside,2}$, which indicates that switching between two different constant levels is impossible. All these observations lead to the conclusion that only switchings between u_{max} and u_{inside} (equivalent to 355.27 K), and between u_{max} and u_{min} are possible. Note that the possible temperature levels are solely determined by the system as the zero of the non-linear equation is independent of the properties in the cost criterion, i.e., the scaling factor K_2 and the weighting factor A .

- Assuming no bounded arcs, the two point boundary value problem can be solved analytically, resulting in: $\lambda_1(1 - x_1) = -(1 - A)C_{in}e^{-BL}$ with $B = \frac{\alpha}{v} e^{\frac{\gamma u}{1+u}}$. Substituting this value in the necessary condition (17) yields the following non-linear relation from which the optimal value for u can be computed numerically:

$$(A - 1)C_{in}Be^{-BL} \frac{\gamma}{(1+u)^2} + 2 \frac{A}{K_2} T_{in}^2 u = 0. \quad (19)$$

Solution of this non-linear equation for $A = 0.5$ and $K_2 = 250000 \text{ K}^2 \cdot \text{mole}^{-1} \cdot \text{L}$ leads to a constant temperature value of 367.74 K. In this case however, the optimal constant control value does depend on the scaling factor K_2 and the weighting factor A .

In summary, the optimal reactor temperature profile consists of one or more

intervals of constant temperature. Without constrained parts, the optimal temperature is the result of a single non-linear relation (19). With bounded parts, the set of possible arcs contains u_{min} , u_{max} and u_{inside} (equivalent to 355.27 K), but only switching between either u_{max} and u_{min} , or u_{max} and u_{inside} is allowed. To obtain the exact sequence of intervals the CVP algorithm is employed. The results are depicted in Figure 3 and indicate that a profile consisting of two constant temperature intervals is optimal, i.e., a first interval at intermediate temperature switching to the upper bound towards the reactor outlet. When zooming in, it is observed that the constant intermediate temperature value obtained through CVP is equal to 355.27 K, the theoretically predicted value. Finally, the AP approach is used to refine the CVP profile. The optimal profile can be interpreted according to both parts in the cost criterion, i.e., the intermediate temperature in the first reactor part limits the global heat loss while the high temperature in the second part enforces an additional conversion towards the reactor outlet. Similar to the pure terminal cost criterion (12), the designed temperature profile yields the lowest cost values, but it is hard to implement in practice due to the jumps.

4.2.2 Jacket fluid temperature $T_w(z)$

Adopting the jacket fluid temperature in a second case as the independent variable for the optimal control problem takes heat transfer limitations into account in order to eliminate temperature jumps. The mathematical model now involves Equations (1) and (2) augmented with Equation (15). The state variables are the dimensionless concentration x_1 , the dimensionless temperature x_2 and the cost state x_{cost} , while the dimensionless jacket fluid temperature x_3 is the control input u (see Table 2).

For path constrained arcs only three types are possible, i.e., u_{max} and u_{min} induced by an active lower and upper limit on the jacket fluid temperature and u_{path} induced by an active temperature constraint. After one differentiation of the active path constraint u_{path} is found: $u_{path} = x_2 - \frac{\alpha\delta}{\beta}(1 - x_1)e^{\frac{\gamma x_2}{1+x_2}}$.

Analysis of the matrix $\mathbf{M} = \left[\frac{\partial \mathbf{f}_a}{\partial u} : \Delta \frac{\partial \mathbf{f}_a}{\partial u} : \Delta \left(\Delta \frac{\partial \mathbf{f}_a}{\partial u} \right) \right]$ based on:

$$\mathbf{f}_a = \begin{bmatrix} c_1 \\ c_2 \\ A \frac{T_{in}^2}{K_2} x_2^2 \end{bmatrix}; \frac{\partial \mathbf{f}_a}{\partial u} = \begin{bmatrix} 0 \\ \beta \\ 0 \end{bmatrix}; \Delta \frac{\partial \mathbf{f}_a}{\partial u} = \frac{\beta}{v} \begin{bmatrix} -c_1 \frac{\gamma}{(1+x_2)^2} \\ c_3 \\ -2A \frac{T_{in}}{K_2} x_2 \end{bmatrix}; \Delta \left(\Delta \frac{\partial \mathbf{f}_a}{\partial u} \right) =$$

$$\frac{\beta}{v} \begin{bmatrix} -c_1 c_2 c_4 \frac{\gamma}{(1+x_2)^2} - c_1 c_3 \frac{\gamma}{(1+x_2)^2} \\ -c_2 c_4 \frac{\alpha \delta}{v} (1-x_1) e^{\frac{\gamma x_2}{1+x_2}} \frac{\gamma}{(1+x_2)^2} - c_3 \frac{\alpha \delta}{v} (1-x_1) e^{\frac{\gamma x_2}{1+x_2}} \frac{\gamma}{(1+x_2)^2} + c_3 \frac{\beta}{v} \\ -2c_2 A \frac{T_{in}^2}{K_2} - 2c_3 A \frac{T_{in}^2}{K_2} x_2 \end{bmatrix}$$

$$\det(\mathbf{M}) = 2A \frac{T_{in}^2}{K_2} c_1 c_2 \frac{\beta^3}{v^3} \frac{\gamma}{(1+x_2)^2} [1 - x_2 c_4] = 0$$

with:

$$c_1 = \frac{\alpha}{v} (1-x_1) e^{\frac{\gamma x_2}{1+x_2}}, \quad c_2 = \frac{\alpha \delta}{v} (1-x_1) e^{\frac{\gamma x_2}{1+x_2}} + \frac{\beta}{v} (u-x_2)$$

$$c_3 = -\frac{\alpha \delta}{v} (1-x_1) e^{\frac{\gamma x_2}{1+x_2}} \frac{\gamma}{(1+x_2)^2} + \frac{\beta}{v} \text{ and } c_4 = \left(\frac{\gamma}{(1+x_2)^2} - \frac{2}{(1+x_2)} \right).$$

leads to an additional arc u_{inside} . The structural rank of $\mathbf{M}$ is three. Since infinite reactor temperatures or an infinite reactor length are not allowed, the only possibility to satisfy the determinant condition is that c_2 or the last factor of the determinant is equal to zero. Again the last factor is only zero for a (constant) reactor temperature of 367.38 K. However, this condition is a degenerate case of $c_2 = 0$, which induces a constant reactor temperature. Hence, this arc inside the feasible region is (by chance) the same as the arc required for following an active temperature constraint, i.e., $u_{inside} = u_{path}$, and is equal to the one obtained in [17]. Figure 4 displays the resulting optimal profiles obtained with the CVP approach. The optimal sequence consists of u_{max} followed by u_{inside} , u_{max} and u_{path} . The resulting reactor temperature profile is similar to the one obtained in the previous case, but (i) the jumps are relaxed and (ii) maintaining a certain temperature is now achieved through a spatially varying jacket fluid temperature. It should be noted that the induced constant intermediate reactor temperature does not differ much from 355.27 K, the optimal value for the temperature control variable, obtained from Equation (18). Using the AP approach yields the final optimal cost values which are only slightly higher than the ones of the previous case. The effect of the reactor temperature constraint is quite obvious, i.e., without an upper temperature limit the second heating phase lasts until the outlet.

5 Practical implementation issues

In this section, first the question is addressed which of the above derived reference temperature profiles has to be selected for a practical reactor configuration with a finite number of isothermal jacket parts, i.e., the one based

on the reaction model only ($T_{ref,T}$), or the one based on the model which also incorporates heat transfer issues (T_{ref,T_w}). Second, the performance loss due to the practical implementation is evaluated. To answer these two questions a reactor with 10 isothermal jacket parts of equal length serves as an illustrative example. Finally, the influence of uncertainty is assessed. It is shown that the nominal reference profiles are hardly influenced by model mismatch and that, consequently, the extracted features exhibit a generic character.

5.1 Selection of the reference temperature profile

To get an idea of which of the analytically derived temperature profiles preferably serves as a reference in a practical configuration with a fixed number of isothermal jackets of equal length, the resulting reactor profiles and performance has to be compared. Hereto, the temperature values of the different jacket parts are selected through CVP in order to minimise $\int_0^L (T_{ref,T \text{ or } T_w}(z) - T(z))^2 dz$ (while taking the bounds into account), and the resulting cost criterion value is calculated.

Figure 5 illustrates for the terminal cost criterion the resulting reactor temperature and concentration profiles for a reactor with 10 isothermal jacket parts of equal length, together with the AP references, i.e., $T_{ref,T}$ and T_{ref,T_w} , respectively. As can be seen, it is difficult to track the $T_{ref,T}$ profile since its infinitely fast temperature jumps at the in- and outlet cannot be induced as such in the practical reactor, but require some *distance*. Since the second reference T_{ref,T_w} already accounts for heat transfer limitations, the deviations between the reference and the actual reactor temperature are much smaller and despite the higher cost value for the T_{ref,T_w} profile, the resulting performance of the practical reactor which tracks this reference is lower than when tracking $T_{ref,T}$ (see also Table 3). Consequently, the more complex model which also accounts for heat transfer restrictions yields more reliable references for the practical situation. When the number of jacket parts is increased the references are more closely followed, generally resulting in a lower cost criterion value. Similar conclusions can be drawn for the combined terminal and integral cost based on Figure 6 which depicts the equivalent results of Figure 5.

5.2 Performance loss due to practical implementation hardware

Since in a practical situation with a fixed number of isothermal jacket parts it is not possible to induce reactor temperature jumps (as in the optimal control solution for the reaction model only) or a continuously spatially varying

jacket fluid temperature profile (as in the optimal control problem which also accounts for the heat transfer between the reactor and its jacket), a performance deterioration is expected. When comparing the cost values (see Table 3), it becomes clear which performance losses can be attributed to which practical restriction. For instance, the presence of a jacket, prohibiting temperature jumps, causes an increase from $8.4662 \cdot 10^{-6}$ to $1.5010 \cdot 10^{-4}$ for the terminal cost criterion, and from $2.7301 \cdot 10^{-3}$ to $2.7899 \cdot 10^{-3}$ for the combined terminal and integral cost criterion. Additionally, the cost values increase for the practical configuration when only a finite number of isothermal jacket can be used, e.g., to $1.7309 \cdot 10^{-4}$ and $2.7940 \cdot 10^{-3}$ for 10 jacket parts. Increasing the number of jacket parts decreases the performance loss as expected (Table 3), but also increases the installation cost. As already remarked before, the performance loss is much smaller for the combined terminal and integral cost criterion. This phenomenon may be attributed to the lower sensitivity induced by the presence of the arc inside the feasible region (see, e.g., [21] for additional details).

5.3 Influence of uncertainty on the reference profiles

An important issue regarding the implementation of the optimal strategies is the way of handling uncertainty [37], e.g., model mismatch and disturbances. Here, the focus is on model mismatch since disturbance rejection has to be assessed under transient conditions and is, hence, out of the direct scope of the current paper.

To evaluate the influence of model mismatch two model parameters, i.e., the reaction rate k_0 and the dimensionless heat transfer coefficient β , of the optimal control problem with as independent variable the jacket fluid temperature are perturbed by -10% and +10%. These parameters are selected as examples of parameters which merely influence the reaction model and the heat transfer model. (Other parameters have also been investigated, e.g., the dimensionless heat of reaction δ , but the observations are similar.) The resulting AP optimal reactor profiles are displayed in Figures 7 and 8. As can be seen, there are only slight deviations from the nominal solutions and the generic features are always present. For the terminal cost criterion, these features always involve the following steps: heating-up as fast as possible until the upper temperature limit of 400 K is reached and cooling down as fast as possible towards 340 K at the reactor outlet. For the combined terminal and integral cost criterion the following steps are always required: a first temperature increase to an intermediate value around 355 K, which has then to be kept constant for almost 50% of the reactor length, followed by a second increase until the upper limit is reached. In order to reduce the risk of violating the upper temperature limit, a certain back-off may be introduced in practice.

To conclude the influence of model mismatch two additional remarks should be made. First, deriving temperature references based on the reaction model only, may at first sight seem more attractive since less parameters, and hence, less uncertainty may be present. However, based on the results of the current paragraph (i.e., the small influence of parameter perturbations on the optimal references) and the previous section (i.e., the problems that a practical configuration experiences to accommodate temperature jumps), the more complex model is recommended for deriving optimal reference temperature profiles. Therefore, only the temperature references based on this model have been displayed here. Second, although different trade-off values A in the cost criterion are generally not regarded as model mismatch, this may also influence the reference profiles. However, it has been illustrated in a recent publication [38], that for these case studies the generic features remain the same even for substantial deviations from $A = 0.5$.

6 Conclusions

In this paper generic reference temperature profiles have been derived for the optimal and safe steady-state operation of an exothermic jacketed tubular reactor by exploiting both analytical and numerical optimal control techniques. Two cost criteria are studied, which both involve a conversion and an energy cost. The first criterion only consists of terminal cost terms, while the second one combines a terminal and an integral cost part.

Based on the combined analytical/numerical approach characteristic features of the induced temperature profiles have been identified and interpreted. For the terminal cost criterion it is important to keep the temperature as high as possible in the first part of the reactor (without violating the maximum temperature constraint) in order to favour conversion. In the second part, however, cooling is required to reduce the terminal heat loss. For the terminal and integral criterion, the temperature is first brought to an intermediate level and kept constant in order to reduce the global heat loss and the risk of the occurrence of hot spots, while in the second part an additional temperature rise induces additional conversion.

Related to a practical implementation with a finite number of isothermal jacket parts, it has first been evidenced that the reactor temperature reference profiles based on the more complex optimal control problem with the jacket fluid temperature as independent variable lead to a better performance since they already account for heat transfer restrictions. Second, the performance loss due to hardware restrictions, i.e., the finite number of isothermal jackets, has been evaluated. Finally, it has been shown that the extracted generic features are robust with respect to model mismatch.

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Figure 1

Fig. 1. Optimal profiles from the optimal control problem with the reactor temperature as independent variable for the terminal cost criterion: control variable (= reactor temperature T) (top) and concentration C (bottom).

Figure 2

Fig. 2. Optimal profiles from the optimal control problem with the jacket fluid temperature as independent variable for the terminal cost criterion: control variable (= jacket fluid temperature T_w) (top) and concentration C and reactor temperature T (bottom).

Figure 3

Fig. 3. Optimal profiles from the optimal control problem with the reactor temperature as independent variable for the terminal and integral cost criterion: control variable (= reactor temperature T) (top) and concentration C (bottom).

Figure 4

Fig. 4. Optimal profiles from the optimal control problem with the jacket fluid temperature as independent variable for the terminal and integral cost criterion: control variable (= jacket fluid temperature T_w) (top) and concentration C and reactor temperature T (bottom).

Figure 5

Fig. 5. Temperature and concentration profiles (—) for a reactor with 10 isothermal jacket parts obtained when tracking the previously optimal temperature references (---) for the terminal cost criterion, i.e., $T_{ref,T}$ (top) and T_{ref,T_w} (bottom).

Figure 6

Fig. 6. Temperature and concentration profiles (—) for a reactor with 10 isothermal jacket parts obtained when tracking the previously optimal temperature references (---) for the terminal and integral cost criterion, i.e., $T_{ref,T}$ (top) and T_{ref,T_w} (bottom).

Figure 7

Fig. 7. Influence of parameter perturbations on the optimal reference profiles for the terminal cost criterion, derived from the optimal control problem with the jacket fluid temperature T_w as independent variable.

Figure 8

Fig. 8. Influence of parameter perturbations on the optimal reference profiles for the terminal and integral cost criterion, derived from the optimal control problem with the jacket fluid temperature T_w as independent variable.

Table 1
Process parameters.

A	$=$	0.5	$[-]$	R	$=$	8.3145	$\text{J} \cdot \text{mole}^{-1} \cdot \text{K}^{-1}$
C_{in}	$=$	0.02	$\text{mole} \cdot \text{L}^{-1}$	T_{in}	$=$	340	K
C_{min}	$=$	0	$\text{mole} \cdot \text{L}^{-1}$	T_{min}	$=$	280	K
C_{max}	$=$	0.02	$\text{mole} \cdot \text{L}^{-1}$	T_{max}	$=$	400	K
E	$=$	47092.5	$\text{J} \cdot \text{mole}^{-1}$	$T_{w,min}$	$=$	280	K
k_0	$=$	10^6	s^{-1}	$T_{w,max}$	$=$	400	K
K_1	$=$	250000	$\text{K}^2 \cdot \text{mole}^{-1} \cdot \text{L}$	v	$=$	0.1	$\text{m} \cdot \text{s}^{-1}$
K_2	$=$	250000	$\text{K}^2 \cdot \text{mole}^{-1} \cdot \text{L}$	β	$=$	0.2	s^{-1}
L	$=$	1	m	δ	$=$	0.25	$[-]$

Table 2

Overview of all optimal control problems (with x_1, x_2, x_3 , the dimensionless concentration, reactor temperature and jacket fluid temperature, and x_{cost} the cost state for the integral cost part).

$T(z)$ independent variable	Terminal criterion	Terminal and integral criterion
Criterion J :	Equation (14)	Equation (16)
Model equations $\mathbf{f}_a$:	Equation (1)	Equations (1) and (15)
Control $u(z)$:	x_2	x_2
States $\mathbf{x}(z)$:	x_1	x_1 and x_{cost}
Bounds on control: u_{min}	$(T_{min} - T_{in})/T_{in}, \quad z \in [0, L[$	$(T_{min} - T_{in})/T_{in}$
u_{max}	$u_{point} = 0, \quad z = L$ $(T_{max} - T_{in})/T_{in}, \quad z \in [0, L[$	$(T_{max} - T_{in})/T_{in}$
Bounds on states: $x_{1,min}$	$u_{point} = 0, \quad z = L$ $(C_{in} - C_{max})/C_{in}$	$(C_{in} - C_{max})/C_{in}$
$x_{1,max}$	$(C_{in} - C_{min})/C_{in}$	$(C_{in} - C_{min})/C_{in}$
$T_w(z)$ independent variable	Terminal cost	Terminal and integral cost
Criterion J :	Equation (12)	Equation (16)
Model equations $\mathbf{f}_a$:	Equations (1) and (2)	Equations (1), (2) and (15)
Control $u(z)$:	x_3	x_3
States $\mathbf{x}(z)$:	x_1 and x_2	x_1, x_2 and x_{cost}
Bounds on control: u_{min}	$(T_{w,min} - T_{in})/T_{in}$	$(T_{w,min} - T_{in})/T_{in}$
u_{max}	$(T_{w,max} - T_{in})/T_{in}$	$(T_{w,max} - T_{in})/T_{in}$
Bounds on states: $x_{1,min}$	$(C_{in} - C_{max})/C_{in}$	$(C_{in} - C_{max})/C_{in}$
$x_{1,max}$	$(C_{in} - C_{min})/C_{in}$	$(C_{in} - C_{min})/C_{in}$
$x_{2,min}$	$(T_{in} - T_{max})/T_{in}$	$(T_{in} - T_{max})/T_{in}$
$x_{2,max}$	$(T_{in} - T_{min})/T_{in}$	$(T_{in} - T_{min})/T_{in}$

Table 3

Cost criterion values for the terminal cost criterion and the combined terminal and integral cost criterion: upper part: results when directly minimising the cost, lower part: results when following the reference profiles.

<i>Performance optimisation</i>			
Optimisation variable		J_{TC}	J_{TIC}
Reactor temperature $T(z)$		$8.4662 \cdot 10^{-6}$	$2.7301 \cdot 10^{-3}$
Jacket temperature $T_w(z)$		$1.5010 \cdot 10^{-4}$	$2.7899 \cdot 10^{-3}$
Isothermal jackets $T_{w,i}$	$N=20$	$1.5500 \cdot 10^{-4}$	$2.7900 \cdot 10^{-3}$
	$N=10$	$1.7309 \cdot 10^{-4}$	$2.7940 \cdot 10^{-3}$
	$N=5$	$2.4866 \cdot 10^{-4}$	$2.8040 \cdot 10^{-3}$
<i>Performance by following reference profiles</i>			
Optimisation variable + reference to be followed		J_{TC}	J_{TIC}
Isothermal jackets $T_{w,i} + T_{ref,T}$	$N=20$	$7.2258 \cdot 10^{-3}$	$2.8509 \cdot 10^{-3}$
	$N=10$	$7.2342 \cdot 10^{-3}$	$2.8518 \cdot 10^{-3}$
	$N=5$	$7.1225 \cdot 10^{-3}$	$2.8513 \cdot 10^{-3}$
Isothermal jackets $T_{w,i} + T_{ref,T_w}$	$N=20$	$1.5479 \cdot 10^{-4}$	$2.7901 \cdot 10^{-3}$
	$N=10$	$1.7362 \cdot 10^{-4}$	$2.7948 \cdot 10^{-3}$
	$N=5$	$2.5060 \cdot 10^{-4}$	$2.8065 \cdot 10^{-3}$

Notation

Physical variables

A	trade-off coefficient	$[-]$
C	reactant concentration	$[\text{mole} \cdot \text{L}^{-1}]$
C_p	specific heat of reactor fluid	$[\text{J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}]$
d	reactor diameter	$[\text{m}]$
E	activation energy	$[\text{J} \cdot \text{mole}^{-1}]$
h	(overall) heat transfer coefficient	$[\text{W} \cdot \text{m}^{-2} \cdot \text{K}^{-1}]$
ΔH	heat of reaction	$[\text{J} \cdot \text{kmole}^{-1}]$
k_0	kinetic constant	$[\text{s}^{-1}]$
K_1	scaling factor for the terminal cost criterion	$[\text{K}^2 \cdot \text{mole}^{-1} \cdot \text{L}]$
K_2	scaling factor for the terminal and integral cost criterion	$[\text{K}^2 \cdot \text{mole}^{-1} \cdot \text{L}]$
L	reactor length	$[\text{m}]$
R	ideal gas constant	$[\text{J} \cdot \text{mole}^{-1} \cdot \text{K}^{-1}]$
T	reactor fluid temperature	$[\text{K}]$
T_w	jacket fluid temperature	$[\text{K}]$
u	operation variable	$[-]$
v	reactor fluid superficial velocity	$[\text{m} \cdot \text{s}^{-1}]$
x_1	$(C_{in} - C)/C_{in}$, dimensionless reactant concentration	$[-]$
x_2	$(T - T_{in})/T_{in}$, dimensionless reactor temperature	$[-]$
x_3	$(T_w - T_{in})/T_{in}$, dimensionless jacket fluid temperature	$[-]$
z	spatial coordinate	$[\text{m}]$
α	$k_0 e^{\frac{-E}{RT_{in}}}$	$[\text{s}^{-1}]$
β	$4h/\rho C_p d$	$[\text{s}^{-1}]$
γ	E/RT_{in}	$[-]$
δ	$-\Delta H C_{in}/\rho C_p T_{in}$	$[-]$
ρ	reactor fluid density	$[\text{kg} \cdot \text{m}^{-3}]$

Mathematical variables

g	terminal cost part
h	integral cost part
J	cost criterion
H	Hamiltonian
$\mathbf{C}$	vector of constraints
$\mathbf{f}$	vector of system equations
$\mathbf{M}$	matrix
$\mathbf{v}$	vector
$\mathbf{x}$	vector of states
$\boldsymbol{\lambda}$	vector of costates
$\boldsymbol{\mu}$	vector of Lagrange multipliers

Subscripts

a	augmented value
$cost$	cost value
min	value at its lower bound
max	value at its upper bound
in	value at the inlet
$inside$	value inside the feasible region
$path$	value determined by path constraint
$point$	value determined by point constraint
ref	reference
TC	value for the terminal cost criterion
TIC	value for the terminal and integral cost criterion
0	initial value