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by *Filip Logist, Alain Vande Wouwer, Ilse Smets and Jan Van Impe*

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Optimal temperature profiles for tubular reactors implemented through a flow reversal strategy

F. Logist^a A. Vande Wouwer^b I.Y. Smets^a J.F. Van Impe^{a,*}

^aBioTeC, Department of Chemical Engineering, Katholieke Universiteit Leuven,
W. de Croylaan 46, B-3001 Leuven, Belgium

^bService d'Automatique, Faculté Polytechnique de Mons,
Boulevard Dolez 31, B-7000 Mons, Belgium

Abstract

Optimal steady-state reactor temperature profiles in classic jacketed tubular reactors often exhibit a (nearly) trapezoidal shape along the reactor, i.e., in a first part the temperature is raised, then a constant temperature interval follows, and (possibly) the temperature is lowered towards the outlet. The practical realisation of these optimal reactor temperature profiles is, however, complex because of the spatially varying jacket fluid temperature profile that is required for the constant reactor temperature part. Since similar trapezoidal temperature profiles are encountered in reverse flow configurations, this reactor type may be an alternative to implement the optimal profiles in practice. This article conceptually proves the feasibility of this concept and provides an optimisation procedure for its design and operation.

Key words: chemical reactors, design, dynamic simulation, optimisation, process intensification, reverse flow reactor

1 Introduction

Since tubular reactors are still important workhorses in chemical process industry, optimising the design and operation of these reactors proves to be highly profitable. For classic jacketed tubular reactors where an exothermic first-order reaction takes place, optimal steady-state temperature profiles have

* Corresponding author: Tel.: +32-16-32.14.66 Fax: +32-16-32.29.91
Email address: jan.vanimpe@cit.kuleuven.be (J.F. Van Impe).

been developed by, e.g., Smets et al. (2002) and Logist et al. (2006). These optimal temperature profiles often exhibit a (nearly) trapezoidal shape, i.e., increasing until a certain temperature is reached, keeping that temperature constant over an interval, and (possibly) decreasing the temperature towards the end. However, inducing the constant temperature part through the surrounding reactor jacket, is often difficult for an exothermic reaction as a spatially varying jacket fluid temperature profile is required. Similar trapezoidal shapes are also encountered in tubular reactors operating under a periodic flow reversal regime, also called *reverse flow reactors* (RFRs). Hence, the objective is to investigate numerically the feasibility of realising optimal temperature profiles for a jacketed tubular reactor by implementing a periodic flow reversal strategy. This analysis shows that the design concept of flow reversal, initially developed so as to recover the heat released by the chemical reaction, is in fact a practical way to implement temperature profiles derived rigorously using optimal control theory. The organisation of this paper is as follows. Section 2 introduces the RFR. Section 3 proves the feasibility of the proposed approach, while in Section 4 a procedure for the optimal design and operation of RFRs is elaborated. Section 5 summarises the main conclusions.

2 Reverse flow reactor

Although the RFR has a long history of periodic reinventions, the recent review by Kolios et al. (2000) gives the credit to Boreskov, Matros and their co-workers, who in the 80's elaborated its scientific understanding, demonstrated its potential up to the industrial scale and communicated its principle all around the world (see, e.g., Matros (1989)). Based on their pioneering work a number of applications have been investigated by other researchers, e.g., (i) the oxidation of SO_2 (e.g., Hong et al. (1997); Xiao et al. (1999)), (ii) the treatment of waste air (e.g., Eigenberger and Nieken (1988); Sapundzhiev et al. (1993); Chaouki et al. (1994); van de Beld et al. (1994); van de Beld and Westerterp (1996, 1997); Züfle and Turek (1997a,b); Nijdam and van der Geld (1997, 1999); Cittadini et al. (2001); Salomons et al. (2004); Kushwaha et al. (2004)), (iii) the synthesis of methanol (e.g., Vanden Bussche et al. (1993)) and (iv) the oxidation of o-xylene for the production of phthalic anhydride (Quinta Ferreira et al., 1999). For additional information on this reactor concept the reader is referred to the review by Kolios et al. (2000).

The classic RFR example of Eigenberger and Nieken (1988), which concerns a combustion reaction of traces of organic compounds in air in a ceramic monolith catalyst, is selected as base for the current case study, because it uses the same model type (i.e., pseudo-homogeneous) and kinetic scheme (i.e., an exothermic, irreversible first-order reaction) as in the papers where the optimal trapezoidal temperature profiles have been derived, i.e., Smets et al. (2002)

and Logist et al. (2006). Consequently, an easy model transformation and a fair comparison between the classic tubular reactor concept and the RFR concept, is ensured. However, to complete the model equivalence, a surrounding cooling jacket is added to the example of Eigenberger and Niesen (1988), allowing to control the reactor temperature, which is often required for safety reasons or for the prevention of catalyst deactivation or product degradation (Khinast et al., 1998). This jacket covers the middle part of the reactor, while the inlet and outlet zones are still insulated. The temperature of the fluid inside the jacket is assumed to be constant along the jacket. Formulating the microscopic reactor, mass and energy balances mathematically results in the following set of coupled nonlinear parabolic partial differential equations (PDEs) for $t \geq 0$ and $0 \leq z \leq L$:

$$\begin{aligned} \frac{\partial C}{\partial t} &= D \frac{\partial^2 C}{\partial z^2} - v \frac{\partial C}{\partial z} - k C e^{-E/RT} \\ \overline{\rho c_p} \frac{\partial T}{\partial t} &= \lambda_{ax,eff} \frac{\partial^2 T}{\partial z^2} - \rho_g c_{pg} v \varepsilon \frac{\partial T}{\partial z} + \varepsilon k C e^{-E/RT} (-\Delta H) - \frac{4h}{d} (T - T_w) \end{aligned} \quad (1)$$

where C [kmole/m³] is the reactant concentration, T [K] the temperature, D [m²/s] the dispersion coefficient, v [m/s] the gas velocity (positive for forward flow and negative for backward flow), k [1/s] the rate constant, E/R [K] the activation temperature, $\overline{\rho c_p}$ [kJ/m³K] the fixed bed heat capacity, $\lambda_{ax,eff}$ [kW/mK] the effective axial heat conductivity, $\rho_g c_{pg}$ [kJ/m³K] the heat capacity of the gas, ε [-] the void fraction, $-\Delta H$ [kJ/kmole] the heat of reaction, L [m] the reactor length, T_w [K] the temperature of the jacket fluid, d [m] the reactor diameter and h [kW/m²K] the (global) heat transfer coefficient. The value of this heat transfer coefficient is constant and positive in the jacketed zone, i.e., $[L/2 - L_j/2, L/2 + L_j/2]$ with L_j [m] the jacket length, and zero in the insulated zones. In appendix, the appropriateness of the pseudo-homogeneous approach for the case under study is substantiated by comparison with a heterogeneous model, which accounts for the difference between the gas and solid phase.

Reversing the flow direction periodically (with a switching time τ [s]) induces each time a temperature front that slowly moves downstream. The aim is to achieve a *cyclic steady-state* (CSS) regime in which the successive reversion cycles (after 2τ) become repeatable. This CSS can be computed by *direct dynamic simulation* (DDS), i.e., calculating the full transient from an initial state to the CSS, which is straightforward but may require a large computation time. To avoid the calculation of the entire transient, the problem can be reformulated as a boundary value problem and solved using *iterative shooting methods* (Gupta and Bhatia, 1991; Salinger and Eigenberger, 1996a,b; Unger et al., 1997). Despite the lower computation time, these methods are more complex. Additionally, DDS directly yields the entire start-up behaviour, which is useful for the operators. Thus, DDS is adopted using a *method of lines*

approach for the solution of the PDEs: first, the spatial derivatives are approximated by finite differences, and then the system of semi-discrete equations is integrated in time. The program by Vande Wouwer et al. (2005), which uses the MatMOL toolbox (Vande Wouwer et al., 2004), is adopted as a starting point. To check whether a CSS is attained, two measures are proposed (Gosiewski, 2004):

$$I_1 = \frac{1}{N} \sum_{i=1}^N |T_j^{start}(z_i) - T_{j-1}^{start}(L - z_i)| \quad (3)$$

$$I_2 = \frac{1}{N} \sum_{i=1}^N |T_j^{start}(z_i) - T_{j-2}^{start}(z_i)| \quad (4)$$

with N the number of discretisation points z_i and T_j^{start} the initial temperature profile after the j -th flow reversal. Measure I_1 assumes that successive half cycles are mirror copies, whereas I_2 assumes that repeatability only occurs for full cycles. To ensure that a symmetric CSS is attained, both measures have to be lower than the integration tolerance (Gosiewski, 2004).

3 Reverse flow implementation of optimal temperature profiles

In this section the feasibility of realising optimal steady-state temperature profiles for classic jacketed tubular reactors based on a periodic flow reversal strategy is studied numerically. First, for the classic jacketed tubular reactor the performance measures and the resulting optimal steady-state temperature profiles derived by Smets et al. (2002) and Logist et al. (2006) are introduced, and equivalent measures for the RFR are defined. Second, similar temperature profiles are induced in a RFR and the parameters influencing these profiles and the performance measures are identified through a sensitivity analysis.

The simulation parameters are summarised in Table 1 and comprise the ones from Eigenberger and Nieken (1988) for an adiabatic RFR, and additional heat transfer related parameters, i.e., the (global) heat transfer coefficient h , the jacket fluid temperature T_w and the jacket length L_j . A similar simulation procedure as in Eigenberger and Nieken (1988) is employed. After 1500 s, during which the gas enters at high temperature (873 K), the feed temperature is decreased to 293 K and periodic flow reversals every 100 s are started. The simulated time is, however, in this study prolonged from 9500 s (80 switchings or 40 cycles) to 21500 s (200 flow reversals or 100 cycles) in order to ensure a CSS is attained. As the jacket is an integral part of the reactor, it is assumed to be already active during the first 1500 s.

3.1 Performance measures and optimal temperature profiles

Two cost criteria are presented, according to the ones proposed in Smets et al. (2002) and Logist et al. (2006), i.e., a *terminal cost criterion* J_{TC} (Equation (5)) and a *combined terminal and integral cost criterion* J_{TIC} (Equation (6)). These criteria both consist of a trade-off between a conversion and an energy cost part, reflecting the current trend in process industry towards process integration and intensification, which relates to enhancing the over all plant performance by (i) optimising the performance of existing reactors or by (ii) adopting novel reactor types, which combine several unit operations into one, and by (iii) interconnecting material and energy streams plantwide.

$$J_{TC} = (1 - A) \underbrace{C(L)}_{J_1} + A \underbrace{\frac{1}{K_1} (T(L) - T_{ref,1})^2}_{J_2} \quad (5)$$

$$J_{TIC} = (1 - A) \underbrace{C(L)}_{J_1} + A \underbrace{\frac{1}{K_2} \int_0^L (T(z) - T_{ref,2})^2 dz}_{J_3} \quad (6)$$

where A [-] is a trade-off coefficient ranging between zero and one, and K_1 [$\text{K}^2 \text{m}^3/\text{kmole}$] and K_2 [$\text{K}^2 \text{m}^3/\text{kmole}$] are scaling factors to bring both cost terms in the same order of magnitude. By varying A , more or less emphasis can be put on either of the cost terms.

The conversion cost J_1 measures the (unconverted) reactant concentration at the reactor outlet ($z = L$). The energy cost J_2 penalises both positive and negative deviations of the exit temperature $T(L)$ from a reference temperature $T_{ref,1}$ [K] in order to limit the terminal heat/energy loss to a certain (acceptable) off-set value, proportional to $(T_{ref,1} - T_{in})$. When combining the macroscopic mass and heat balances for a classic jacketed tubular reactor

$$0 = v(C_{in} - C(L)) \frac{\pi d^2}{4} - \text{reaction} \frac{\pi d^2}{4} L \quad (7)$$

$$0 = v\rho_g c_{pg}(T_{in} - T(L)) \frac{\pi d^2}{4} + \text{reaction} \frac{\pi d^2}{4} L(-\Delta H) - \int_0^L \pi dh(T(z) - T_w(z)) dz \quad (8)$$

into Equation (9), clarifies the meaning of the terminal energy cost term J_2 .

$$v\rho_g c_{pg}(T(L) - T_{ref,1}) = \underbrace{v(-\Delta H)(C_{in} - C(L))}_{\text{heat generated through reaction}} - \underbrace{\int_0^L \frac{4h}{d}(T(z) - T_w(z))dz}_{\text{heat recuperated via jacket}} - \underbrace{v\rho_g c_{pg}(T_{ref,1} - T_{in})}_{\text{acceptable heat loss}} \quad (9)$$

$T(L) - T_{ref,1} = \sqrt{J_2}$ corresponds to the amount of energy generated by the reaction that has not been recuperated by the jacket (assuming this energy can be reused elsewhere), nor has been considered as acceptable heat loss. Consequently, decreasing J_2 towards zero also reduces the heat loss to the off-set value considered acceptable. The energy cost J_3 penalises temperature deviations from a reference temperature $T_{ref,2}$ along the reactor to minimise the risk of excessive temperatures or hot spots, which may cause undesired effects (catalyst deactivation, product degradation or side-reactions) and/or hazardous situations. However, apart from this integral cost which implicitly limits the temperature (i.e., a *soft* constraint), an upper bound $T(z) \leq T_{ul}$ can be imposed to explicitly limit the reactor temperature (i.e., a *hard* constraint).

Based on the results derived by Smets et al. (2002) and Logist et al. (2006) by exploiting optimal control theory, an optimal temperature profile for the first criterion J_{TC} often exhibits a trapezoidal shape, i.e., the temperature is first increased until the upper temperature limit T_{ul} is attained which then has to be kept constant over a certain interval, and finally it is lowered again towards the outlet to attain $T_{ref,1}$. The first two parts are driven by a minimisation of the conversion cost, whereas the last part is used to minimise the energy cost. For the second cost criterion J_{TIC} , similar optimal temperature profiles are often encountered, e.g., increasing the temperature to an intermediate value T_{int} which has to be maintained over an interval. Possibly, a decrease towards the outlet is observed. The initial increase favours conversion, while the constant intermediate temperature is driven by a minimisation of the risk of hot spots. The optimal temperature profiles and the required spatially varying jacket fluid temperature profiles are depicted schematically in Figure 1.

To compare several reverse flow implementations, equivalent cost criteria J'_{TC} and J'_{TIC} have to be defined. Due to the transient effects within one cycle an averaged value is computed. Note that the reactor outlet also switches due to the flow reversals (i.e., $z = L$ for forward and $z = 0$ for backward flow):

$$J'_{TC} = (1 - A) \underbrace{\frac{\int_0^{2\tau} C_{out}(t) dt}{2\tau}}_{J'_1} + A \underbrace{\frac{1}{K_1} \left(\frac{\int_0^{2\tau} (T_{out}(t) - T_{ref,1}) dt}{2\tau} \right)^2}_{J'_2} \quad (10)$$

$$J'_{TIC} = (1 - A) \underbrace{\frac{\int_0^{2\tau} C_{out}(t) dt}{2\tau}}_{J'_1} + A \underbrace{\frac{1}{K_2} \frac{\int_0^{2\tau} \int_0^L (T(z, t) - T_{ref,2})^2 dz dt}{2\tau}}_{J'_3} \quad (11)$$

Term J'_1 represents the time averaged conversion cost. The use of the macroscopic mass and heat balances over an entire cycle leads to Equation (12) and enables an equivalent interpretation of the terminal energy cost term J'_2 .

$$\begin{aligned} v\rho_g c_{pg} \frac{\int_0^{2\tau} (T_{out}(t) - T_{ref,1}) dt}{2\tau} &= \underbrace{\frac{\int_0^{2\tau} v(-\Delta H)(C_{in} - C_{out}(t)) dt}{2\tau}}_{\text{heat generated through reaction}} \\ &- \underbrace{\frac{\int_0^{2\tau} \int_0^L \frac{4h}{d} (T(z, t) - T_w) dz dt}{2\tau}}_{\text{heat recuperated via jacket}} \\ &- \underbrace{v\rho_g c_{pg} \frac{\int_0^{2\tau} (T_{ref,1} - T_{in}) dt}{2\tau}}_{\text{acceptable heat loss}} \end{aligned} \quad (12)$$

Again forcing the left hand side term towards zero, yields a heat recuperation equal to the generated heat minus a certain (acceptable) off-set or heat loss. Term J'_3 indicates the time averaged value of deviations from the reference temperature $T_{ref,2}$ along the reactor over the entire cycle.

Most of the cost parameters are user-defined and, hence, have to be adapted for a specific process according to information about the process itself, the prices of resources, energy and products, etc. The procedures illustrated in the following sections are, however, generally applicable. In the following simulation studies, A is taken equal to 0.5 (i.e., an equal weighting of conversion and energy costs), K_1 and K_2 to $5.0 \cdot 10^8 \text{ K}^2 \text{ m}^3/\text{kmole}$ (to bring both cost terms in the same order of magnitude), and $T_{ref,1}$ and $T_{ref,2}$ to 440 K and 700 K, respectively. The upper limit for the reactor temperature T_{ul} is assumed to be 900 K. As mentioned before, $T_{ref,1}$ concerns the allowable amount of energy lost. The temperature $T_{ref,2}$ relates to an appropriate penalisation of high temperatures in order not to exceed the upper temperature limit T_{ul} .

3.2 Sensitivity analysis of a jacketed RFR

In this subsection numerical simulations illustrate that trapezoidal temperature profiles with a controllable constant part can be induced in a jacketed RFR. Additionally, a sensitivity analysis reveals the influence of several design and control variables on (i) the temperature and concentration CSS profiles inside the reactor and at the reactor outlet and (ii) the different cost terms J'_1 , J'_2 and J'_3 . Hereby, the variables which are specific to the jacketed RFR are mainly focussed on, i.e., the heat transfer coefficient h , the jacket fluid temperature T_w , the jacket length L_j and the switching time τ . Although other variables have been investigated, e.g., the feed temperature T_{in} , the feed concentration C_{in} and the gas velocity v , their results are omitted, as they are similar to previous sensitivity studies for the *adiabatic* RFR (e.g., Eigenberger and Niekens (1988); van de Beld et al. (1994); Züfle and Turek (1997a); Wu et al. (1999)).

Switching time τ . Values of 100, 200, 300, 500, 750 and 1000 s are selected for the switching time τ . As depicted in Figure 2, there is a maximum switching time (of about 500 s) to have an ignited reactor. For longer switching times the maximum reactor temperature is lower than the jacket fluid temperature, meaning that the jacket is now used for heating the reactor instead of cooling it, which is of course not the intended operation regime. Whenever an ignited steady-state is obtained, the maximum temperature hardly depends on the switching time, whereas the conversion slowly decreases. The outlet temperature $T_{out}(t)$ remains almost constant for half the cycle period and shows an almost linear rise afterwards. The outlet concentration $C_{out}(t)$ attains a constant value almost immediately after a flow reversal, due to the small gas residence time. For all ignited operation regimes, the conversion cost J'_1 increases with increasing switching times due to lower averaged temperatures, the energy cost J'_2 exhibits a minimum, whereas a monotonous increase is observed for the energy cost J'_3 .

The strong dependence of the energy cost J'_2 can be explained analytically, assuming the RFR is ignited and the temperature profile within one cycle can be approximated by a symmetric trapezoidal shaped profile moving forward during the first half-cycle and backward during the second half-cycle. A linear increase in the outlet temperature after an initial delay of half the switching time, results in the following approximation:

$$T_{out}(t) = \begin{cases} T_{in} & \text{for } 0 \leq t \leq \frac{\tau}{2} \\ T_{in} + bt & \text{for } \frac{\tau}{2} \leq t \leq \tau \text{ with } b = \omega \cdot \left. \frac{dT}{dz} \right|_{out}. \end{cases}$$

Calculating the full-cycle cost yields the following second-order polynomial:

$$J'_2(2\tau) = \left(\frac{1}{\tau} \left[\int_0^{\frac{\tau}{2}} (T_{out}(t) - T_{ref,1}) dt + \int_{\frac{\tau}{2}}^{\tau} (T_{out}(t) - T_{ref,1}) dt \right] \right)^2 \\ = \left((T_{in} - T_{ref,1}) + \frac{b}{8}\tau \right)^2.$$

Its minimum of zero is reached for $\tau = 8 \frac{T_{ref,1} - T_{in}}{b}$, which may be an appropriate first guess of the optimal switching time.

Heat transfer coefficient h . The selected (reduced) heat transfer coefficients $\beta = \frac{4h}{\rho c_p d}$ are 0, 2.5, 5, 12.5 and $25 \cdot 10^{-4}$ 1/s. As can be seen in Figure 3, increasing the heat transfer coefficient causes a significant decrease in the maximum temperature and the conversion. For the conversion cost J'_1 , lower heat transfer coefficients are preferred because they induce higher reactor temperatures. This situation is also desirable from the energy cost J'_3 point of view as the temperature profile along the reactor deviates less from the reference temperature $T_{ref,2}$. Because the outlet temperature is hardly affected, the energy cost J'_2 remains virtually unaltered.

Jacket fluid temperature T_w . The jacket fluid temperature range (from 700 to 900 K) is divided into equal intervals of 50 K. From Figure 4, it is clear that decreasing the jacket fluid temperature causes a decrease not only in the maximum reactor temperature (which ranges from 710 to 950 K) but also in the slope at the outlet. This feature makes the jacket fluid temperature an excellent parameter to control and limit the temperature inside the reactor. For the conversion cost J'_1 , higher jacket fluid temperatures, resulting in higher reactor temperatures and higher reactant conversion levels, are favoured. In contrast with the energy cost J'_2 , which only slightly decreases due to the limited effect of the jacket fluid temperature on the outlet temperatures, the energy cost J'_3 exhibits a shallow minimum as both (too) low and (too) high temperature profiles inside the reactor are penalised.

Jacket length L_j . Six jacket lengths are studied: from the adiabatic case with no jacket at all, i.e., L_j equals 0 m, in steps of 0.2 m, to L_j equals 1 m, which covers the entire reactor. However, as can be observed in Figure 5, the jacket has to be long enough in order to avoid asymmetric temperature profiles (indicated by a high I_1 value), e.g., the one encountered for a jacket length of 0.2 m. As a matter of fact, the jacket has to cover the zone of flow reversal. Once the minimal length has been exceeded (i.e., $L_j \geq 0.4$ m), symmetric profiles are obtained and the maximum temperature is hardly affected by the jacket length. Due the additional heating near the reactor inlet and

outlet, the constant temperature zone expands and the temperature gradients become sharper for increasing jacket lengths. Due to the extended high temperature plateau the conversion cost J'_1 decreases. In addition, lower values are also observed for both energy costs. The decrease in energy cost J'_3 is easily explained by the fact that the extended high temperature plateau induces less deviations from the desired reference temperature $T_{ref,2}$, whereas the decrease in energy cost J'_2 is related to the sharper gradients at the outlet causing the optimal switching time to shift towards lower values, i.e., closer to the here used fixed value of 100 s.

In summary, the jacketed RFR gives rise to trapezoidal temperature profiles with a controllable maximum temperature. Consequently, this reactor configuration may be an adequate alternative to implement the optimal temperature profiles obtained by Smets et al. (2002) and Logist et al. (2006) for a classic jacketed tubular reactor in practice, without the need for a spatially varying jacket fluid temperature profile. The most important variables which influence this maximum temperature value are the heat transfer coefficient and the jacket fluid temperature. From a cost point of view, the most important parameters to be determined in practice are the jacket temperature (i.e., to minimise mainly the conversion cost J'_1 and the energy cost J'_3) and the switching time and jacket length (i.e., to minimise mainly the energy cost J'_2).

It should also be noted that complex dynamic phenomena (e.g., asymmetric and/or aperiodic solutions) are hardly observed. Solely for a short jacket length an asymmetric solution is found. The existence of such complex behaviour in cooled RFRs is, however, reported by Khinast et al. (1998, 1999). Although the same reaction kinetics and similar model types are employed in the current study, complex behaviours are not evidenced here, probably due to the fact that the switching times are generally (much) longer than the ones for which complex solutions have been found by Khinast et al. (1998).

4 Optimisation of a jacketed RFR

In this section a procedure for an optimal design and operation of a jacketed RFR is elaborated and illustrated. Assuming the inlet concentration C_{in} , the gas velocity v and the heat transfer coefficient h are process specifications, the only variables left to be determined based on a minimisation of the cost criteria (Equations (10) and (11)) while taking an upper temperature limit inside the reactor into account $T(z) \leq 900$ K, are the jacket fluid temperature T_w , the switching time τ and the jacket length L_j . (This choice is, however, not restrictive as the other parameters can equally be added as degrees of freedom.) Using the previous sensitivity results allows to impose simple bounds on all optimisation variables. For the switching time a range from 100 to 500 s

is adopted. For lower values the number of flow reversals to reach the CSS becomes too high to be feasible in practice and complex dynamic phenomena may arise, whereas for higher values the risk of extinction exists. The jacket length has to exceed 0.4 m in order to ensure a symmetric solution and maximally equals the reactor length of 1 m. As the jacket fluid temperature has the largest influence on the maximum reactor temperature, bounds on the jacket fluid temperature can be selected depending on the constant temperature level envisioned. For instance, based on Figure 6, a range of jacket fluid temperatures T_w from 800 to 850 K can be used to obtain maximum reactor temperatures T_{max} of about 875 to 900 K.

The abovementioned optimisation problem encompasses, however, a discrete decision variable L_j due to the spatial discretisation. Therefore, the following optimisation strategy is proposed, which combines discrete grid search and continuous optimisation procedures. For a grid of jacket length values, continuous optimisations inside the selected ranges are performed for the switching time and the jacket fluid temperature. To solve each time these constrained nonlinear programming problems the Matlab routine `fmincon` is employed. For each cost function and constraint evaluation, direct dynamic simulation is adopted. Termination tolerances on the decision variables are specified as stop criteria for `fmincon`. These tolerances are set in relation to the length of the admissible interval, i.e., 0.001 and 0.01 of the interval length for the switching time τ and the jacket fluid temperature T_w , respectively. For the cases under study, these values give rise to a maximal deviation of 0.5 s and 0.5 K, respectively. Since it is hard to apply more accurate values in practice, lowering these tolerances is useless and only results in an increased computation time. The optimal set of parameters is finally determined by selecting the lowest cost value over all jacket lengths in the grid.

4.1 Terminal cost criterion J'_{TC}

For a grid of jacket lengths, i.e., from 0.4 to 1 m in steps of 0.1 m, continuous optimisations are performed with the jacket fluid temperature and the switching time as decision variables. As already mentioned, the admissible intervals for these variables range from 800 to 850 K (since reactor temperatures close to 900 K are envisioned) and from 100 to 500 s, respectively. In Figure 7, the evolution of the terminal cost J'_{TC} and its two constituting parts is displayed. As can be seen, the lowest cost value ($J'^*_{TC} = 6.8389 \cdot 10^{-6}$) is obtained for a jacket length of 1 m, which means that the jacket covers the entire length of the reactor. The corresponding optimal values for the jacket fluid temperature T_w^* and the switching time τ^* are 846.9 K and 169.1 s, respectively. The resulting optimal CSS profiles are depicted in Figure 8: a trapezoidal shaped temperature profile which does not violate the temperature constraint, is ob-

tained. In addition, it can be checked that during the entire transient phase towards the CSS the constraint is never violated.

4.2 Combined terminal and integral cost criterion J'_{TIC}

Assuming a desired intermediate reactor temperature level of about 875 K, a range of admissible jacket fluid temperatures for the continuous optimisation is easily selected from Figure 6, i.e., from 800 to 850 K. 100 and 500 s are again selected as lower and upper bounds for the switching time and the grid of jacket lengths also remains unchanged. The evolution of the different cost values is depicted in Figure 9 for increasing jacket lengths. Again a decrease in the total cost is observed for increasing jacket lengths and a jacket length L_j of 1 m yields the lowest cost value ($J'^*_{TIC} = 0.3978 \cdot 10^{-4}$). The optimal CSS profiles, obtained with the corresponding optimal values $T_w^* = 821.4$ K and $\tau^* = 100$ s, are illustrated in Figure 10. It should be noted that the maximum temperature constraint of 900 K is not attained in this case due to the *soft constraint*, i.e., energy cost J'_3 . Consequently, constraint violation never occurs. The conversion cost J'_1 is, however, somewhat higher than for the pure terminal cost criterion J'_{TC} due to these slightly lower temperature profiles. The resemblance between the optimal temperature profiles for the RFR and for the classic tubular reactor is again remarkable.

5 Conclusions

In this paper the feasibility of realising optimal temperature profiles for tubular reactors through the use of a flow reversal strategy, is investigated through a profound simulation study. It is shown that (nearly) trapezoidal shaped temperature profiles can be obtained in a jacketed RFR, similar to the optimal profiles derived by Smets et al. (2002) and Logist et al. (2006) using optimal control theory for steady-state classic jacketed tubular reactors. Additionally, the most important design and control variables are identified through a sensitivity analysis. Based on the results of this analysis a procedure for optimising the RFR design is proposed and illustrated. The resemblance between the resulting optimal temperature profiles for the jacketed reverse flow and the classic jacketed tubular reactor is remarkable. Although the reverse flow implementation exhibits inherent transient characteristics, the global process is simplified with regard to the steady-state classic jacketed tubular reactor, because spatially varying jacket fluid temperature profiles are no longer required. This concept may open the door to a new area of RFR applications.

Future work may evolve in several directions. First, based on the here pre-

sented proof of feasibility, both the classic jacketed tubular and the jacketed RFR configuration can be compared quantitatively for the case studied by Smets et al. (2002) and Logist et al. (2006). Second, a further validation can be performed through the use of more rigorous and complex models (e.g., 2D and/or heterogeneous) and/or through an experimental set-up.

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Parameter	Value	Units
Parameters from Eigenberger and Niekem (1988) for an adiabatic RFR		
C_{in}	$= 1.21 \cdot 10^{-4}$	kmole/m ³
D	$= 5 \cdot 10^{-3}$	m ² /s
E/R	$= 8000$	K
k	$= 29732$	1/s
L	$= 1$	m
T_{in}	$= 293$	K
v	$= 1$	m/s
ε	$= 0.8$	[-]
$-\Delta H$	$= 206000$	kJ/kmole
$\lambda_{ax,eff}$	$= 2.06 \cdot 10^{-3}$	kW/mK
$\overline{\rho c_p}$	$= 400$	kJ/m ³ K
$\rho_g c_{pg}$	$= 0.5$	kJ/m ³ K
τ	$= 100$	s
Heat transfer related parameters for a jacketed RFR		
T_w	$= 800$	K
L_j	$= 0.6$	m
$\beta = \frac{4h}{\overline{\rho c_p} d}$	$= 12.5 \cdot 10^{-4} \text{ 1/s}$	1/s

Table 1

Parameter values used in the simulations. (Details about the evaluation of the heat transfer coefficient are given in Appendix A).

Fig. 1. Schematic view of the optimal jacket fluid temperature and reactor temperature profiles for a classic jacketed tubular reactor under steady-state conditions: terminal cost criterion J_{TC} (left) and combined terminal and integral cost criterion J_{TIC} (right).

Fig. 2. CSS temperature and concentration profiles (top), outlet temperature and concentration profiles (middle) and cost values (bottom) for different switching times: $\tau = 100$ s (x), 200 s ($\square$), 300 s ($\diamond$), 500 s (o), 750 s ($\triangle$) and 1000 s (∇).

Fig. 3. CSS temperature and concentration profiles (top), outlet temperature and concentration profiles (middle) and cost values (bottom) for different heat transfer coefficients: $\beta = 0 \text{ s}^{-1}$ (x), $2.5 \cdot 10^{-4} \text{ s}^{-1}$ ($\square$), $5 \cdot 10^{-4} \text{ s}^{-1}$ ($\diamond$), $12.5 \cdot 10^{-4} \text{ s}^{-1}$ (o) and $25 \cdot 10^{-4} \text{ s}^{-1}$ ($\triangle$).

Fig. 4. CSS temperature and concentration profiles (top), outlet temperature and concentration profiles (middle) and cost values (bottom) for different jacket fluid temperatures: $T_w = 700$ K (x), 750 K ($\square$), 800 K ($\diamond$), 850 K (o) and 900 K ($\triangle$).

Fig. 5. CSS temperature and concentration profiles (top), outlet temperature and concentration profiles (middle) and cost values (bottom) for different jacket lengths: $L_j = 0$ m (x), 0.2 m ($\square$), 0.4 m ($\diamond$), 0.6 m (o), 0.8 m ($\triangle$) and 1.0 m (∇).

Fig. 6. Evolution of the maximum reactor temperature as a function of the jacket fluid temperature.

Fig. 7. Evolution the optimised terminal cost values as a function of the jacket length.

Fig. 8. Optimal jacketed RFR for the terminal cost criterion J'_{TC} : cyclic steady-state profiles after 80 flow reversals with $\tau^* = 169.1$ s and $T_w^* = 846.9$ K: concentration (top) and temperature (bottom).

Fig. 9. Evolution the optimised combined terminal and integral cost values as a function of the jacket length.

Fig. 10. Optimal jacketed RFR for the combined terminal and integral cost criterion J'_{TIC} : cyclic steady-state profiles after 80 flow reversals with $\tau^* = 100$ s and $T_w^* = 821.4$ K: concentration (top) and temperature (bottom).

A Validation of the 1D pseudo-homogeneous jacketed RFR model

The study of Eigenberger and Nieken (1988) on an adiabatic RFR contained both simulation results (for a 1D pseudo-homogeneous model) and experimental results. The simulation values *referred to a combustion reaction for traces of organic compounds in air in a ceramic monolith catalyst*. For the experiments these authors used a commercial ceramic monolith for exhaust purification. The square channels of the monolith were 1 mm wide with a wall thickness of 0.2 mm (resulting in a void fraction of $\varepsilon = 0.8$) and contained a wash coat with precious metals as catalyst.

In this appendix the validity and real-life value of the current jacketed RFR simulations is substantiated. Hereto, (i) the selection of the global heat transfer coefficient value is justified and (ii) a comparison is made between the pseudo-homogeneous model and a corresponding heterogeneous model (see Khinast et al. (1999) for the model equations) accounting for gas-solid transport processes. Values for these transfer properties are estimated based on correlations and design information of the experimental set-up of Eigenberger and Nieken (1988). To estimate the physical properties, a cordierite solid phase

(Lepperhoff et al., 2000) with a heat conductivity of 1 W/mK and a gas phase (mainly consisting of air) at 900 K and 1 bar are assumed.

For the *radial heat transfer*, a classic *heat transfer resistances model* (see Dixon (1996) for an overview) is used to capture all 2D heat transfer effects into the sole heat transfer related 1D variable, i.e., the heat transfer coefficient h . Assuming perfect thermal contact between the catalyst matrix and the reactor wall as in Groppi and Tronconi (2000), results in a negligible resistance at the reactor wall. Hence, only the resistance inside the reactor remains, leading to $\frac{1}{h} = \frac{d}{8\lambda_{rad,eff}}$ with d [m] the reactor tube diameter and $\lambda_{rad,eff}$ [kW/mK] the effective radial heat conductivity. An estimate of $\lambda_{rad,eff}$ can be obtained for the monolith using: $\lambda_{rad,eff} = G \cdot \lambda_s$ (Groppi and Tronconi, 2000) with G [-] a correction factor based on the monolith's void fraction ε , e.g., $G = 0.10$ for $\varepsilon = 0.8$. This leads together with the parameters of Eigenberger and Nieken (1988) to the following relation for the (reduced) heat transfer coefficient: $\beta = \frac{4h}{\overline{\rho c_p} d} = \frac{32G\lambda_s}{\overline{\rho c_p} d^2}$ with the reactor diameter d as the only unknown. Figure A.1 displays the (reduced) heat transfer coefficient β as a function of the reactor tube diameter. For β values ranging between $2.5 \cdot 10^{-4}$ and $25 \cdot 10^{-4}$ 1/s, the reactor diameter varies between 0.19 and 0.06 m, which are suitable values for industrial implementations. In addition, conversion of the heat transfer coefficient values Δ used in Khinast et al. (1998) into corresponding β values yields a similar range: $\beta \in [0; 18 \cdot 10^{-4}]$ 1/s.

For the *gas-solid transfer* properties of the *heterogeneous model*, the heat and mass gas solid transfer coefficients h_{gs} [W/m²K] and k_{gs} [m/s] are estimated based on the following semi-theoretical correlations for the Nusselt number Nu and the Sherwood number Sh (Groppi and Tronconi, 2000)

$$Sh = 2.977 + 6.874 \left(\frac{1000}{Gr_m} \right)^{-0.488} \exp \left(-\frac{57.2}{Gr_m} \right) \quad (A.1)$$

$$Nu = 2.977 + 6.874 \left(\frac{1000}{Gr_t} \right)^{-0.488} \exp \left(-\frac{57.2}{Gr_t} \right) \quad (A.2)$$

with Gr_m and Gr_t the mass and heat Graetz number. The specific monolith surface to reactor volume ratio a_v [m²_{monolith}/m³_{reactor}] is given by $a_v = 4\varepsilon/d_h$ with d_h the hydraulic channel diameter. With the design specifications of the set-up of Eigenberger and Nieken (1988) the calculations yield for h_{gs} , k_{gs} and a_v , values of 154 W/m²K, 0.37 m/s and 2666 m²_{monolith}/m³_{reactor}, respectively, which are in the same order as the parameters used by Khinast et al. (1998).

Simulated CSS temperature and concentration profiles based on the heterogeneous model of the experimental set-up are depicted in Figure A.2. Clearly, hardly any difference between the solid and gas phase is observed, justifying

the homogeneous approach. When comparing the simulated CSS profiles for the set-up (i.e., from the heterogeneous model) and for the simulation values of Eigenberger and Niekens (1988) (i.e., the pseudo-homogeneous model), it is observed that the maximum temperature and velocity of the temperature front are similar. However, steeper temperature and, hence, lower concentration profiles are found for the simulated experimental set-up. This difference can be attributed to a different effective axial heat dispersion $\lambda_{ax,eff}$, i.e., $0.65 \cdot 10^{-3}$ W/mK for the simulated set-up according to the correlation of Vortmeyer and Schaeffer (1974): $\lambda_{ax,eff} = (1 - \varepsilon)\lambda_s + \lambda_g + \frac{(\rho_g c_{pg} \varepsilon v)^2}{h_{gs} a_v}$, and $2.06 \cdot 10^{-3}$ W/mK used by Eigenberger and Niekens (1988) in their simulations. Increasing the channel width and wall thickness to 2.2 mm and 0.4 mm, results in $h_{gs} = 71$ W/m²K, $k_{gs} = 0.17$ m/s, $a_v = 1231$ m²_{monolith}/m³_{reactor} and $\lambda_{ax,eff} = 2.09 \cdot 10^{-3}$ W/mK. The corresponding heterogeneous model CSS profiles now indeed agree with the homogeneous model simulations, indicating the validity of the simulation parameters used. In practice, of course, the inverse road has to be followed: a heterogeneous model may be the starting point and should, if possible, be reduced to a homogeneous model by tuning the effective axial dispersion $\lambda_{ax,eff}$. In addition, it should be noted that for the case under study the same stable behaviour is predicted by both the pseudo-homogeneous and the heterogeneous model, despite the large difference between the inlet temperature and the jacket fluid temperature (Khinast et al., 1999).

Fig. A.1. Figure A.1. (Reduced) heat transfer coefficient β as a function of the reactor diameter.

Fig. A.2. Figure A.2. CSS temperature and concentration profiles for different models: pseudo-homogeneous model using the simulation values of Eigenberger and Nieken (1988) ($\square$), heterogeneous model for the experimental set-up of Eigenberger and Nieken (1988) (∇ solid, $+$ gas) and heterogeneous model after tuning of the experimental set-up of Eigenberger and Nieken (1988) (o solid, x gas).

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Notation

a_v	specific monolith surface to reactor volume ratio	$[\text{m}_{monolith}^2 \cdot \text{m}_{reactor}^{-3}]$
A	trade-off coefficient	$[-]$
C	reactant concentration	$[\text{kmole} \cdot \text{m}^{-3}]$
d	reactor diameter	$[\text{m}]$
d_h	hydraulic diameter	$[\text{m}]$
D	dispersion coefficient	$[\text{m}^2 \cdot \text{s}^{-1}]$
E	activation energy	$[\text{J} \cdot \text{mole}^{-1}]$
G	correction factor	$[-]$
Gr_m	mass Graetz number	$[-]$
Gr_t	temperature Graetz number	$[-]$
h	(global) heat transfer coefficient	$[\text{kW} \cdot \text{m}^{-2} \cdot \text{K}^{-1}]$
h_{gs}	gas solid heat transfer coefficient	$[\text{kW} \cdot \text{m}^{-2} \cdot \text{K}^{-1}]$
h_w	heat transfer coefficient at the reactor wall	$[\text{kW} \cdot \text{m}^{-2} \cdot \text{K}^{-1}]$
I_1	half cycle CSS measure	$[\text{K}]$
I_2	full cycle CSS measure	$[\text{K}]$
J	cost criterion	$[\text{kmole} \cdot \text{m}^{-3}]$
k	rate constant	$[\text{s}^{-1}]$
k_{gs}	gas solid heat transfer coefficient	$[\text{s}^{-1}]$
K_1	scaling factor for the terminal cost criterion	$[\text{K}^2 \cdot \text{kmole}^{-1} \cdot \text{m}^3]$
K_2	scaling factor for the terminal and integral cost criterion	$[\text{K}^2 \cdot \text{kmole}^{-1} \cdot \text{m}^3]$
L	reactor length	$[\text{m}]$
L_j	jacket length	$[\text{m}]$
N	number of discretisation points	$[-]$
Nu	Nusselt number	$[-]$
R	ideal gas constant	$[\text{J} \cdot \text{mole}^{-1} \cdot \text{K}^{-1}]$
Sh	Sherwood number	$[-]$
t	time	$[\text{s}]$
T	reactor temperature	$[\text{K}]$
T_w	jacket fluid temperature	$[\text{K}]$
v	gas velocity	$[\text{m} \cdot \text{s}^{-1}]$
z	spatial coordinate	$[\text{m}]$

Greek letters

β	(reduced) heat transfer coefficient, $\frac{4h}{\rho c_p d}$	$[\text{s}^{-1}]$
Δ	(dimensionless) heat transfer coefficient used by Khinast et al. (1998)	$[-]$
$-\Delta H$	heat of reaction	$[\text{kJ} \cdot \text{kmole}^{-1}]$
ε	void fraction	$[-]$
λ	heat conductivity	$[\text{kW} \cdot \text{m}^{-1} \cdot \text{K}^{-1}]$
$\overline{\rho c_p}$	fixed bed heat capacity	$[\text{kJ} \cdot \text{m}^{-3} \cdot \text{K}^{-1}]$
$\rho_g c_{pg}$	gas heat capacity	$[\text{kJ} \cdot \text{m}^{-3} \cdot \text{K}^{-1}]$
τ	switching time	$[\text{s}]$
ω	velocity of the temperature front	$[\text{m} \cdot \text{s}^{-1}]$

Subscripts

ax	axial value
eff	effective value
i	value at the i -th discretisation point
j	value after the j -th flow reversal
in	value at the inlet
int	intermediate value
ll	lower limit
max	maximum value
out	value at the outlet
rad	radial value
ref	reference value
s	value for the solid
TC	value for the terminal cost criterion
TIC	value for the terminal and integral cost criterion
ul	upper limit

Superscripts

start start value

*** optimal value

' value adapted to RFR

Acronyms

CSS cyclic steady-state

DDS direct dynamic simulation

RFR reverse flow reactor