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# OPTIMAL JACKETED TUBULAR REACTOR OPERATION: CLASSIC VS. FLOW REVERSAL

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Abstract: Optimal steady-state temperature profiles for jacketed tubular reactors often exhibit a trapezoidal shape along the reactor, i.e., first an increase, then a constant part and finally a decrease (Smets *et al.*, 2002; Logist *et al.*, 2006*b*). The practical realisation of these reactor temperature profiles is complex given the (infinite dimensional) spatially varying jacket fluid temperature profile required for the constant reactor temperature part. Therefore, based on simulations this paper compares two practically feasible alternatives with a near-optimal performance. The first splits the jacket into a finite number of isothermal zones. The second considers a flow reversal strategy, which also induces trapezoidally shaped temperature profiles (Logist *et al.*, 2006*a*). 

Keywords: chemical reactors, computer-aided design, computer simulation, distributed-parameter systems, model-based control, optimal control

## 1. INTRODUCTION

For classic *jacketed tubular reactors* optimal temperature profiles often exhibit a trapezoidal shape, i.e., increasing until a certain reactor temperature is reached, keeping that temperature constant over an interval, and decreasing the temperature towards the end (Smets *et al.*, 2002; Logist *et al.*, 2006*b*). However, inducing the constant temperature part via the jacket is difficult for an exothermic reaction, because a spatially varying jacket fluid temperature profile is required.

The flow reversal or *reverse flow reactor* is an alternative configuration which integrates reaction and heat exchange. Periodically reversing the flow causes the fixed bed inside the reactor to act as a regenerative heat exchanger, typically yielding trapezoidal temperature profiles. (For an overview see the review by Kolios *et al.* (2000).) A quantitative comparison between the classic and the reverse flow reactor has been reported for the adiabatic case (Gawdzik and Rakowski, 1988). However, a surrounding cooling jacket is required when certain temperature limits must not be exceeded or when the constant temperature level has to be controlled (Khinast *et al.*, 1998). A comparison for this nonadiabatic case is still lacking.

Therefore, this paper focusses on the performance optimisation and comparison of two practically feasible jacketed reactor configurations, which lead to (near-)optimal temperature profiles. First, a classic tubular reactor with a finite number of isothermal jacket zones is selected. As second option a cooled reverse flow reactor is studied. The organisation is as follows. Section 2 introduces the reactor configurations. Section 3 describes the numerical procedures, while Section 4 discusses the results. Section 5 summarises the conclusions.

## 2. JACKETED TUBULAR REACTORS

This study concentrates on reactors in which an exothermic reaction takes place and where the produced heat is removed via a surrounding jacket. The studied classic and reverse flow configurations both involve a simple 1D model and an irreversible first-order reaction with Arrhenius kinetics enabling a fair comparison between the (cyclic) steady-state performance. Additionally, the start-up procedure is checked to ensure a safe convergence to the desired (cyclic) steady-state.

### *2.1 Classic tubular reactor configuration*

Assuming plug flow reactor (PFR) behaviour under transient conditions results in the following set of coupled nonlinear first-order hyperbolic PDEs with as independent variables the time $t$ [s] and the spatial coordinate $z$ [m]:

$$\frac{\partial C}{\partial t} = -v\frac{\partial C}{\partial z} - k_0 C e^{\frac{-E}{RT}}$$
$$\frac{\partial T}{\partial t} = -v\frac{\partial T}{\partial z} - \frac{\Delta H}{\rho_f c_{pf}} k_0 C e^{\frac{-E}{RT}} + \frac{4h}{\rho_f c_{pf} d}(T_w - T)$$

with $C(0,t) = C_{in}$ and $T(0,t) = T_{in}$. $C$ [kmole/m$^3$] and $T$ [K] indicate the reactant concentration and the temperature, $v$ [m/s] the fluid velocity, $\Delta H$ [J/kmole] the heat of reaction ($\Delta H < 0$ for an exothermic reaction) and $\rho_f$ [kg/m$^3$], $c_{pf}$ [J/kg/K], $k_0$ [1/s], $E$ [J/mole], $R$ [J/mole/K], $h$ [W/m$^2$/K], $T_w$ [K] and $d$ [m], the fluid density, the specific heat, the kinetic constant, the activation energy, the ideal gas constant, the heat transfer coefficient, the jacket fluid temperature and the reactor diameter, respectively. The steady-state balances are readily obtained by equating all derivatives with respect to time to zero.

In previous work by Smets *et al.* (2002) and Logist *et al.* (2006b) optimal infinite dimensional steady-state jacket fluid profiles $T_w(z)$ have been derived analytically (based on *optimal control theory*) for cases with and without an explicit upper reactor temperature limit. Two cost criteria (Equations (1) and (2)) have been considered, which 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:

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

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

where $A$ [-] is a trade-off coefficient ranging from zero to one, and $K_1$ [K$^2$m$^3$/kmole] and $K_2$ [K$^2$m$^3$/kmole] are scaling factors. The conversion cost $J_1$ measures the reactant concentration at the reactor outlet ($z$=$L$). The terminal energy cost $J_2$ penalises the deviation of the exit temperature $T(L)$ from a reference temperature $T_{ref,1}$ [K] in order to limit the terminal energy loss to a certain off-set value by controlling the heat recovery through the jacket. The integral type cost $J_3$ penalises temperature deviations from a reference temperature $T_{ref,2}$ along the reactor, in order to minimise the chance of hot spots. More information on the interpretation of both cost criteria can be found in Logist *et al.* (2006a). To avoid hazardous situations an upper temperature limit $T(z) \leq T_{ul}$ is often required. The resulting optimal reactor temperature profiles exhibit a trapezoidal shape, requiring for the constant reactor temperature part a spatially varying temperature inside the jacket. For a practical realisation, the jacket will be split into a finite number of isothermal parts, whose temperature level has to be optimised to achieve a (near-)optimal performance.

### *2.2 Reverse flow configuration*

The classic example of Eigenberger and Nieken (1988) deals with an insulated (adiabatic) reverse flow reactor (RFR). The addition of a cooling jacket (Khinast *et al.*, 1998) results in the following set of coupled nonlinear parabolic PDEs for the mass and energy balances:

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial z^2} - v\frac{\partial C}{\partial z} - kCe^{-E/RT}$$
$$\frac{\partial T}{\partial t} = \frac{\lambda_{eff}}{\overline{\rho c}_p}\frac{\partial^2 T}{\partial z^2} - \frac{v\varepsilon\rho_f c_{pf}}{\overline{\rho c}_p}\frac{\partial T}{\partial z} + \frac{\varepsilon}{\overline{\rho c}_p}kCe^{-E/RT}(-\Delta H) + \frac{4h}{\overline{\rho c}_p d}(T_w - T)$$

with additionally $D$ [m$^2$/s] the dispersion coefficient, $\overline{\rho c}_p$ [kJ/m$^3$/K] the fixed bed heat capacity, $\lambda_{eff}$ [kW/m/K] the effective heat conductivity and $\varepsilon$ [-] the void fraction.

The introduction of the fixed bed induces dispersion of mass and heat reflected by the axial dispersion coefficient and effective heat conductivity. As the jacket mainly covers the middle part of the reactor, while the inlet and outlet zones are still insulated, the value of the heat transfer coefficient $h$ 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. The temperature of the fluid inside the jacket is assumed to be constant along the jacket. For the classic example of Eigenberger and Nieken (1988) this approach has been successfully applied in previous work (Logist *et al.*, 2006*a*) to achieve a trapezoidal (cyclic) steady-state reactor temperature with a controllable constant temperature part. The reverse flow performance will be optimised according to the time-averaged cost criteria $J'_{TC}$ and $J'_{TIC}$ equivalent to Equations (1) and (2):

$$J'_{TC} = (1-A)\underbrace{\frac{\int_0^{2\tau} C_{out}(t)dt}{2\tau}}_{J'_1} + \underbrace{A\,\frac{1}{K_1}\left(\frac{\int_0^{2\tau}(T_{out}(t)-T_{ref,1})dt}{2\tau}\right)^2}_{J'_2} \quad (3)$$

$$J'_{TIC} = (1-A)J'_1 + \underbrace{A\,\frac{1}{K_2}\frac{\int_0^{2\tau}\int_0^{L}(T(z,t)-T_{ref,2})^2 dz\, dt}{2\tau}}_{J'_3} \quad (4)$$

Most cost parameters are user-defined and, hence, have to be adapted for a specific process. In this simulation study, $A$ is taken equal to 0.5, $K_1$ and $K_2$ to 250000 $K^2m^3$/kmole, and $T_{ref,1}$ and $T_{ref,2}$ to 350 K and 360 K, respectively. The upper reactor temperature limit $T_{ul}$ is assumed at 400 K.

In summary, this cooled reverse flow reactor and the classic jacketed tubular reactor with a finite number of isothermal jacket parts are compared as practically feasible alternatives for implementing (near-)optimal trapezoidal temperature profiles.

## 3. NUMERICAL SIMULATION AND OPTIMISATION TECHNIQUES

### *3.1 Classic tubular reactor configuration*

Since the classic *steady-state* plug flow reactor model for the jacketed tubular reactor consists of ODEs with initial conditions, the steady-state can easily be computed via a standard integrator. For the *transient* simulations of the reactor during start-up, requiring the solution of hyperbolic PDEs, an *operator splitting* algorithm (Renou *et al.*, 2003) is adopted due to its excellent behaviour in the presence of steep gradients.

The steady-state optimisation problem has a finite number of degrees of freedom (i.e., the temperatures for each of the isothermal jackets) and is subject to ODE constraints. Such problems can be solved numerically using direct *dynamic optimisation* techniques, which typically require the solution of a nonlinear programming (NLP) problem. Here, a *sequential approach* is adopted, using standard integrators to calculate the objective function at each iteration step of the optimisation routine. A stochastic NLP solver (Integrated Controlled Random Search (Banga *et al.*, 1998)) is first applied to localise the vicinity of the global minimum, while a deterministic gradient based SQP solver (`E04UCF` from NAG Fortran library) ensures in a second phase a fast convergence to this global optimum.

### *3.2 Reverse flow configuration*

Due to the periodic flow reversals, the low inlet temperature and the fixed bed, acting as a regenerative heat exchanger, a slowly moving temperature front is induced. The aim is to achieve an operation regime in which the successive reversion cycles become repeatable, i.e., the *cyclic steady-state* (CSS). *Direct dynamic simulation* (DDS) refers to the computation of the CSS by calculating the full transient from an initial state to the CSS. This approach is straightforward as accurate simulation codes for nonlinear parabolic PDEs are available, e.g., the MatMOL toolbox (Vande Wouwer *et al.*, 2004). Despite the possibly large computation time, DDS yields the entire transient behaviour towards the CSS related to a certain start-up procedure, which is useful for the operators. Initially, a uniform reactor temperature equal to the jacket temperature is assumed. To check whether a symmetric CSS is attained, two measures from Gosiewski (2004) are employed.

Based on preliminary sensitivity analysis results, the jacket fluid temperature $T_w$, the jacket length $L_j$ and the switching time $\tau$ are selected as the most important parameters to optimise the reactor performance, while accounting for the upper reactor temperature constraint $T_{ul}$. $T_w$ and $\tau$ are continuous variables, whereas $L_j$ is inherently discrete due to the spatial discretisation. Therefore, a continuous nonlinear optimisation problem in $T_w$ and $\tau$ is solved repeatedly with the Matlab routine `fmincon` for a grid of jacket lengths. The optimal parameters are finally determined by selecting the lowest cost value over all jacket lengths.

Table 1. Parameter values.

| Parameter | | Value | Units |
|---|---|---|---|
| Values selected from Smets *et al.* (2002) | | | |
| $C_{in}$ | = | 0.02 | kmole/$m^3$ |
| $E/R$ | = | 5681 | K |
| $L$ | = | 1 | m |
| $k_0$ | = | $10^6$ | 1/s |
| $T_{in}$ | = | 340 | K |
| $v$ | = | 0.1 | m/s |
| $\frac{4h}{\rho_f c_{pf}}$ | = | 0.2 | 1/s |
| $\frac{-\Delta H}{\rho_f c_{pf}} C_{in}$ | = | 85 | K |
| Values selected from Eigenberger and Nieken (1988) | | | |
| $D$ | = | $5 \cdot 10^{-3}$ | $m^2$/s |
| $\varepsilon$ | = | 0.8 | [-] |
| $\lambda_{eff}$ | = | $2.06 \cdot 10^{-3}$ | kW/m/K |
| $\overline{\rho c}_p$ | = | 400 | kJ/$m^3$/K |
| $\rho_f c_{pf}$ | = | 0.5 | kJ/$m^3$/K |

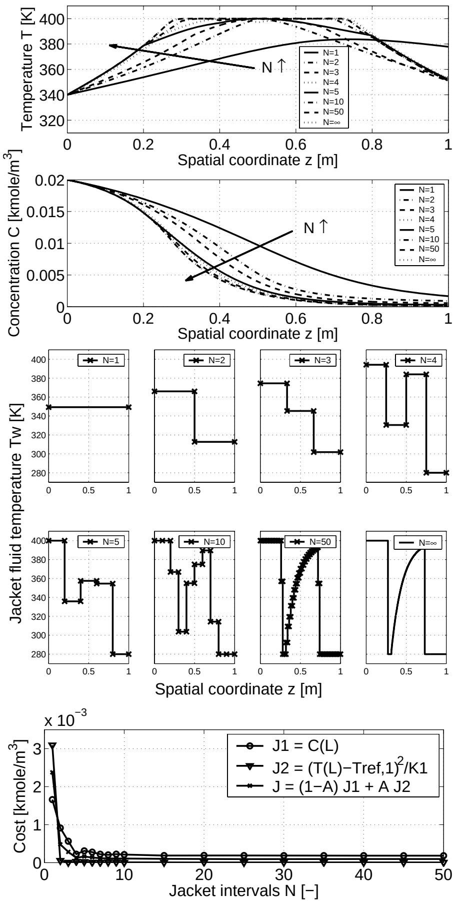


Fig. 1. Influence of the number of isothermal jackets of equal length on ($i$) the concentration and reactor temperature (top), ($ii$) the jacket fluid temperature (middle), and ($iii$) the minimal TC criterion value (bottom).

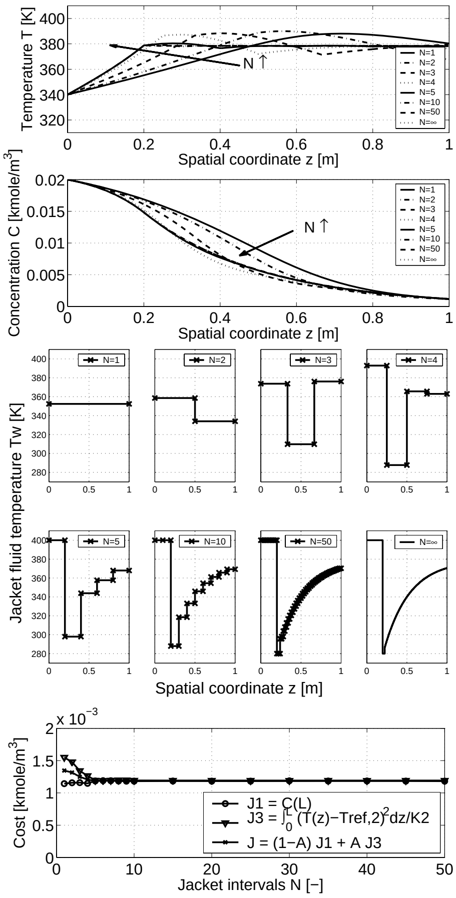


Fig. 2. Influence of the number of isothermal jackets of equal length on ($i$) the concentration and reactor temperature (top), ($ii$) the jacket fluid temperature (middle), and ($iii$) the minimal TIC criterion value (bottom).

## 4. NUMERICAL RESULTS

The parameter values are displayed in Table 1. As the aim is to compare both strategies for the case studied by Smets *et al.* (2002), most parameters originate from this study, while missing parameters for the RFR case have been filled up by values used by Eigenberger and Nieken (1988).

### *4.1 Classic tubular reactor configuration*

For the steady-state optimisation of the classic tubular reactor the influence of the number of isothermal intervals of equal length $N$ is studied. The resulting profiles for the jacket fluid temperature $T_w$, the reactor temperature $T$ and the concentration $C$ are displayed for increasing $N$ together with the evolution of the different costs.

For the *terminal cost* (TC) criterion (see Fig. 1), the jacket fluid temperature profile consisting of different isothermal jacket parts converges towards the optimal infinite dimensional one as the number of jacket parts $N$ increases, inducing ($i$) reactor temperature profiles closer to the desired trapezoidal shape and ($ii$) lower terminal cost values. A local higher value can be found, whenever the discretisation grid corresponds less to the optimal switching structure. An acceptable approximation can be obtained with 15 equidistant parts, yielding cost values within 5% of the optimal cost of $0.93733 \cdot 10^{-4}$ kmole/m$^3$.

For the *terminal and integral cost* (TIC) criterion (see Fig. 2), the optimal temperature profile conceptually consists of two parts. In the first part the temperature is raised to an intermediate value of about 378 K, which has to be kept constant in the second part. Again, increasing the number of isothermal jackets induces temperature profiles approaching the optimal infinite dimensional one. Accordingly, the cost values decrease, but only slightly. It should be noted that for both criteria a drastic cost decrease is observed when using a distributed jacket temperature ($N \geq 2$) instead of an isothermal jacket ($N = 1$), illustrating the efficiency of the here proposed operation policy.

Checking the start-up behaviour from an empty reactor at 340 K ensures for all values of $N$ a safe convergence towards the previously optimised steady-state. It is observed that the fluid flows as a plug through the reactor inducing sharp fronts without exceeding the temperature bounds. Because of space limitations the transient figures are not displayed.

### *4.2 Reverse flow configuration*

For the optimisation of the RFR, a value of 1000 s is selected as initial guess for the switching time in order to have the same ratio of residence time over switching time as in Eigenberger and Nieken (1988). The initial guess for the jacket fluid temperature $T_w$ is selected based on $(i)$ the maximum value of the previously optimised trapezoidal temperature profiles for the classic reactor and $(ii)$ the RFR sensitivity results for the maximum reactor temperature $T_{max}$ as a function of the jacket fluid temperature $T_w$ (see Fig. 3). This procedure yields 360 K and 350 K as initial $T_w$ guesses for the terminal and the combined terminal and integral cost criterion, respectively. For both criteria the evolution of the costs, the optimal switchings and the optimal jacket fluid temperatures as a function of the jacket length are depicted together with the optimal CSS profiles.

For the *terminal cost* criterion a continuous optimisation of the jacket fluid temperature $T_w$ and the switching time $\tau$ is performed for a grid of jacket lengths from 0.4 to 1 m in steps of 0.1 m. The bounds on the switching time and the jacket fluid temperature are respectively, 100 and 2500 s, and 330 and 370 K. As can be seen in Fig. 4, once a minimum jacket length has been exceeded, hardly any influence on the cost is visible. However, a minimum cost of $2.0884 \cdot 10^{-4}$ kmole/m$^3$ is obtained with a jacket of 0.9 m, a switching time of 590.1 s and a jacket fluid temperature of 368.5 K. The resemblance between the resulting optimal CSS concentration and temperature profiles (see Fig. 4) and the profiles for the classic tubular reactor (see Fig. 1) is remarkable. The minimal RFR cost value corresponds to a cost value obtained with 4 isothermal jacket parts of equal length ($1.2586 \cdot 10^{-4}$ kmole/m$^3$) for the classic reactor. The corresponding conversion costs are $4.1473 \cdot 10^{-4}$ kmole/m$^3$ for the RFR, and $2.2259 \cdot 10^{-4}$ kmole/m$^3$ for the classic reactor, respectively. Thus, although a similar trapezoidal shape is induced by the RFR, a higher (conversion) cost is found because the upper temperature limit cannot be reached as closely as with a classic configuration with a limited number ($N \geq 4$) of isothermal jackets.

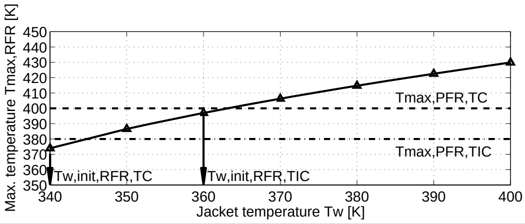


Fig. 3. RFR: Influence of the jacket fluid temperature on the maximum reactor temperature.

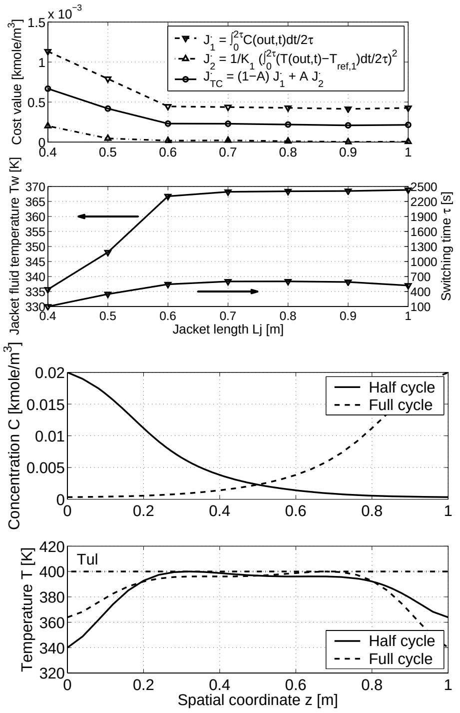


Fig. 4. TC criterion, optimal jacket fluid temperatures and switching times vs. the jacket length (top). Optimal CSS concentration and reactor temperature profiles (bottom).

A similar procedure is applied to the *terminal and integral cost* criterion. The bounds for $T_w$ remain unaltered, but the upper bound for $\tau$ is now increased to 5000 s. As also observed for the classic configuration, the terminal and integral cost is less sensitive with respect to the parameters (see Fig. 5). Although the minimum cost is found for a jacket length $L_j = 0.5$ m ($J'_{TIC} = 1.4615 \cdot 10^{-3}$ kmole/m$^3$), it is possible to find for each jacket length an appropriate switching time and jacket fluid temperature which result in nearly the same performance. Therefore, a jacket length of 0.9 m is here selected as the optimal one although it does not yield the best overall performance ($J'_{TIC} = 1.5273 \cdot 10^{-3}$ kmole/m$^3$). However, this configuration leads to the lowest conversion cost value ($J'_1 = 1.4593 \cdot 10^{-3}$ kmole/m$^3$) and the longer jacket allows to directly influence a larger reactor part, which is advantageous from a control and safety point of view. The corresponding optimal switching time and jacket fluid temperature are 1007.3 s and 352.9 K, respectively.

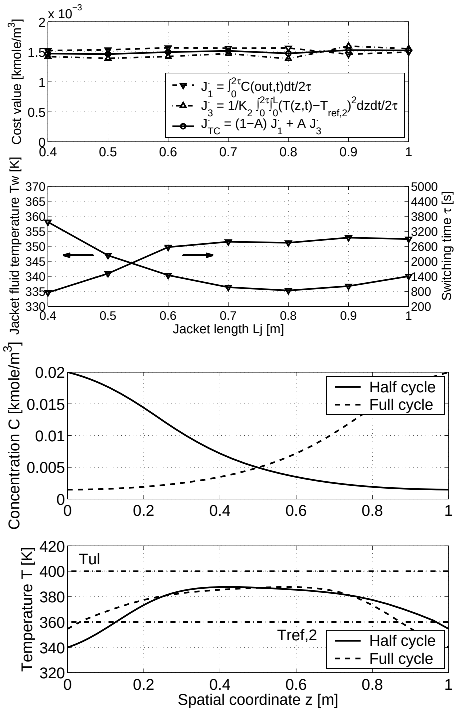


Fig. 5. TIC criterion, optimal jacket fluid temperatures and switching times vs. the jacket length (top). Optimal CSS concentration and reactor temperature profiles (bottom).

Comparing the resulting RFR profiles with the ones for the classic reactor, highlights two observations. First, there is a temperature decrease towards the end, and second, the constant temperature level is higher. These observations can easily be explained because the intrinsic decrease at the reactor end leads to smaller contributions to the energy cost at the end, which can be compensated by higher temperatures in the middle of the reactor. However, the corresponding cost values ($J_1' = 1.4593 \cdot 10^{-3}$ kmole/m$^3$ and $J_3' = 1.5941 \cdot 10^{-3}$ kmole/m$^3$) are larger than all equivalents for a classic reactor, indicating that the higher temperatures in the middle (and the higher conversion), cannot compensate for the temperature decrease and conversion loss at the end. Checking the start-up behaviour reveals no complex phenomena and ensures a safe convergence to the CSS. The corresponding figures are omitted due to space limitations.

## 5. CONCLUSIONS

In this paper two jacketed tubular reactor configurations have been compared for a practical realisation of (near-)optimal trapezoidal temperature profiles. As a first option the jacket is discretised in a finite number of isothermal parts of which the temperatures have to be optimised. A second option is to implement a flow reversal strategy which directly leads to trapezoidal temperature profiles and has the jacket fluid temperature, the switching time and the jacket length as main degrees of freedom. Two cost criteria have been considered which both involve a conversion and an energy cost. For both options a reasonable approximation of the optimal steady-state profiles and a safe convergence during start-up has been obtained. The classic configuration outperforms the reverse flow reactor, when a sufficient number of isothermal intervals is installed. However, the more isothermal intervals, the more complex the control scheme will be. The flow reversal strategy requires more equipment and its more complex (cyclic) behaviour may hamper the control design.

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