OPTIMAL CONTROL OF DISPERSIVE TUBULAR CHEMICAL REACTORS: PART I

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Abstract: This paper, the first of a series of two, deals with the determination of optimal steady-state jacket fluid temperature profiles for dispersive tubular chemical reactors, ranging from plug flow to perfectly mixed reactors. According to Pontryagin's minimum principle, the optimal control is of the bang-bang type for the proposed terminal cost criterion. The bang-bang switching position is numerically optimised, by means of a weighted shooting-type procedure for the determination of the reactor profiles, ensuring the Danckwerts boundary conditions are satisfied. Following this procedure the impact of dispersion on the optimised profiles is illustrated. In the second paper, the performance of the obtained optimal control laws will be compared with that of practically more feasible controls. Furthermore, the transient behaviour will be assessed.

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

1. INTRODUCTION

An intriguing and persistent challenge for chemical process control engineers is to master the problem of optimal control of (nonlinear) chemical processes. Over the past decades optimisation of chemical reactors has indeed received considerable attention in the published literature. However, only a very limited number of analytical optimal control profiles has been reported, and this merely for either *(i) plug flow* reactors (PFR) (see, e.g., Chou *et al.* (1967) and Smets *et al.* (2002)), or *(ii) continuous stirred tank* reactors (CSTR) (see, e.g., Ray (1981)). The influence of axial dispersion, introducing a certain degree of backmixing, on optimal tubular reactor performance is almost never taken into account. This lack does not come as a surprise since the axial dispersion model with Danckwerts boundary conditions, introduces second-order terms in the mass and energy balance equations, which already encompass highly nonlinear reaction terms. Hence, sophisticated techniques for steady-state simulation of the reactor behaviour become indispensable.

Several techniques to solve second-order nonlinear boundary value problems are mentioned in literature (Kubíček and Hlaváček, 1983).

A first option is converting the set of differential equations and boundary conditions into a set of nonlinear algebraic equations by rewriting all derivatives as *finite differences*. Since most often a large number of grid points are required for an accurate simulation, this method becomes computationally quite expensive when the resulting large set of nonlinear equations has to be solved.

A second technique, called the *false transient method*, assumes the steady-state problem to be solved whenever all transient features have disappeared from the transient simulation of the dynamic mass and heat balances. Hereto, the

solution of a set of parabolic partial differential equations (PDEs) is required, for which most simulation methods are primarily based on the approximation of the set of partial differential equations by a set of ordinary differential equations (Hundsdorfer and Verwer, 2003). Although this approach is generally applicable, it is again rather time consuming.

A third alternative are *shooting methods*, which are iterative procedures based on the solution of a corresponding initial value problem. This type of method is attractive because of the availability of powerful routines for numerical integration of initial value problems and it can often successfully be applied to boundary value problems of any complexity as long as the corresponding initial value problem is stable and a good set of starting values is available. A procedure of this last type is implemented in this paper, enabling the simulation and optimisation of the dispersive tubular reactor, while the (false) transient behaviour is studied in Part II (Logist *et al.*, 2005).

The organisation of the paper is as follows. Section 2 introduces the mathematical model. Section 3 states the optimal control problem and derives the analytical control law. Section 4 outlines the weighted shooting-type procedure, which is applied in Section 5. In this section, the influence of dispersion on the optimal control profiles and the sensitivity of the optimal control with respect to the trade-off parameter in the cost criterion, are illustrated. Finally, Section 6 summarises the main conclusions.

2. THE AXIAL DISPERSION MODEL

The reactor under study is a classical tubular reactor in which an irreversible, exothermic, first-order reaction takes place. A surrounding heating/cooling jacket is used to control the reactor temperature, as depicted in Figure 1. Describing the reactor under steady-state conditions by a 1D-model with axial mass and heat dispersion results in the following system of two second-order differential equations with respect to the spatial coordinate z and four Danckwerts boundary conditions (Danckwerts, 1953):

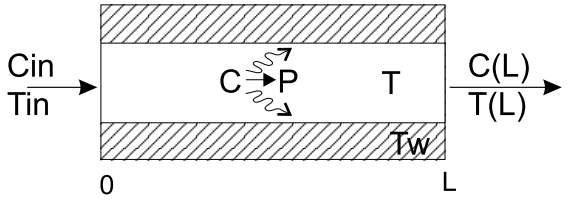

Fig. 1. Tubular reactor with surrounding heating/cooling jacket.

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

$$D_2\frac{d^2C}{dz^2} - v\frac{dC}{dz} - k_0 C e^{\frac{-E}{RT}} = 0$$

$$\begin{aligned} D_1\frac{dT}{dz} &= v(T - T_{in}) \quad \text{at} \quad z = 0 \\ D_2\frac{dC}{dz} &= v(C - C_{in}) \quad \text{at} \quad z = 0 \\ \frac{dT}{dz} &= 0 \qquad\qquad \text{at } z = L \\ \frac{dC}{dz} &= 0 \qquad\qquad \text{at } z = L \end{aligned}$$

where T [K] and C [mole/L] are the temperature and the reactant concentration, respectively, D_1 [m^2/s] and D_2 [m^2/s] are the energy and mass dispersion coefficients, respectively, v [m/s] is the fluid superficial velocity, ΔH [cal/mole] is the heat of reaction ($\Delta H < 0$ for an exothermic reaction), ρ [kg/m^3], C_p [cal/kg/K], k_0 [1/s], E [cal/mole], R [cal/mole/K], h [cal/s/m^2/K], d [m] and T_w [K] are the 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. T_{in} and C_{in} are the respective feed temperature and concentration (at $z = 0^-$).

The dispersion parameters D_1 and D_2 take deviations from the plug flow behaviour, caused by *(i)* turbulence, *(ii)* a nonuniform velocity profile over the cross-section and *(iii)* molecular diffusion, into account and introduce a certain amount of back-mixing (i.e., upstream material and heat transport) (Kramers and Westerterp, 1963). Interestingly, these parameters can also be used to describe an entire range of reactors. When no dispersion is present, the model reduces to the idealised plug flow model, while an infinite amount of dispersion induces infinite back-mixing, yielding perfectly mixed continuous reactor behaviour, encountered in an idealised continuous stirred tank reactor. Observe that in literature, dimensionless Peclet numbers Pe ($Pe \triangleq v \cdot L/D$), indicating the relative importance of convective over dispersive transport, are often used to describe the reactor behaviour instead of the dispersion coefficients themselves.

3. THE OPTIMAL CONTROL PROBLEM

The ingredients for developing a model-based, optimal control strategy are *(i)* a mathematical model that captures the main process characteristics, *(ii)* the determination of constraints imposed on the states and/or controls and *(iii)* the specification of a cost criterion that has to be minimised (Kirk, 1970). For the studied system, these elements are elaborated in the following subsections.

3.1 Mathematical model

Before optimal control theory is applied, the axial dispersion model is adapted in two ways. To compensate for the large differences in order of magnitude between the temperature and the concentration (possibly resulting in numerical simulation inaccuracies), the dimensionless states x_1 and x_2, the dimensionless control input u, and the constants α, β, γ and δ are first introduced.

$$x_1 \triangleq \frac{T - T_{in}}{T_{in}}, \quad x_2 \triangleq \frac{C_{in} - C}{C_{in}}, \quad u \triangleq \frac{T_w - T_{in}}{T_{in}},$$
$$\alpha \triangleq k_0 e^{\frac{-E}{RT_{in}}}, \quad \beta \triangleq \frac{4h}{\rho C_p d}, \quad \gamma \triangleq \frac{E}{RT_{in}} \text{ and}$$
$$\delta \triangleq -\frac{\Delta H}{\rho C_p}\frac{C_{in}}{T_{in}}$$

Secondly, since optimal control theory is preferably applied to systems of first-order differential equations, the system of two second-order differential equations is transformed by the introduction of two new variables x_3 and x_4, representing the respective gradients,

$$x_3 \triangleq \frac{dx_1}{dz} \quad \text{and} \quad x_4 \triangleq \frac{dx_2}{dz}$$

into a system of four first-order differential equations and four boundary conditions.

$$\begin{aligned}
\frac{dx_1}{dz} &= x_3 \\
\frac{dx_2}{dz} &= x_4 \\
\frac{dx_3}{dz} &= \frac{v}{D_1}x_3 - \frac{\alpha\delta}{D_1}(1 - x_2)e^{\frac{\gamma x_1}{1+x_1}} - \frac{\beta}{D_1}(u - x_1) \\
\frac{dx_4}{dz} &= \frac{v}{D_2}x_4 - \frac{\alpha}{D_2}(1 - x_2)e^{\frac{\gamma x_1}{1+x_1}}
\end{aligned} \tag{1}$$

$$D_1 x_3 - v x_1 = 0 \quad \text{at} \quad z = 0 \tag{2}$$
$$D_2 x_4 - v x_2 = 0 \quad \text{at} \quad z = 0 \tag{3}$$
$$x_3 = 0 \quad \text{at} \quad z = L$$
$$x_4 = 0 \quad \text{at} \quad z = L$$

These equations will form the basis for the derivation of optimal jacket temperature profiles.

3.2 Cost criterion

In this study the following terminal cost criterion is proposed:

$$\mathcal{J}[u] = (1 - A')\underbrace{(1 - x_2(L))}_{\mathcal{J}_1[u]} + A'\underbrace{\frac{x_1^2(L)}{K_1'}}_{\mathcal{J}_2[u]}$$

or equivalently:

$$\mathcal{J}[u] = (1 - A)\underbrace{C(L)}_{\mathcal{J}_1[u]} + A\underbrace{\frac{(T(L) - T_{in})^2}{K_1}}_{\mathcal{J}_2[u]} \tag{4}$$

with A' (or equivalently A) the trade-off coefficient between the conversion cost $\mathcal{J}_1$ and the energy consumption cost $\mathcal{J}_2$, and K_1' (or equivalently K_1) a user defined weighting factor to equalise the order of magnitude of both costs.

The conversion cost part is a measure for the process efficiency (reactor outlet concentration), while the energy consumption cost part penalises for excessive heating or cooling.

3.3 Constraints

Constraints on the state variables are imposed for physical reasons: $T(z) > 0$, $0 < C(z) < C_{in}$ and $\frac{dC}{dz}(z) \leq 0$, which induce $x_1(z) > -1$, $0 < x_2(z) < 1$ and $x_4(z) \geq 0$. Furthermore, an upper and a lower limit are taken into account for the jacket fluid temperature: $T_{w,min} \leq T_w \leq T_{w,max}$ which induces $u_{min} \leq u(z) \leq u_{max}$.

3.4 Solution

The optimal control problem statement is the following. *Find an admissible control which causes the given reactor system to follow an admissible trajectory while at the same time minimising the performance criterion* (Kirk, 1970). It has been shown by the authors that for the above mentioned optimal control problem, the extremal control law is of the bang-bang type (Logist *et al.*, 2004). Intuitively, a maximum-minimum (max-min) step profile is proposed to first favour the reaction and then lower the temperature to decrease the energy cost.

4. NUMERICAL SIMULATION TECHNIQUE

As already mentioned, the steady-state reactor profiles are calculated by means of a weighted shooting-type procedure. Since both at the reactor inlet and outlet only two boundary conditions are specified, the boundary value problem under study is of order two. The shooting-type procedure consists of selecting two missing values, both either at the inlet or at the outlet and then integrating the system of first-order differential equations. At the other boundary the boundary conditions have to be checked and as long as they are not satisfied, the choice of the missing values has to be adapted.

To avoid numerical instabilities, often encountered with forward integration of the system under study (Kubíček and Hlaváček, 1983), the problem formulation is reversed with respect to previous work (Logist *et al.*, 2004): the two missing conditions at reactor outlet ($x_1(L)$ and $x_2(L)$) have to be chosen in such a way that after backward integration of the system of first-order differential equations (1), the resulting inlet values satisfy boundary conditions (2) and (3). This problem is formulated as a minimisation problem of an objective function G, which is a weighted sum of the squared inlet boundary conditions. K is a strictly positive scaling factor in order to bring both parts in the same order of magnitude, here equal to 100.

$$G = K(D_1 x_3(0) - v x_1(0))^2 + (D_2 x_4(0) - v x_2(0))^2$$

Evidently, the correct outlet boundary values are found when G reaches its minimum value of 0. The integration of the ODE system is performed with the `ode15s` routine while the optimisation routine is the `e04jaf` routine from the NAG-toolbox, both in `Matlab`© (The Mathworks Inc., Natick).

5. NUMERICAL SIMULATIONS

In this section, the reactor performance under the analytically derived bang-bang control is numerically investigated. The calculations are performed for tubular reactors ranging from nearly plug flow reactors ($Pe = 10^8$) to almost perfectly mixed continuous stirred tank reactors ($Pe = 0.01$), using the weighted shooting-type procedure. It should be noted here, that for the simulations both mass and heat dispersion are assumed to have the same importance, reflected by equal values for the heat and mass dispersion coefficients D_1 and D_2 (and consequently also the same heat and mass Peclet numbers). In addition, the optimal values for the idealised, limit cases, i.e., the PFR and the CSTR, are computed. Afterwards the sensitivity of the optimised control profile on the trade-off coefficient A is studied.

The (fixed) process parameters values used for the numerical simulations originate from Smets *et al.* (2002) and are summarised in Table 1.

Table 1. Parameter values.

Parameter		Value	Units
v	=	0.1	m/s
L	=	1	m
δ	=	0.25	[-]
E	=	11250	cal/mole
k_0	=	10^6	1/s
β	=	0.2	1/s
C_{in}	=	0.02	mole/L
R	=	1.986	cal/(mole · K)
T_{in}	=	340	K
$T_{w,min}$	=	280	K
$T_{w,max}$	=	400	K
K_1	=	250000	[-]

5.1 Max-min control law

The practical implementation of the max-min control profile involves a sequence of two heat exchangers, a first one with a constant temperature $T_{w,max}$ from the inlet to a switching position z_1, followed by a second one with a constant temperature $T_{w,min}$ from z_1 to the reactor outlet.

In this section, the optimal switching positions are numerically determined. For 101 equally distributed switching positions z_1 and for 15 different Peclet values (10^8, 10^7, 10^6, 10^5, 10^4, 10^3, 200, 100, 20, 10, 2, 1, 0.5, 0.1 and 0.01), the exact initial conditions are calculated and used to compute the corresponding reactor profiles and the value of the cost criterion $\mathcal{J}$ (Equation (4)). Figure 2 illustrates the evolution of this cost and its constituting parts (i.e., the conversion cost $\mathcal{J}_1$ and the energy cost $\mathcal{J}_2$), as a function of the switching position z_1. Without loss of generality, only the results for five Peclet numbers (0.1, 1, 2, 10 and 20) and for the PFR and CSTR cases, are depicted, to avoid overloading the plots. Figure 3 displays the resulting optimal reactor profiles. The following observations can be made.

- A first important observation is that in all plots, for increasing Peclet numbers, the results for the axial dispersion model converge towards the plug flow values, while for decreasing Peclet numbers, a convergence towards the CSTR results is visible. Broadly speaking, the transition from a more or less perfectly mixed behaviour to a more or less plug flow regime is observed within a narrow range of Peclet numbers ($Pe = 0.1$ to 10).
- The conversion cost $\mathcal{J}_1$ always reaches its minimum at a switching position z_1 equal to 1 m, which means that the optimal temperature profile is $T_w(z) = T_{w,max}$ for all values of z. This is not surprising because higher (reactor) temperatures favour the conversion.
- The energy consumption cost $\mathcal{J}_2$ first decreases with increasing switching position z_1, then reaches almost zero and finally increases again. This tendency reflects the fact that both too high and too low reactor (and hence jacket fluid) temperatures are penalised.
- When both the conversion cost $\mathcal{J}_1$ and the energy consumption cost $\mathcal{J}_2$ are equally weighted ($A = 0.5$) in the total cost criterion $\mathcal{J}$, an intermediate optimal switching position results. Increasing Peclet numbers give rise to a lower optimal cost and a lower switching position.

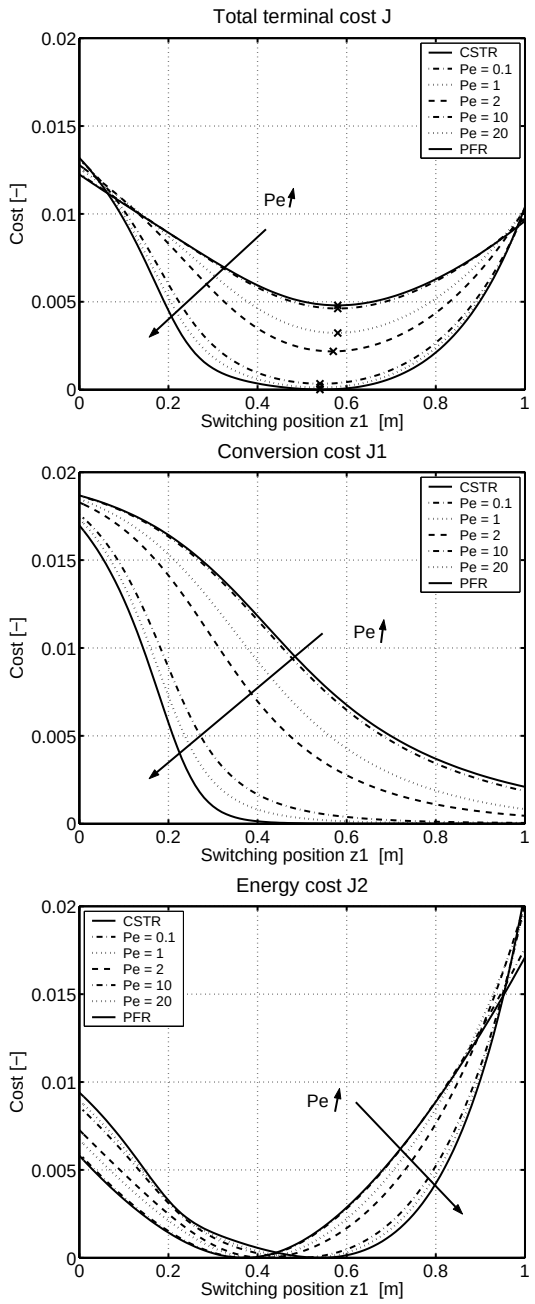

Fig. 2. Total terminal cost $\mathcal{J}$ (top), conversion cost $\mathcal{J}_1$ (middle) and energy cost $\mathcal{J}_2$ (bottom) as a function of the switching position z_1 for various Peclet numbers Pe.

This observation is more elaborated in Figure 4, where the minimal cost value and the optimal switching positions are shown as a function of the Peclet number. Two conclusions can be deduced.

- For very high Peclet numbers the results converge towards the plug flow results described by Smets *et al.* (2002). The very low cost value, encountered for high Peclet numbers can be explained by the more plug flow like behaviour of the tubular reactor if dispersion is not well pronounced. The conversion in a plug flow reactor is known to be higher than in a perfectly mixed configuration for a reaction with monotonically increasing kinetics, e.g., an irreversible first-order reaction.
- An important practical feature is the fact that dispersion has almost no effect on the optimal switching position z_1^*. This is a very useful conclusion for tubular reactor design because the dispersion coefficients D_1 and D_2 are hard to measure or predict accurately.

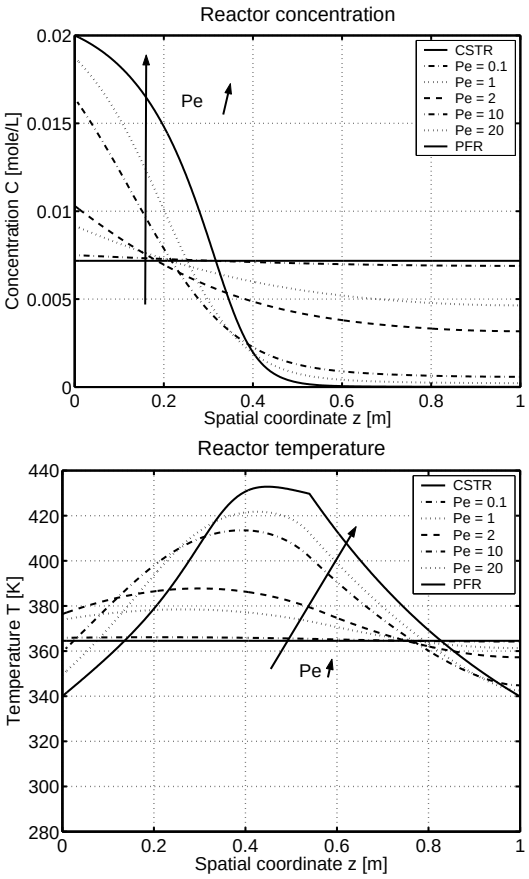

Fig. 3. Optimal concentration (top) and temperature (bottom) profiles for different Peclet numbers Pe.

5.2 The influence of the trade-off coefficient A

In this section, the influence of the trade-off coefficient A is assessed. It is clear that for higher values of A (e.g., A=0.7), more attention is paid to a low energy cost, while for lower A values (e.g., A=0.3), a low outlet concentration is emphasised.

Similar to Figure 4 the influence of dispersion on the minimal cost and the optimised control variables is depicted in Figure 5, but here for two additional values of A (i.e., 0.3 and 0.7). It can easily be seen that, for the bang-bang control, increasing the value of A causes a decrease in the optimal switching position and the minimal cost, but only for low Peclet numbers ($Pe < 10$).

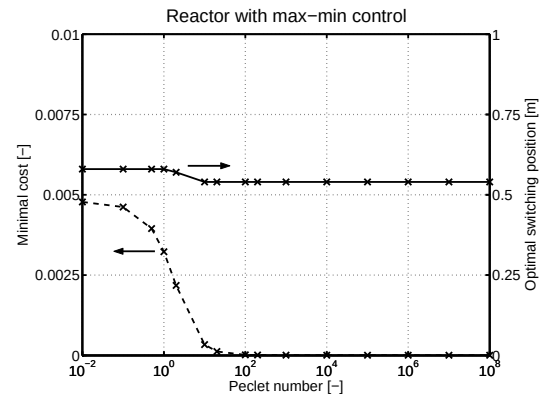

Fig. 4. Minimal cost value (- - -) and optimal switching position (—) as a function of the Peclet number Pe.

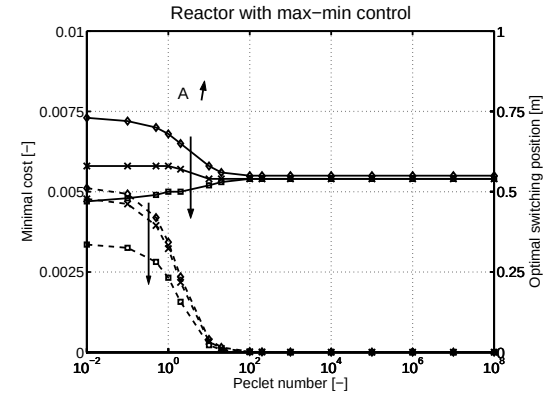

Fig. 5. Minimal cost value (- - -) and optimal switching position (—) as a function of the Peclet number Pe for various trade-off coefficient A (A=0.3 (♢), 0.5 (x) and 0.7 (□)) in the reactor with max-min control.

6. CONCLUSIONS

In the context of optimal control of chemical reactors, this paper, being the first paper of a series of two, focusses on optimal jacket fluid temperature profiles for exothermic tubular chemical reactors with axial mass and heat dispersion. By applying Pontryagin's minimum principle, the optimal control law for the system under steady-state, i.e., a bang-bang control was previously determined. Since the minimum principle provides only the type of extremal control sequences and not the exact switching positions, the latter have to be found by numerical optimisation. Hereto, an efficient shooting-type procedure is developed to obtain the temperature and concentration profiles which satisfy the Danckwerts boundary conditions. Based on this procedure, simulations are performed to determine the optimal switching position for a max-min jacket fluid temperature for whole family of tubular reactors, i.e, from almost perfectly mixed reactors with a CSTR behaviour to nearly plug flow reactors. The results show only a weak influence of dispersion on the optimal switching position, which is an extremely useful conclusion for practical reactor design. In addition, the influence of the trade-off coefficient A in the cost criterion is investigated. Here, an increased emphasis on the energy cost results in a decrease of the optimal switching position and the minimal cost. In the second paper (Logist *et al.*, 2005), the performance of the here introduced optimal control solutions will be compared with that of practically more feasible control inputs and the transient behaviour will be assessed.

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