REVERSE FLOW CATALYTIC COMBUSTORS: COMPARISON BETWEEN CLOSED AND OPEN LOOP DYNAMICS

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Abstract
In this paper the dynamics of reverse flow catalytic combustor with and without control are compared and discussed. The dynamics of the closed-loop system is much more complex. In particular, the closed-loop system may admit periodic regimes with non-constant switching time which cannot be found in the open-loop one. The most intriguing dynamics of the closed-loop RFR is found to appear when a multiplicity of periodic regimes of the open-loop RFR is compatible with a single prescribed set-point temperature.

Introduction
Periodically forced tubular catalytic reactors have been successfully used for the combustion of volatile organic compounds concentrations of industrial exhaust gas (VOCs) and to improve equilibrium limited exothermic reactions such as Methanol Synthesis [1]. The most common configuration, called Reverse Flow Reactors (RFRs), involves the periodic reversion of the flow direction. For VOCs catalytic combustion the problems related to the use of RFRs are the reaction extinction and the hot spots formation, caused by variations of the feed temperature and/or concentration. To deal with the problem, a simple feedback control scheme (a one point controller) has been proposed where the flow is reversed when the temperature as measured at specific points inside the reactor falls below a certain value [2]. In our previous work, we have demonstrated that such closed loop system is a hybrid dynamical system characterized by discrete events (the inversions of the flow direction) and continuous dynamics between two successive switches [2,3]. Moreover, we showed numerically and theoretically the occurrence of typical phenomena of hybrid systems (Zeno executions) which cannot be observed in the open loop (continuous) system. The system exhibits unusual and intriguing dynamics [3,4], like: 1) A period-adding sequence not comparable to the well-studied period-doubling cascade and those originated by frequency-locking; 2) A novel route to chaos, different from those reported previously in the literature for continuous systems and for hybrid ones; 3) The coexistence of Zeno state with quasi-periodic and chaotic regimes.
In this paper, we give an explanation of this complex dynamics by comparing the dynamics of the closed loop RFR with the dynamics of the open loop RFR. In particular, we compute the periodic regimes as the switch time is varied for the open loop RFR and we evaluate if the temperature profiles are compatible with the set point temperature value in the closed loop RFR. From this comparison, we found that the most intriguing dynamics of the closed loop RFR appears when a multiplicity of periodic regimes (of the open loop RFR) are compatible with a single prescribed set point temperature value.

**Model equations and hybrid automaton of the closed loop system.**

The model used for the catalytic reactor is a heterogeneous two-phase model of the one dimensional catalytic fixed bed reactor, with a uniform distribution of catalyst. The model takes into account heat and mass transfer resistance between gas and solid phase, axial dispersion in the gas phase, axial conduction in the solid phase and cooling through reactor wall. Pseudo-steady state of mass balance in solid phase is assumed. Mass and energy balances give the following dimensionless partial differential equations:

- In the gas phase:

\[
\frac{\partial y_g}{\partial \tau} = \frac{1}{Pe_m^g} \frac{\partial^2 y_g}{\partial z^2} + (1-2IO) \frac{\partial y_g}{\partial z} + J_m^g (y_s - y_g) \tag{1}
\]

\[
\frac{\partial \theta_g}{\partial \tau} = \frac{1}{Pe_h^g} \frac{\partial^2 \theta_g}{\partial z^2} + (1-2IO) \frac{\partial \theta_g}{\partial z} + J_h^g (\theta_s - \theta_g) \tag{2}
\]

- In the solid phase:

\[
\frac{\partial \theta_s}{\partial \tau} = \frac{1}{Pe_h^s} \frac{\partial^2 \theta_s}{\partial z^2} - J_m^s (\theta_s - \theta_g) + B\eta Da (1-y_s) \exp \frac{\theta_s}{1+y_s} \tag{3}
\]

\[
J_m^s (y_s - y_g) = \eta Da (1-y_s) \exp \frac{\theta_s}{1+y_s} \tag{4}
\]

Dimensionless parameters and state variables are the same adopted in [2,3]. Danckwerts boundary conditions are assumed for concentration and temperature in the gas phase:

\[
\left. \frac{\partial y_g}{\partial z} \right|_{0} - IOPe_m^g y_g (0,t) = 0; \left. \frac{\partial \theta_g}{\partial z} \right|_{0} - IOPe_h^g (\theta_g (0,t) - \theta_{feed}) = 0; \left. \frac{\partial \theta_s}{\partial z} \right|_{0} = 0 \tag{5}
\]

\[
\left. \frac{\partial y_g}{\partial z} \right|_{1} - (1-IO) Pe_m^g y_g (1,t) = 0; \left. \frac{\partial \theta_g}{\partial z} \right|_{1} - (1-IO) Pe_h^g (\theta_g (1,t) - \theta_{feed}) = 0; \left. \frac{\partial \theta_s}{\partial z} \right|_{1} = 0 \tag{6}
\]
Equations and boundary conditions depend on a discrete variable IO, which takes into account the flow inversions. In particular, the variable IO assumes value 1 when the flow is from left to right and value 0 when the flow is from right to left. For the open loop system, the flow direction is changed at constant time equal to the switch period T, whereas for the closed loop system the flow inversion is dictated by the control law. Indeed, when the gas temperature inside the reactor, very close to the inlet of reactor, decrease up to a fixed value (i.e. set-point temperature), the control system reverses the flow direction. The continuous dynamics, described by partial differential equations, is thus interrupted by discrete events regulated by the control law. The whole system is a hybrid (discrete/continuous) spatial extended system that in abstract form can be written as \( \dot{x} = f(x, IO) \) where \( x \equiv (y_g(z,t), \theta_g(z,t), \theta_g(z,t)) \) is the continuous state variable and \( f \) is the discontinuous vector field. Schematically, the evolution of this hybrid system can be represented by a graph called hybrid automaton. The hybrid automaton of the controlled RFR is reported in Fig. 1. It describes all the possible system evolutions, during both continuous and discrete dynamics and it represents a skeleton of the numerical algorithm necessary for the numerical simulation.

\[
\begin{align*}
\theta_g(0,t) \geq &\; \theta_{\text{set-point}} \quad \text{if } IO = 1 \\
\theta_g(1,t) \geq &\; \theta_{\text{set-point}} \quad \text{if } IO = 0
\end{align*}
\]

**Figure 1.** Hybrid automaton of the controlled reverse flow reactor.

**Results and discussions**

A typical simple symmetric periodic regime, both for the closed and open loop system, is reported in Fig.2. In particular, in figure Fig.2a are reported the temporal series of temperature in two points placed at same distance from the reactor center, whereas in Fig.2b are reported spatial profiles at two successive switch instants.
Figure 2. Periodic symmetric regime at $\theta_{set\text{-point}} = -8.113$. (a) Temporal series of solid temperature in two symmetric point inside the catalytic bed; (b) Spatial profiles at two successive switch instants.

For the system symmetry [5], two points at the same distance from the reactor center have the same temporal behavior, but shifted of the switch time $T$, being the solution period $2T$. Moreover, the spatial profiles of the temperature of catalytic bed at a time $T$ are the mirror reflection of the spatial profiles at $t+T$. However, while single periodic regimes may be very similar in the open and the closed system, the overall dynamics, and thus the bifurcations of such regimes, are very different in both systems. First, in the open loop system, the period of the periodic regimes is equal or multiple of the period action, whereas in the closed loop system the regimes period is the result of the dynamics of the whole system, that is the switch time is itself a state time-dependent variable. The reason is that, although the forcing action is assumed instantaneous, the open loop is periodically forced system with a dynamics that is typical of smooth dynamical systems. On the contrary, in the closed loop system the dynamics is much more complex and peculiar of non-smooth dynamical system. Indeed, varying the set point temperature $\theta_{set\text{-point}}$, the controlled reverse flow reactor shows a very complex behavior: Zeno phenomena (see [2] for the description), multiplicity of symmetric and asymmetric regimes, quasi-periodic regimes and strange attractors. Moreover, intriguing bifurcation scenario, never observed before in smooth system, has been observed in this system, like a novel route to chaos and an unusual period adding cascade. Detailed discussions of these scenarios are reported in [2-4]. Here, we want to explain some of this complexity by searching and comparing the periodic regimes of the open loop system which are compatible with set-point temperature and the control of the closed loop system. Our guideline is this conjecture: if the closed loop system admits a periodic regime with a constant switch period, such regime must be found in the open loop system for the same parameters values and its temperature profile should be compatible with the set-point temperature in the closed loop system. We focus this comparison in the particular range of the set-point temperature values where, in the closed loop system, a multiplicity of periodic regimes has been observed. In Fig.3, the solution diagram for the closed loop system is reported as the set-point temperature is varied.
Stable periodic regimes are shown as solid lines; unstable ones are shown as short dashed lines. In all the investigated parameter range there are non-ignited regimes which are not reported in Fig.3. Four regions are located into the diagram: R1 is a region characterized by the symmetric period-1 periodic solution; A is the region in which the period-adding phenomenon appends; R2 is a region in which several periodic solutions coexist; R3 is a region in which more complex behavior are generated from periodic solutions. In regions R1,A,R2 no Zeno executions occur while in region R3 Zeno executions coexist with ignited regimes. For values of the bifurcation parameter in the interval (−8.166,−8.112) (the R1 zone), the regime is periodic with a constant value of the switch period and the typical temporal series and spatial profiles are those reported in Fig.2, where in particular the switch period is 1160. Such kind of solutions exists also in the open loop system. Indeed, for example, by imposing in the open system the switch period at 1160, we verified that such solution regimes have a temperature profile that is compatible with $\theta_{\text{set-point}} = -8.113$. These calculations have been systematically conducted varying the switch period $T$ and the results are reported in Fig.4. As it appears from the comparison of Fig.3 and Fig.4, the dynamics of the closed loop system is much more complex respect to the open loop one. Indeed, as we enter into zone A and R2 of Fig.3 we observe a multiplicity of periodic regime characterized by different switch times which alternate in the solution period. The reason is that the closed loop system has more “degrees of freedom” as the switch time is dependent variable which may have its own evolution in time. This never occurs, of course, in the open loop system where the switch time is chosen a priori and independently.
from the system dynamics. Nevertheless, in our opinion, it is not a coincidence that the zone A and zone R2 where has been observed such complexity in the closed loop system correspond to a range of the set-point temperature where, for the open loop system, more periodic regimes have a temperature profile which is compatible with a fixed value of the set-point temperature. In the open loop system, each one of these periodic regimes corresponds to a different switch time value. Thus, we may argue that the closed system has the opportunity to choose between different regimes which are compatible with the imposed control and in such way it composes more complex periodic regimes.

Figure 4. Solution diagram varying the switch time for the open loop system.

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References