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On the Dynamics of Open and Closed Loop Reverse Flow Catalytic Reactors

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In this paper the dynamics of a reverse flow catalytic reactor with and without control are compared and discussed. The control system acts on the inlet gas temperature. Depending on flow direction the temperature is measured at one specific point at the inlet edges of the reactor. The dynamics of the closed-loop system is much more complex. In particular, varying set-point temperature, we found that 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.

1. 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 (Matros and Bunimovich, 1996). The most common configuration, called Reverse Flow Reactors (RFRs), involves the periodic reversion of the flow direction, although several others periodic actions have been proposed in the literature (Russo et al. 2004; Mancusi et 2007a; Russo et al. 2006; Russo et al 2007; Mancusi et 2011; Altimari et al.2012).

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 (Mancusi et al., 2007b). In our previous works, 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 (Brasiello et al., 2005; Mancusi et al., 2007b), showing, numerically and theoretically, the occurrence of typical phenomena of hybrid systems (Zeno executions) which cannot be observed in the open loop (continuous) system.
Moreover, the system exhibits unusual and intriguing dynamics (Brasiello et al., 2010; Brasiello et al., 2005), 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

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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.

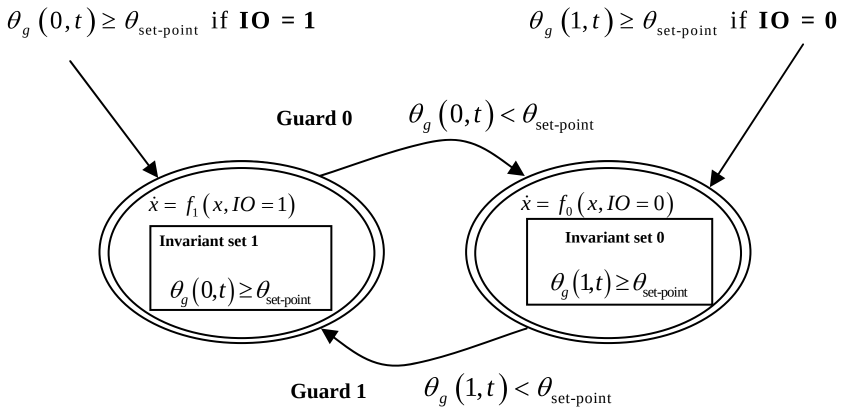

Figure 1: hybrid automaton of the controlled reverse flow reactor.

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

The model used for the description of the continuous dynamics of 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 the 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:

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

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

- In the solid phase:

$$\frac{\partial \theta_s}{\partial \tau} = \frac{1}{Pe_h^s} \frac{\partial^2 \theta_s}{\partial z^2} - J_h^s (\theta_s - \theta_g) + B\eta Da (1 - y_s) \exp \frac{\theta_s}{1 + \theta_s / \gamma} \tag{3}$$

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

Dimensionless parameters and state variables are the same adopted in (Brasiello et al., 2010; Brasiello et al., 2005). The following Danckwerts boundary conditions are assumed for concentration and temperature in the gas phase:

$$\left.\frac{\partial y_g}{\partial z}\right|_0 - IOPe_m^g y_g\left(0,t\right)=0;\left.\frac{\partial \theta_g}{\partial z}\right|_0 - IOPe_h^g\left(\theta_g\left(0,t\right)-\theta_{feed}\right)=0;\left.\frac{\partial \theta_s}{\partial z}\right|_0=0 \tag{5}$$

$$\left.\frac{\partial y_g}{\partial z}\right|_1 -\left(1-IO\right)Pe_m^g y_g\left(1,t\right)=0\,;\left.\frac{\partial \theta_g}{\partial z}\right|_1 -\left(1-IO\right)Pe_h^g\left(\theta_g\left(1,t\right)-\theta_{feed}\right)=0\,;\left.\frac{\partial \theta_s}{\partial z}\right|_1=0 \tag{6}$$

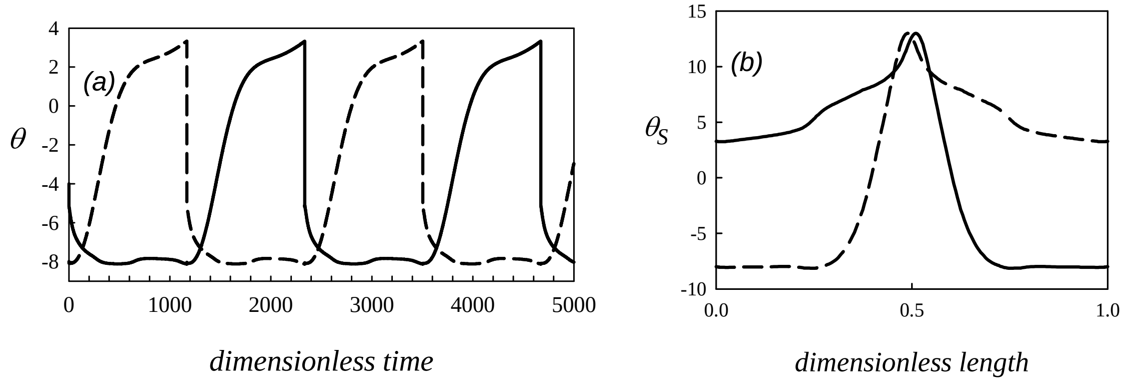

Figure 2. Periodic symmetric regime at $\theta_{\text{set-point}} = -8{,}113$. (a) Temporal series of gas temperature in two symmetric point inside the catalytic bed; (b) Spatial profiles at two successive switch instants.

Equations and boundary conditions depend on a discrete variable IO, which takes into account the flow inversions (i.e. discrete events). 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 (at about 3% of the reactor length from the edge), decrease up to a fixed value (i.e. set-point temperature), the control system reverses the flow direction. Hence, two point control devices, acting alternatively, are necessary. 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 $\dot{x} = f\left(x, IO\right)$ where $x \equiv \left(y_g\left(z,t\right), \vartheta_g\left(z,t\right), \vartheta_s\left(z,t\right)\right)$ is the continuous state variable and *f* is the discontinuous vector field.

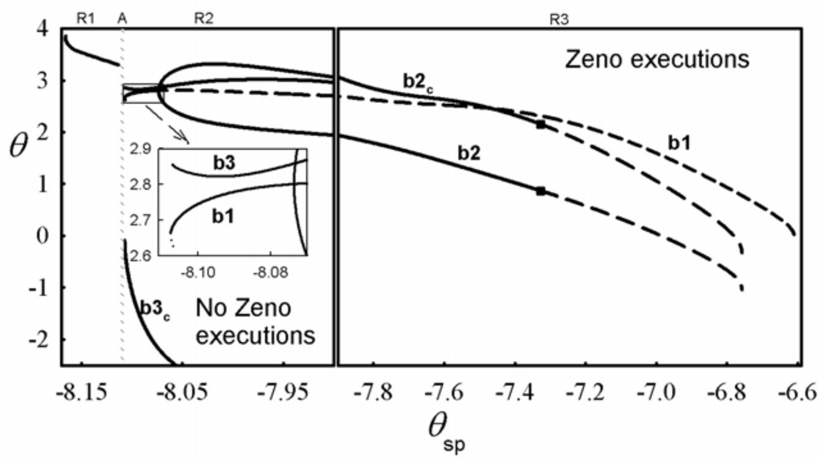

Figure 3. Solution diagram varying set-point temperature for the closed loop system.

3. Results and discussions

A typical simple symmetric periodic regime, both for the closed and open loop system, is reported in Figure 2. In particular, in Figure 2a are reported the temporal series of temperature in two points placed at same distance from the reactor centre, whereas in Figure 2b are reported spatial profiles at two successive switch instants.

For the system symmetry (Russo et al., 2002), two points at the same distance from the reactor centre have the same temporal behaviour, 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 a periodically forced system with dynamics that is typical of a smooth periodically forced dynamical system. 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, the controlled reverse flow reactor shows a very complex behaviour: Zeno phenomena (see Mancusi et al. 2007b 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 Brasiello et al. (2010) and Mancusi et al. (2007b). 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 Figure 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 Figure 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 Figure 2, where in particular the switch period is 1160.

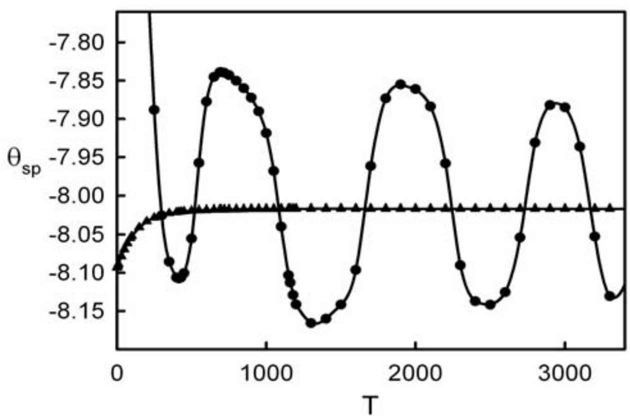

Figure 4. Solution diagram varying the switch time for the open loop system.

Such kind of solutions exist 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 $\theta_{\text{set-point}} = -8,113$.

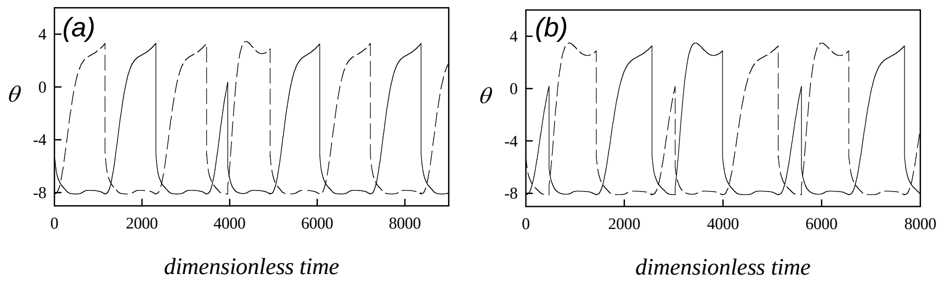

Figure 5. Periodic regimes in A region: Temporal series of gas temperature in two symmetric point inside the catalytic bed. (a) asymmetric periodic regime with 3 switch times; (b) symmetric periodic regime with 3 switch times.

These calculations have been systematically conducted varying the switch period T and the results are reported in Figure 4.
As it appears from the comparison of Figure 3 and Figure 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 Figure 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 a dependent variable which may have its own evolution in time. For instance, in Figure 5, temporal series related to two periodic regimes each characterized by 3 different switch times which follow one other periodically are reported. The two regimes differs each other in spatial symmetry.
This situation 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. It is worth to note that complex behaviours involving the period adding phenomenon and the symmetry breaking bifurcation of the periodic solutions in the closed-loop scheme correspond in the open-loop scheme to a temperature region in which multiple periodic solutions coexist with a non-ignited periodic regime.

4. Conclusions

In this paper the comparison between open-loop and closed-loop system dynamics of a reverse flow reactor is discussed. The objective is to underline common points between the two configurations to derive some explanation of the occurrence of complex dynamics in the controlled RFR. It has been observed that the two schemes admit the same periodic regimes in the case of constant switch period. However, the closed-loop scheme shows a richer dynamics as well as periodic regimes characterized by several switch periods with alternates each other periodically. We observe that such dynamics manifest for set-point temperatures which are compatible with several periodic regimes and one periodic non-ignited regime in the open-loop scheme. Although these observations are promising to construct a guideline for enhancing process conduction and control, further studies are needed to understand more accurately the relationship between the dynamics of the open-loop and closed-loop system.

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