Spatio-temporal patterns in loop reactors with different switch strategies

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We consider a NTW of three fixed bed reactors forced with two different switch strategies characterized by different feed/discharge permutations. We show that different forcing strategies induce very different stability ranges of the T-periodic regimes in the NTW. This is demonstrated both by bifurcation analysis, and by comparing the related spatio-temporal patterns generated by the two strategies. The bifurcation analysis is carried out choosing as bifurcation parameter the switch time. The stability limits are then compared with those predicted by the criterium proposed by Sheintuch and Nekhamkina (2005).

1. Introduction

Several studies have shown that many irreversible exothermic catalytic processes can be efficiently carried out in periodically-forced fixed-bed reactors. The reverse flow reactor (RFR) (Matros, 1989) is by far the most extensively studied periodically-forced reactor. In the RFRs the flow inversion traps the reaction heat in the central part of the reactor, the edges remaining colder. In optimal conditions, after an initial transient, a bell shaped temperature profile is attained, and cold and very lean mixtures can be conveniently processed. The main shortcoming of RFRs is the loss of reactants immediately upon flow reversal: the so-called washout effect. Matros (1989) has explored several other reactor configurations aimed at trapping reaction heat inside the catalytic bed and avoiding washout. One proposed configuration is a network (NTW) of two or three reactors with feed and discharge positions periodically shifted in time (Haynes and Caram 1994; Brinkmann et al. 1999). In a NTW, the heat front propagates in a virtually closed cycle while keeping constant the flow direction in each reactor. From the theoretical point of view, Sheintuch and Nekhamkina (2005) analyzed a NTW consisting of an arbitrary number of reactors, and showed that its performance converge to that of a continuum loop reactor as the number of reactors is increased.

When the NTW consists of fixed-bed reactors, the periodic forcing generates spatio-temporal patterns that are strictly related to the spatio-temporal symmetry of the system (Russo et al. 2006; Altimari et al. 2006). Indeed, it was demonstrated that periodic forcing of a NTW of identical reactors determines specific spatio temporal symmetries (Russo et al. 2002) which do not depend on the specific model used to describe the reactor. It was also shown that the symmetry properties of the system have substantial effects on the bifurcation behavior of the NTW.

In the present paper we consider a NTW of three fixed bed reactors forced with two different switch strategies characterized by different feed/discharge permutations. The analysis is aimed at evaluating how the switch strategy affects the NTW stability through simulations and bifurcation analysis. The first strategy, which was already considered in the literature, consists into changing at each switch time the feed/discharge positions so that the first reactor of the NTW is moved to the last place, whereas with the second strategy the last reactor is periodically moved to the first place of the NTW sequence.

2. Mathematical model equations and switch strategies

Two different switch strategies as sketched in Fig.1 are studied. In both cases the sequence of reactors is changed periodically following a cyclic permutation.
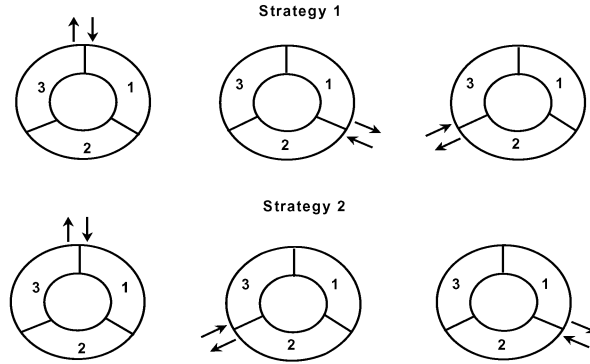

Figure1. The three catalytic reactors with two different strategies of permutation of the feed and discharge positions.

We consider a first order exothermic reaction occurring in a fixed catalytic reactor. Each fixed-bed reactor is modeled as a heterogeneous system with heat and mass transfer resistance between the gas and the solid phase, axial dispersion in the gas phase, axial heat conduction in the solid phase, and cooling at the reactor wall.
The mathematical model (Russo et al. 2006) for the NTW reads:

$$
\begin{cases}
\dfrac{\partial y_{g,i}}{\partial t} = \dfrac{1}{Pe_g^m}\dfrac{\partial^2 y_{g,i}}{\partial z^2} - \dfrac{\partial y_{g,i}}{\partial z} + J_g^m\left(y_{s,i}-y_{g,i}\right) \\[2ex]
J_s^m\left(y_{s,i}-y_{g,i}\right) = \eta Da\left(1-y_{s,i}\right)\exp\dfrac{\theta_{s,i}}{1+\theta_{s,i}/\gamma} \\[2ex]
\dfrac{\partial \theta_{g,i}}{\partial t} = \dfrac{1}{Pe_g^h}\dfrac{\partial^2 \theta_{g,i}}{\partial z^2} - \dfrac{\partial \theta_{g,i}}{\partial z} + J_h^g\left(\theta_{s,i}-\theta_{g,i}\right) - \varphi\left(\theta_{g,i}-\theta_{w,i}\right) \\[2ex]
\dfrac{\partial \theta_{s,i}}{\partial t} = \dfrac{1}{Pe_s^m}\dfrac{\partial^2 \theta_{s,i}}{\partial z^2} - J_s^h\left(\theta_{s,i}-\theta_{g,i}\right) + B\eta Da\left(1-y_{s,i}\right)\exp\dfrac{\theta_{s,i}}{1+\theta_{s,i}/\gamma}
\end{cases}
\qquad i=1,2,3. \qquad (1)
$$

In Eq.(1) the first two equations represent the mass balance in gas phase and in the solid phase, respectively. The third and the fourth equations are the heat balance in gas phase and in the solid phase, respectively. The index i identifies the reactor. The definition of all dimensionless state variables and parameters are the same of Russo et al. (2006). The forcing enters the model through the boundary conditions, which hence differentiate the two strategies. The forcing is implemented with a discontinuous periodic wave function $f(t)$ given by:

$$f(t) = \begin{cases} 0 & if \ \ 0 \leq \dfrac{t}{\tau}(\mathrm{mod}\,3) < 1 \\ 1 & if \ \ \ \dfrac{t}{\tau}(\mathrm{mod}\,3) > 1 \end{cases} \tag{2}$$

Thus, the boundary conditions reads:

$$\begin{cases} \left.\dfrac{\partial y_{g,i}}{\partial z}\right|_0 = \dfrac{-\left[1 \pm f\left(t-(i-1)\tau\right)\right]y_{in} - f\left(t \pm (i-1)\tau\right)y_{g,i-1}(1,t) + y_{g,i}(0,t)}{Pe_g^m} \\ \left.\dfrac{\partial \theta_{g,i}}{\partial z}\right|_0 = \dfrac{-\left[1 \pm f\left(t-(i-1)\tau\right)\right]\theta_{in} - f\left(t \pm (i-1)\tau\right)\theta_{g,i-1}(1,t) + \theta_{g,i}(0,t)}{Pe_g^h} \\ \left.\dfrac{\partial \theta_{s,i}}{\partial z}\right|_0 = 0 \end{cases} \tag{3}$$

$$\left.\dfrac{\partial y_{g,i}}{\partial z}\right|_1 = \left.\dfrac{\partial \theta_{g,i}}{\partial z}\right|_1 = \left.\dfrac{\partial \theta_{s,i}}{\partial z}\right|_1 = 0$$

Where the sign – should be used for the strategy-1 and + for the strategy-2.
It is worth remarking that the NTW mathematical model is a discontinuous periodically forced system with a minimal period $T=3\tau$ for both switching strategies.

3. Effect of the switch strategies

The effect of the switch strategy on the dynamic behavior of the NTW has been studied through spatio-temporal patterns and bifurcation analysis in the case of T-periodic regimes. The switch time is chosen as bifurcation parameter. Bifurcation analysis is carried out with a continuation algorithm that allows the construction of solution diagrams representing the locus of T-periodic regime solution. We implemented this procedure as described in details by Russo et al. (2002); this technique exploits symmetry properties (induced by the periodic forcing) of the model. Stability ranges of ignited T-periodic regimes and their bifurcations are thoroughly analyzed as the switch time is varied for both switching strategies. In such a way, ignition/extinction ranges of the forced network are efficiently detected.

3.1 Effect of the switch strategy on the spatio-temporal patterns.

The occurrence of temperature and conversion waves traveling along the NTW appears as T-periodic regimes.
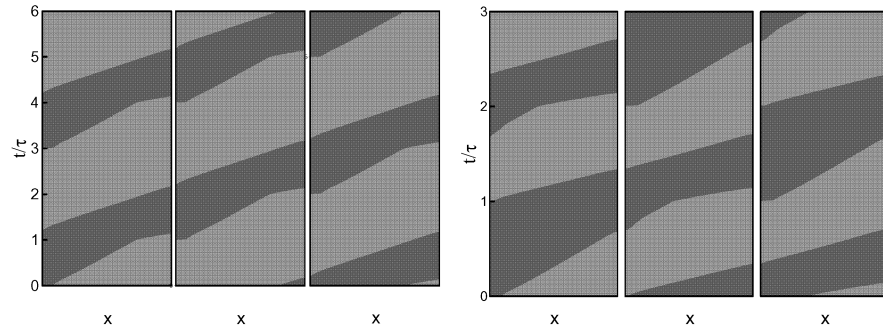

Figure 2. Spatio-temporal grey-scale patterns of the gas phase temperature. Left: strategy-1,T_{in}=60, τ=2500; Right: strategy-2, T_{in}=60 C, τ=5000. Each box corresponds to a reactor unit.

For both strategies, examples of traveling temperature waves are given by the spatio-temporal pattern reported in Fig. 2, where each panel represents a single reactor. The temperature level is grey coded, and the non-dimensional axial coordinate is reported in abscissa. The scaled time (t/τ) is in ordinate. For the sake of clarity, only two temperature levels are considered: light grey represents points with T>500C, and dark grey T<500C. For the strategy-1, it is then apparent the path of the temperature traveling wave through the NTW: the temperature front forms in the first reactor, and then moves along axially; after the first switch a new front forms and moves along the second reactor; then after the second switch the same happens in the third reactor. It is evident that the spatio-temporal patterns (Fig. 2-left) in any of the three reactor is equal to the spatio-temporal pattern in the successive (according the flow direction) reactor but shifted forward in time of τ. This feature is the manifestation of the spatio-temporal symmetry of the system (Russo et al. 2002).The differences in the spatio-temporal symmetries induced by the two forcing strategies are readily evident from the comparison of the spatio-temporal patterns of the T-periodic regime (Figs. 2). The situation for the strategy-2 (Fig.2-right) shows that the spatio temporal pattern in any of the three reactors is equal to the spatio-temporal pattern in the preceding (according the flow direction) reactor but shifted forward in time of τ.

3.2 Bifurcation analysis: The influence of the switch time τ.

Figure 3 shows solution diagrams for both strategies with the switch time as the bifurcation parameter. In these solution diagrams the locus of T-periodic regimes is shown. Stable solutions are represented with solid lines, and unstable solutions are plotted with dashed lines. The feed temperature value is 60 C. It should be remarked that at this feed temperature value the unforced system does not exhibit any stable ignited regime.
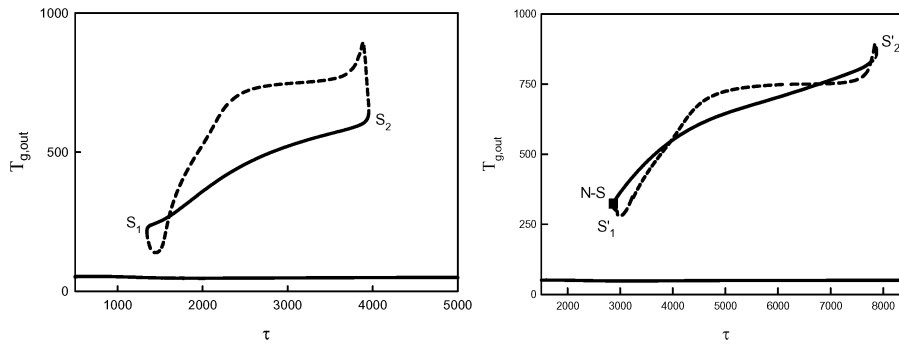
Figure 3. The symmetric *T*-periodic solution diagram with the switch time, τ, as bifurcation parameter at T$_{in}$=60 C. Left: strategy-1. Right: strategy-2. The solution is reported with the gas temperature at the network exit, T$_{g,out}$ C. Solid lines: stable *T*-periodic regimes; dashed lines: unstable *T*-periodic regimes.

For both strategies, a nonignited *T*-periodic regime is present throughout the investigated switch time range (the horizontal line on the bottom of Figs. 3). The ignited (high conversion) solutions form an isola bounded by two catastrophic saddle node bifurcation points: S$_1$ (at τ=1386) and S$_2$ (at τ=3950) for strategy-1 and S'$_1$ (at τ=2810) and s'$_2$ (at τ=7900) for strategy-2. When the switch time is increased above τ$_{S2}$ (or τ$_{S'2}$), forcing is not able to sustain auto thermal operation anymore, and, correspondingly, no high conversion solution is found. Conversely, for switch times lower than τ$_{S1}$ (or τ$_{S'1}$), *T*-periodic ignited solutions disappear but the system still possesses ignited solution until τ approaches to zero, which are stable multi-periodic, quasi-periodic and chaotic regimes. Those solutions can be detected via numerical simulation. A detailed description of the bifurcations occurring in this switch time range is beyond the scope of this work (Russo et al. 2006). For the strategy-2, the stable high conversion *T*-periodic regimes on the isola become unstable at τ=2870 through a Neimark-Sacker bifurcation N-S. This bifurcation is supercritical, and leads to the onset of stable symmetric quasi-periodic regimes. It is important to note the switch time value at which the Neimark-Sacker occurs is very close to the switch time value at which the saddle-node bifurcation S'1 occurs. The saddle-node bifurcation S'2 is catastrophic whereas S'1 is not. The most important difference of the solutions diagrams shown in Fig3. is the significantly larger stability range of *T*-periodic ignited solutions for the strategy-2 with respect to that predicted for strategy-1. In particular, the switch time range (τ∈[2810; 7900]) where stategy-2 exhibits stable traveling waves is almost doubled with respect to that found for strategy 1 (τ∈[1386, 3950]). This fact seems to suggest that an intelligent choice of the switch strategy can overcome the stability limitations of NTW reported in the literature. A physical interpretation of these limits is possible making use of the analytic criteria of Sheintuch and Nekhamkina (2005), who proposed and tested these criteria for a NTW of adiabatic reactors treated with a pseudo-homogeneous model. These criteria define the range of existence of traveling waves, and are based on the comparison of the thermal and reaction front velocity with the switch velocity, that is,

the velocity of the feed position movement. The range of existence of T-periodic ignited solution is defined by the following inequalities:

$$V_{fr} \le V_{sw} \le V_{th} \tag{4}$$

where V_{th}, the velocity of a thermal front in absence of reaction, V_{fr} the velocity of a reaction front that represents the velocity at which the reaction section moves along the reactor and V_{sw} is the switch velocity. V_{th} and V_{fr} are slightly affected by the switch strategy, as they mainly depend on the heat capacities of solid and gas, on the gas velocity, on the adiabatic temperature rise, and on the inlet temperature. On the contrary, the switch velocity does depend on the switch strategy, as $V_{sw} = n/\tau$, where n is the number of the reactor units jumped upon switching (in the flow direction) by the feed position. It can be easily noted that the value of n is 1 in the case of strategy 1, and 2 in the case of strategy-2. Thus the Eq.4 explains the doubling of the stability limits of the stategy-2 respect to the strategy-1.

Conclusions

We analyzed and compared the spatio-temporal patterns and the stability of T-periodic regimes of a network of three nonadiabatic catalytic fixed bed subjected to two different periodic switch strategies: one in which after one switch time the first reactor of the sequence is moved at the last place and the other in which after one switch time the last reactor of the sequence is moved at the first place. For both strategies, the switch time ranges of T-periodic regimes are delimited by saddle node bifurcations. Consistently with the theoretical predictions of Sheintuch and Nekhamkina (2005), obtained in adiabatic conditions, the bifurcation switch time values of the strategy-2 are doubled with respect to the strategy-1. This fact show that the switch strategy can be used to enlarge the narrow stability ranges of NTW.

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