

3-D TRANSVERSAL PATTERNS IN MOVING OR OSCILLATORY-FRONTS OF PACKED-BED REACTORS

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Summary

We derive criteria for emergence of patterns in the direction transversal to advection of planar fronts propagating in catalytic tubular bed reactors, in which an exothermic activated reaction is conducted. Patterns may emerge when $Pe_C/Pe_T < (1-V_f)/(1-LeV_f)$. The criterion is confirmed by simulations of the 2-D cylindrical shell and full 3-D models.

The study is extended to the case of an oscillatory kinetics and three distinct patterns are traced: rigid rotating, non-rotating transversal patterns superimposed on the axially oscillating front and mixed structures. The possible reduction of 3-D model to the 2-D cylindrical shell model is discussed.

Keywords

Fronts, advection-reaction-diffusion system, stability, transversal patterns.

Introduction

Heterogeneous catalysts are extensively used in chemical and petrochemical reactors and for reduction of environmental pollution generated by automobiles, electrical power stations and other stationary engines. It is usually assumed that the temperature and concentrations of the reactants at any transversal cross section of an adiabatic packed-bed reactor are uniform. However, various industrial and laboratory experimental observations revealed formation of non-uniform temperature and concentration in the reactor cross section. Such patterns may pose severe safety hazard when present next to the reactor wall. Understanding which operating conditions and reactions may lead to spatiotemporal pattern formation is essential for the development of control strategies that circumvent them. Significant modeling efforts have been directed to predict formation of transversal patterns during the last decade (see a recently published review¹). The analysis and simulations of a three-dimensional model with complex kinetics is very intricate. Thus, previous studies have been conducted with simplified models using several assumptions concerning either the physical and kinetics models or geometrical symmetry assumed.

In the present study we consider patterns formation in 3-D PBRs using two different kinetic models: i) the generic kinetics that accounts for a single exothermic reaction with Arrhenius kinetics and is described by two state variables: the temperature (y) and the concentration of the limiting species (x); ii) the minimalistic model that accounts for an oscillatory behavior and is constructed by coupling such a

two-variable model with a third variable (θ) that describes a reversible change of a catalytic activity. Both models are governed by the following balance equations:

$$Le y_t + y_z - \Delta y / Pe_T = B f(x, y, \theta); \quad x_t + x_z - \Delta x / Pe_C = f(x, y, \theta); \\ f = Da(1-x)\theta \exp(y/(1+y/\gamma)) \quad (1)$$

Here Pe_C , Pe_T are the mass and heat Pe numbers and $Le \gg 1$.

Regular kinetics ($\theta = \text{const} = 1$)

With the two-variable thermo-kinetic model ($\theta = 1$) the 1-D systems may exhibit a stable "frozen" reaction front propagating with a constant velocity. For this case we have recently showed² using a linear stability analysis that a planar front may bifurcate into non-rotating transversal patterns when

$$Pe_C/Pe_T < (1-V_f)/(1-LeV_f) \quad (2)$$

where V_f is the front velocity. A similar condition was obtained using an alternative approach based on the relation between the front velocity (V_f) and the local curvature (K) using the critical condition $dV_f/dK|_{K=0} = 0$ and a simplified model describing two 1-D reactor channels with heat and mass exchanges between them². For stationary fronts criterion (2) implies that $Pe_C/Pe_T < 1$, which is exactly the Turing condition. Since in practice $Pe_C/Pe_T > 1$, this novel condition can be satisfied only for an upstream propagating front (toward the cold reactor inlet).

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Criterion (2) was verified by numerical linear stability analysis of certain 1-D front solutions. The constructed bifurcation diagrams show, that the planar front is unstable with respect to small perturbations with small wave number ($k \sim 0$). Direct numerical simulations of a cylindrical shell model² as a learning model show formation of stable non-rotating transversal patterns on a shell of large diameter ($D > L$) and transient structures with small D of practical interest. Here we present complex non-rotating patterns with the full 3-D reactor model.

Oscillatory kinetics ($\theta = \text{var}$)

With the three-variable kinetics the system can exhibit a 1-D oscillating front. In such a case the basic 1-D solution varies both with time and with space. Any analytical results concerning the front velocity, the maximal temperature rise to the best of our knowledge were not published yet. Numerical simulations using the three-variable (x, y, θ) kinetics of the learning 2-D cylindrical shell³ and the full 3-D reactor model revealed that symmetry breaking of a planar front can emerge even with $Pe_C/Pe_T \gg 1$.

Three types of patterns were detected in simulations: rigid rotating fronts, oscillating fronts with superimposed (non-rotating) oscillations and mixed rotating-oscillating fronts. Non-rotating patterns. Typical non-rotating patterns simulated in a 2-D case are presented in Fig.1 showing several temperature snapshots on an unfolded surface. The transversal front divergence gradually enlarges and shrinks during the downstream and upstream section of the front motion and reaches its minimum close to points of the front direction reversal. At these points the cold- and the hot domains are interchanged forming antiphase oscillations. In the 3-D case such type of patterns resemble the transversal eigenfunctions which are the Bessel functions of the 1-st kind (due to no-flux boundary conditions) multiplied by periodic functions in the azimuthal direction.



Fig.1 Typical non-rotating patterns superimposed on an axial oscillating front in a 2-D cylindrical shell model with oscillatory kinetics showing several temperature snapshots during one period.

Rotating patterns. We detected both one-wave and multiperiodic rotating patterns in simulations of the cylindrical shell model². To describe the 3-D patterns we defined within the each cross-section the tip as a point of maximal gradients of the state variables. To classify patterns we traced the tip motion in the cross-section around the front position. Rigid rotating fronts exhibit simple circles of the tip in the transversal cross-section. Mixed rotating-oscillating patterns appear as rotations superimposed on the axial front oscillations. The

corresponding tip motion exhibit meandering behavior which reflects the coexistence of several frequencies.

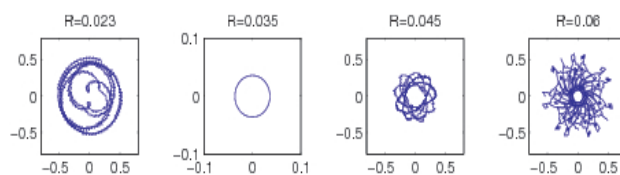


Fig.2 Typical rotating ($R=0.035$) and mixed rotating patterns in a 3-D model with oscillatory kinetics showing effect of radius on the tip motion in the reactor cross-section.

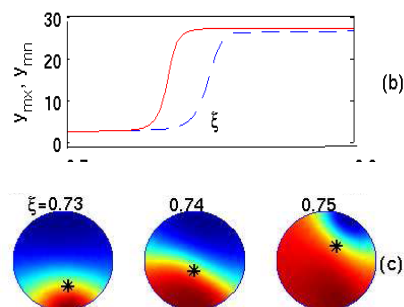


Fig.3 Typical rigid rotating patterns in a 3-D model with oscillatory kinetics showing the axial profiles of the maximal and minimal temperature (b) and several axial snapshots (c) that rotate with time.

We map bifurcation diagrams that account for the two main (rotating and non-rotating) patterns using the reactor radius as a bifurcation parameter. The pattern selection crucially depends on the choice of initial conditions.

The possible reduction of the 3-D model to the 2-D cylindrical shell model is discussed.

References

- (1) Viswanathan, G.A. Sheintuch, M.; Luss, D. Transversal Hot Zones Formation in Catalytic Packed Bed Reactor, *Ind. Eng. Chem. Res.* **2008**, 47, 7509.
- (2) Nekhamkina, O.; Sheintuch, M. Transversal moving-front patterns: Criteria and simulations for two-bed and cylindrical shell packed-bed reactors. *Chemical Engineering Science* **2008**, 63, 3716
- (3). Sheintuch, M., Nekhamkina, O.; Thermal patterns in simple models of cylindrical reactors. *Chemical Engineering Science* **2003**, 59, 1441.

