Wall Temperature Modulates Transversal Pattern Formation and Dynamics in Shallow, Non-adiabatic Packed-bed Reactors

K Narendiran and Ganesh A Viswanathan*
Department of Chemical Engineering
Indian Institute of Technology Bombay
Powai, Mumbai – 400076, India
*Corresponding author (Email: ganeshav@iitb.ac.in)

Abstract

Transversal temperature pattern formation has been observed in catalytic packed-bed reactors used for conducting exothermic reactions. These patterns or non-uniform states can strongly affect the performance of the reactor and pose severe safety issues. Recent studies show that, for a fixed wall temperature, small wall heat transport is sufficient to trigger a transition between characteristically different transversal spatiotemporal patterns in a shallow reactor. We ask a question as to how wall temperature may influence selection and dynamics of spatiotemporal patterns exhibited by a shallow reactor used to conduct ethylene hydrogenation reaction governed by periodic blocking-reactivation kinetics. Using a fitting combination of linear stability analysis and extensive numerical simulations, we predict the dependence of wall temperature on different types of targets and rotating patterns that may form (and co-exist) in the shallow reactor. We compare the dependency of these two varieties of spatiotemporal patterns on the reactor diameter.
Keywords: Non-adiabatic packed-bed reactor, Spatiotemporal temperature pattern formation, Heat-exchange dynamics, Targets, Spirals

Introduction

Formation of spatial and spatiotemporal patterns or non-uniform states has been observed in several industrial and laboratory catalytic packed-bed reactors (PBRs). Presence of a non-uniform state can pose safety concerns, can have detrimental effects on the performance and may trigger undesired side-reactions affecting product selectivity. Understanding spatiotemporal pattern formation may offer insights to circumvent them during regular operation by appropriately tweaking the operating conditions.

Several experimental and theoretical studies have been conducted to detect and predict the formation of spatiotemporal patterns. Experimental methods typically include monitoring the temperature by measuring it in appropriate locations using thermocouples or infrared camera (Marwaha and Luss, 2003; Digilov *et al*., 2004]. Laboratory shallow reactor studies were conducted by placing alumina wool that, at best only minimizes the heat transport from the reactor to the surroundings (Marwaha and Luss, 2003). Theoretical studies to predict these were performed by assuming perfect adiabatic conditions (Viswanathan and Luss, 2006; Balakotaiah *et al*., 1999; Agarwal *et al*., 2007; Nekhamkina and Sheintuch, 2014).

Objectives

Real reactors are seldom adiabatic. Heat exchange between the reactor and surroundings is inevitable. Thus, there is a need to understand how heat exchange may affect the spatiotemporal pattern formation. Dimensionless wall heat transport rate depends on Biot number or Bi and the dimensionless wall temperature θ_w. We recently showed that small non-zero Bi at constant θ_w is sufficient to drastically alter pattern formation in a shallow reactor, whose length to diameter ratio is small (Narendiran and Viswanathan, 2015). In this study, we consider the effect of wall temperature on the spatiotemporal pattern formation in a shallow, non-adiabatic PBR.

Mathematical model

In this study, we model a shallow reactor for investigating the spatiotemporal pattern formation and dynamics during a single catalytic ethylene hydrogenation reaction

* To whom all correspondence should be addressed

governed by a blocking-reactivation kinetic mechanism. Shallow reactor, an asymptote of a tall reactor, has been used, in both theoretical and experimental approaches to study pattern formation in PBRs under various conditions (Balakotaiah *et al*., 2002, Narendiran and Viswanathan, 2015). Assuming ethylene is present in excess and first-order with respect to hydrogen, we write a mathematical model capturing dynamics of local temperature, conversion and catalyst activity.

Results

We conduct linear stability analysis by introducing azimuthally non-uniform perturbations to the (symmetric) base states of the model and map regions in parameter space where symmetry-breaking spatiotemporal patterns may form. Figure 1 shows, for R/d_p=10, the Hopf neutral stability curves in the planes of Da and $\theta_w (=T_w/T_{in})$ for the symmetric base state (*m*=0) and the first three azimuthal modes (*m*=1,2,3). Neutral stability curves show that the region where symmetry-breaking patterns may form for different types of perturbations increases with increasing wall temperature suggesting that θ_w may have a strong role to play in the pattern formation and dynamics.

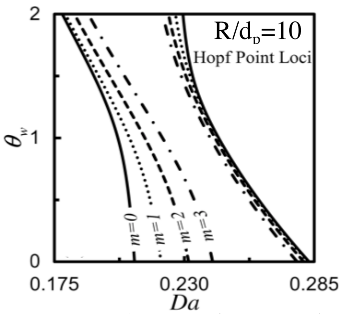

Figure 1: Hopf neutral stability curves for the base state (m=0) and the first three azimuthal modes.

Since regions in the parameter space bounded by the neutral stability curves are only indicative of pattern formation, we considered prediction of the actual spatiotemporal patterns. Eigenfunctions and neutral stability curves guided initial conditions were used for extensive numerical simulations of the model to predict a rich variety of two classes of spatiotemporal patterns, viz., targets and rotating patterns. In Fig. 2, we show dynamics of typical rotating and target patterns for one oscillation period for a certain set of parameters.

We next consider the effect of wall temperature on these two classes of spatiotemporal patterns. We predict in parameter space, regions where different targets/rotating patterns were found or where both target & rotating patterns co-existed. In Fig. 3, for the sake of brevity, we only show the partitions in the plane of Da vs θ_w where different target patterns exist.

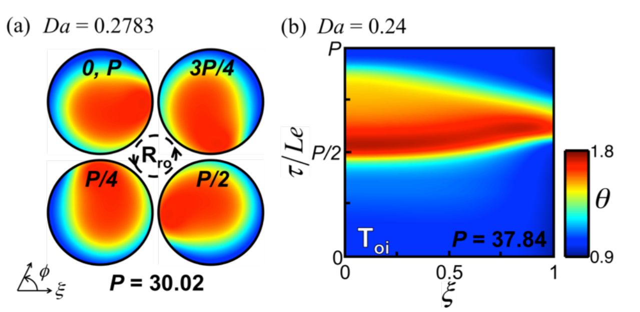

Figure 2: (a) Snapshot of the rotating patterns at 0, P/4, P/2, 3P/4, P time points in one period (P). (b) Spatiotemporal dynamics of outward-inward moving targets. R/d_p=10, θ_w=0.1.

Conclusions

We show that wall temperature for a certain reactor diameter may modulate the spatiotemporal target and rotating patterns observed during catalytic ethylene hydrogenation reaction governed by blocking-reactivation kinetics occurring in a shallow, non-adiabatic packed-bed reactor. A combined effect of the reactor diameter and the wall temperature on the targets and rotating patterns will also be presented.

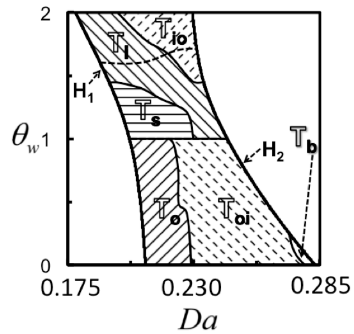

Figure 3: Regions where different target patterns exist. $\mathbb{T}_i$ *– inward moving,* $\mathbb{T}_{io}$ *– inward-outward moving,* $\mathbb{T}_s$ *– standing wave,* $\mathbb{T}_o$ *– outward moving,* $\mathbb{T}_{oi}$ *– outward-inward moving,* $\mathbb{T}_b$ *– breathing.* H_1 *and* H_2 *are the Hopf point loci of the base symmetric state.*

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