

# EFFECT OF NON-UNIFORM CATALYST LOADING ON STATIONARY AND MOVING TEMPERATURE FRONTS IN CATALYTIC MONOLITH REACTORS

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## Summary

We study the effect of non-uniform catalyst distribution along the length of the channel on the performance of the monolith catalytic reactors using a one dimensional two-phase model with position dependent heat and mass transfer coefficients. We present the steady-state bifurcation diagrams illustrating the effect of different non-uniform catalyst distributions, solid phase conductivity and the flow conditions near the inlet to the channel on the behavior of the monolith. It is shown that the ignition and extinction points are very sensitive to the catalyst distribution, solid conductivity as well as the correlations used to calculate the heat and mass transfer coefficients.

## Keywords

Novel reactor technologies, Dynamics and control of chemical reacting systems

## Introduction

Monolith catalytic reactors are used widely in automobile emission control. At present, the required conversion of pollutants is greater than 95%. Approximately 50-80% of emissions from a light duty automobile on an average journey occur before the cold converter ignites, which takes about one minute. An almost instantaneous light-off of the converter is required to meet stringent EPA regulations in the coming years. Many alternative designs like hydrocarbon traps, electrically heated catalytic converters, and pre-igniters have been proposed to reduce the cold start emissions. The main drawback of these techniques is the requirement of additional equipment. To avoid this, some researchers have proposed using non-uniform distribution of the catalyst along the length of the channel (or several bricks with different precious metal loading) to reduce the light off time. In this work, we investigate in some detail, the effect of non-uniform catalyst distribution, solid phase conductivity and the flow development length on the steady-state and transient (light-off) behavior of the monolith catalytic reactor. It is hoped that the results and the analysis presented here will be useful in the selection of optimum designs for various applications of monolith catalytic reactors.

## Mathematical Model

We consider a one dimensional two-phase mathematical model of a straight channeled, washcoated catalytic monolith with the following assumptions:

i) conduction/diffusion in the axial direction is negligible in the fluid phase, ii) laminar flow in the channel with negligible pressure drop, iii) constant physical properties, iv) uniform cross sectional area, v) a single first order irreversible exothermic reaction of the type  $A \rightarrow B$  occurs only in the washcoat and catalyst activity remains constant with time. With these assumptions, the mathematical model in dimensionless form is described by the following equations and boundary conditions:

$$\frac{\partial C_m}{\partial \tau} = -\frac{\partial C_m}{\partial z} - a(z) Da r_e(C_m, \theta_s), \quad (1)$$

$$\frac{\partial \theta_m}{\partial \tau} = -\frac{\partial \theta_m}{\partial z} + Nu_\Omega(z) \frac{Le_f}{P} (\theta_s - \theta_m), \quad (2)$$

$$\sigma \frac{\partial \theta_s}{\partial \tau} = \frac{1}{Pe_h} \frac{\partial^2 \theta_s}{\partial z^2} - Nu_\Omega(z) \frac{Le_f}{P} (\theta_s - \theta_m) + a(z) B Da r_e(C_m, \theta_s), \quad (3)$$

$$c_m(0, \tau) = c_{in}(\tau), \quad \theta_m(0, \tau) = \theta_{in}(\tau),$$

$$\partial \theta_s(0, \tau) / \partial z = \partial \theta_s(1, \tau) / \partial z = 0,$$

where  $r_e(C_m, \theta_s)$ , the effective dimensional reaction rate, is given by

$$r_e(C_m, \theta_s) = \frac{C_m X}{\frac{\Phi(z)}{\tanh \Phi(z)} + \frac{\Phi(z)^2}{\Lambda Sh_\Omega(z)}}, \quad (4)$$

where

$$\Phi(z) = \phi_s \sqrt{a(z) \Lambda X}; \quad X = \exp\left(\frac{\theta_s}{1 + \theta_s / \gamma}\right).$$

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Here,  $a(z)$  is the activity profile along the length. We refer to Ramanathan et al.<sup>1</sup> for a detailed explanation of dimensionless parameters.

## Results and Discussion

Steady state bifurcation diagram with non-uniform catalyst loading (two zone catalyst distribution where the initial 10% of the channel length has 52.6% of the catalyst) for three different heat and mass transfer coefficient correlations (we refer to Gupta and Balakotaiah<sup>2</sup> for complete description of heat and mass transfer correlations) is shown in Fig. 1. We see that both ignition and extinction points are very sensitive to the type of correlation used. We observe that only the ignition point is sensitive in the case of uniform catalyst loading. Hence, the details of the flow development are important for the case of non-uniform catalyst loading to correctly predict the monolith behavior.

Fig. 2 compares the steady-state bifurcation diagrams with uniform and non-uniform catalyst loading for hydrogen oxidation. We see that non-uniform catalyst loading eliminates the hot spot which exists for uniform loading. The effect of solid heat conduction on the monolith behavior is shown in Fig. 3 for two zone distribution of the catalyst.

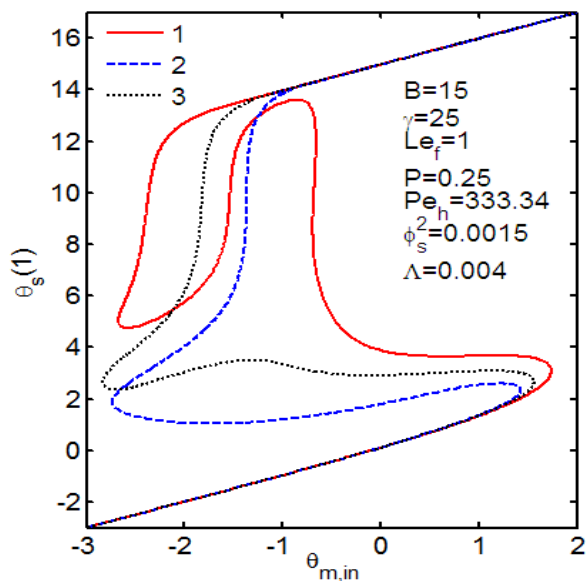


Fig. 1. Steady-state bifurcation diagram of the exit monolith temperature versus inlet fluid temperature with non-uniform catalyst loading. (Curves 1 and 3 represent position dependent transfer coefficients for fully developed and developing flow, respectively. Curve 2 represents constant transfer coefficients.)

The number of steady-state solutions and their stability varies depending on the catalyst distribution and other system parameters. We present the detailed analysis of the effect of various parameters and different catalyst distributions on the steady state and transient performance (moving temperature fronts) of the monolith reactor in the full length paper.

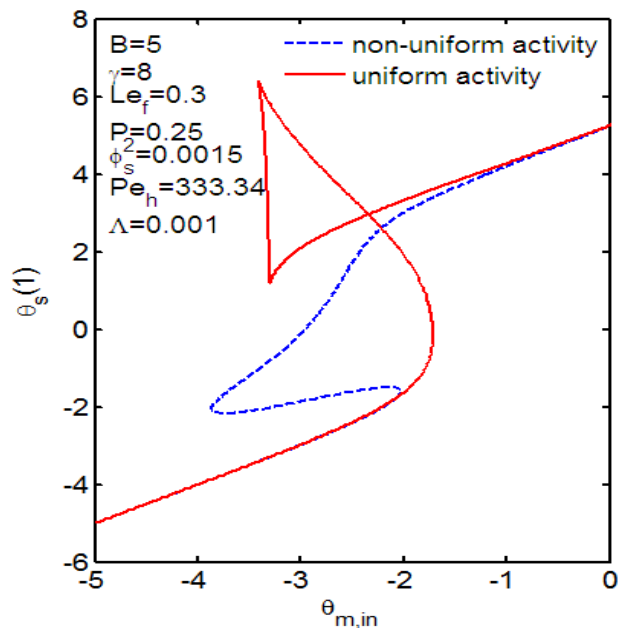


Fig. 2. Steady-state bifurcation diagrams illustrating the effect of non-uniform catalyst loading on the behavior of monolith for the case of fluid Lewis number less than unity (Hydrogen oxidation).

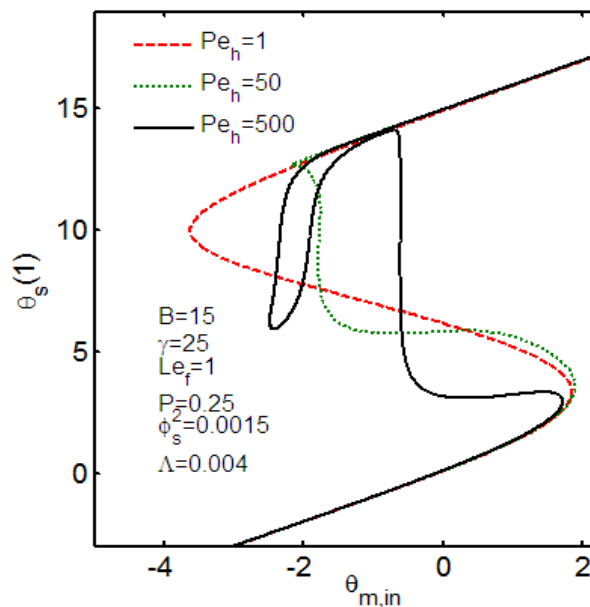


Fig. 3. Steady-state bifurcation diagram illustrating the effect of solid heat conduction on the behavior of monolith with non-uniform catalyst loading.

## References

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