Analysis of the dynamics of pseudohomogeneous chemical reactor by DAE model

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1 Introduction

Dynamical analysis of pseudohomegeneous reactors with thermal feedback was presented by a number of authors, among others [1,2]. It was shown that in autothermal systems various dynamical behaviors like periodic, multiperiodic or chaotic oscillations could take place, as well as different routes to chaos under specific work conditions can occur. In most cases numerical analysis is necessary for modeling the dynamical behavior of these chemical reactors. Unfortunately, engineering computations necessary for designing and controlling these systems are very time-consuming and this creates the needs to find faster and more efficient numerical methods to solve the differential equation systems which describe these models. Different way of simplification and optimization of design of chemical reactors can be found in the literature [3]. In the present work, a semi-analytical method is adopted for investigating the dynamics of autothermal pseudohomogeneous chemical tubular reactor, with thermal feedback realized as tubular external heat exchanger. For this system, it was observed that increasing the value of a parameter related to the thermal capacity, namely the Lewis number, causes appearance of stable oscillations. More precisely the phenomenon of "relaxation oscillations" takes place. This phenomenon was observed in different types of dynamical systems, among others in chemical reactors [4]. It was shown earlier [5] that, for sufficiently high values of the Lewis number, the differential balance equation system can be replaced by a differential-algebraic equation system (DAE) by the CSTR cascade approximation and the Damköhler number was explored as the bifurcation parameter. In the present work, the approximate DAE model is employed for computations and the influence of the influence of parameters f, which corresponds to heat recycle factor, β, which corresponds to dimensionless heat of reaction, and γ, which corresponds to dimensionless activation energy, is studied.

2 Mathematical background

The model of a pseudohomegeneous tubular reactor with heat recycle and axial mass dispersion is considered (Figure 1).

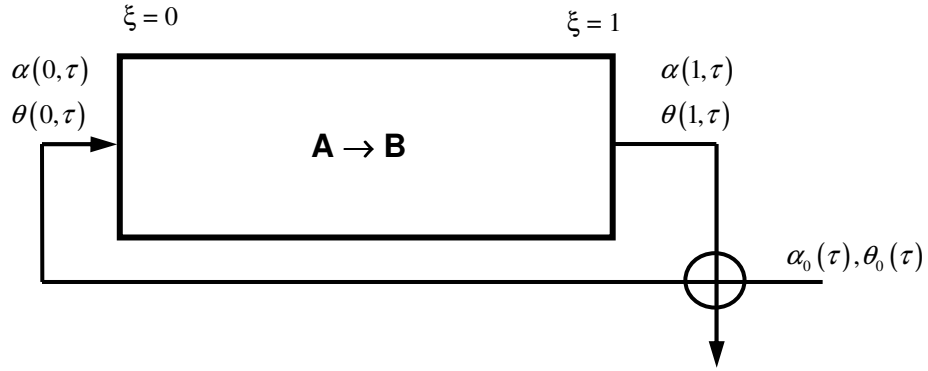

Figure 1. Schematic diagram of autothermal reactor with external heat exchanger

The model consists of the following mass and energy balance equations:

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$$\frac{\partial \alpha}{\partial \tau}+\frac{\partial \alpha}{\partial \zeta}=\frac{1}{Pe_M}\frac{\partial^2 \alpha}{\partial \xi^2}+Da(1-\alpha)^n \exp\left(\gamma\frac{\beta\theta}{1+\beta\theta}\right) \tag{1}$$

$$Le\frac{\partial \theta}{\partial \tau}+\frac{\partial \theta}{\partial \xi}=\frac{1}{Pe_H}\frac{\partial^2 \theta}{\partial \xi^2}+\mathrm{Da}(1-\alpha)^n \exp\left(\gamma\frac{\beta\theta}{1+\beta\theta}\right) \tag{2}$$

with boundary conditions:

$$\alpha(0^+)=\frac{1}{Pe_M}\frac{d\alpha}{d\xi}\Big|_{\xi=0^+}\ ;\ \theta(0^+)=f\theta(1)+\frac{1}{Pe_H}\frac{d\theta}{d\xi}\Big|_{\xi=0^+} \tag{3a,b}$$

The differential equation system can be reduced by perturbation. In the limit of $Le\to\infty$, i.e. $1/Le\to 0$, dimensionless time can be rescaled as:

$$\tau_{Le}=\frac{\tau}{Le} \tag{4}$$

After a substitution and some simple rearrangements, the reduced form of the model of the autothermal pseudohomegeneous tubular reactor with feedback is described by the following balance equations:

$$\frac{\partial \alpha}{\partial \xi}=\frac{1}{Pe_M}\frac{\partial^2 \alpha}{\partial \xi^2}+Da(1-\alpha)^n \exp\left(\gamma\frac{\beta\theta}{1+\beta\theta}\right) \tag{5}$$

$$\frac{\partial \theta}{\partial \tau_{Le}}+\frac{\partial \theta}{\partial \xi}=\frac{1}{Pe_H}\frac{\partial^2 \theta}{\partial \xi^2}+Da(1-\alpha)^n \exp\left(\gamma\frac{\beta\theta}{1+\beta\theta}\right) \tag{6}$$

whereas boundary conditions (3a,b), being time-independent, remain unchanged. It was shown [5] that for relatively high values of the Lewis number ($Le>10$), the reduced model (5-6) could be used instead of the full differential model (1-2). Moreover, an approximation of the reduced model (5-6) by cascade of CSTR, for first-order reaction, i.e. n=1, allows for a semi-analytical solution of differential-algebraic equation system (DAE) which is generated by that kind of approximation.

3 Results

The CSTR method was chosen to approximate the reduced system (5-6) valid in the limit of $Le\to\infty$. The following values have been assigned to the other parameters: Pe_M=Pe_H=100, γ=12.0, f=0.2, β=0.75 and n=1.0. Figure 2 reports a bifurcation diagram of steady–state solutions with branches of extrema of periodic solutions, showing the relationship between the dimensionless temperature θ at the outlet and the Damköhler number Da.

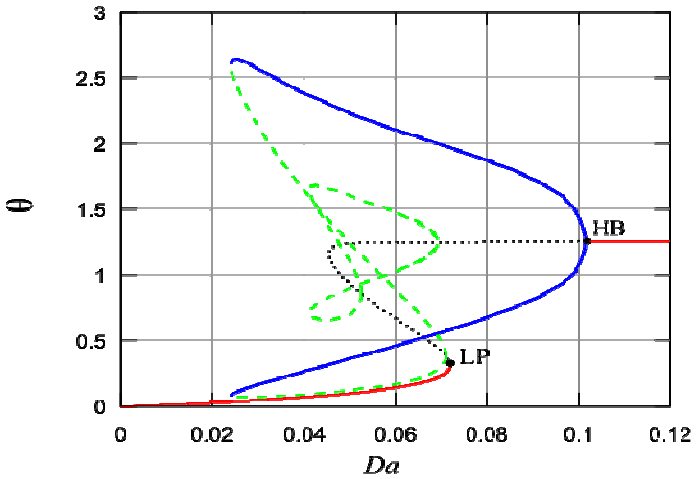

Figure 2. Diagram of steady-state solutions obtained by DAE system, where HB denotes the Hopf bifurcation while LP denotes the limit point.

The following notation is used for different types of solutions: red line for stationary states, black dots for unstable steady–states, blue line for stable periodic solutions, and green line for unstable periodic solutions. Two characteristic points, where changes of stability of dynamical solutions take place, are distinguished on this diagram (Fig. 2). Particularly, the Hopf bifurcation (HB), which causes the only stable solution change between stationary state and periodic, as well as the limit point (LP), where stability is lost and the branch of stable stationary states becomes unstable. LP corresponds to Da=0.071982, whereas HB corresponds to Da=0.101368.

Numerical analysis executed by the DAE system allows for the determination of boundaries between regimes of stable stationary states and regimes of periodical solutions. Locations of both the Hopf bifurcation (HB) and the limit point (LP) detected in the system were traced as a 2-parameter loci. Figures 3 show the relationships between γ and other parameters, namely the Damköhler number Da, the recycle coefficient f and the coefficient related to dimensionless adiabatic temperature increase β. Lines presented on the plots correspond to loci of the Hopf bifurcation (solid line) and the limit point (dashed line).

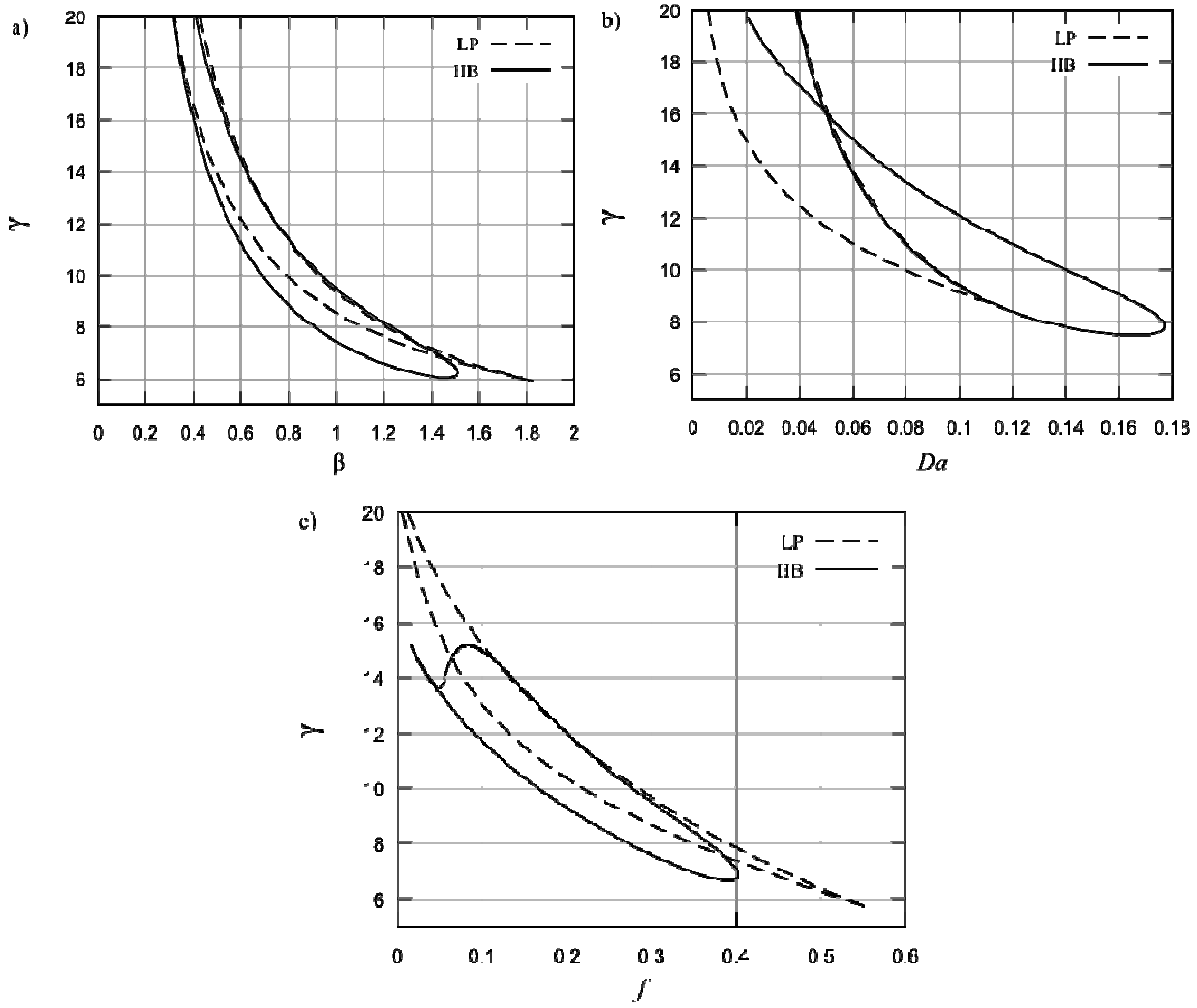

Figure 3. Bifurcation diagrams obtained by DAE system; (a) the γ-β plane, f=0.2, Da_{LP}=0.071982, Da_{HB}=0.101368; (b) the γ-Da plane, β=0.75, f=0.2; (c) the γ-f plane, β=0.75, Da_{LP}=0.071982, Da_{HB}=0.101368.

As seen, the model produces solutions in the range of γ-values much wider than those met in practice. For this reason, the range of this parameter is shown just to γ=20. It could be observed that decreasing of each of

these three mentioned parameters causes both types of bifurcations to appear for larger values of γ. However, an interesting structure could be observed in the plot γ vs. f (Fig. 3c), where the locus of the Hopf bifurcation creates a loop in the phase–space, while branches given by tracing of the limit point approach higher and higher γ–values when f–values get smaller. In each case two regions delimiting periodical solutions appear in the parameter space. Figure 4 reports a 3D bifurcation diagram summarizing the overall aspect of the oscillation region in the three parameter space.

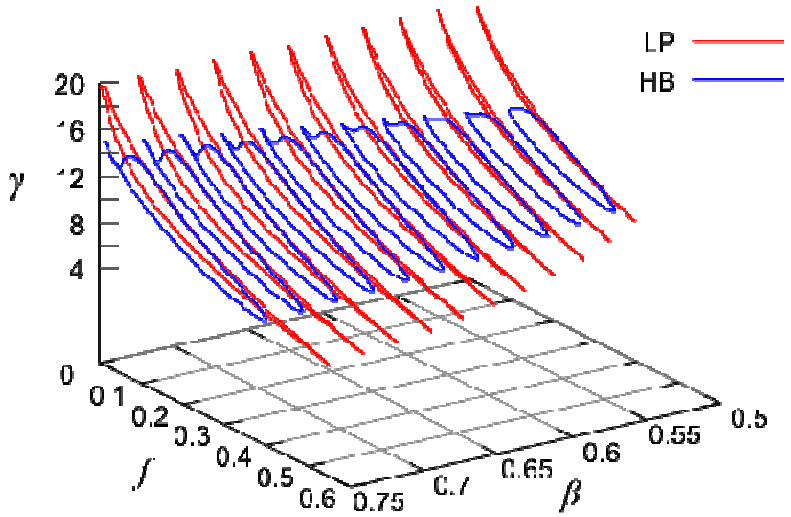

Figure 4. Three-dimensional bifurcation diagrams obtained by DAE system.

4 Conclusions

In the present work, the dynamics of autothermal pseudohomegeneous chemical reactor was analyzed. The DAE system, which allows for meaningful reduction of computational time, was applied to numerical computations. Relationships between dimensionless activation energy γ and other parameters were studied. The 2-parameter bifurcation diagrams allows for determination of subspaces where periodical solutions take place.

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