Dynamic Steady States of Distributed-Parameter Models of Self-Igniting Packed Beds

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Abstract

The dynamic steady-states of reaction-diffusion systems, constructed as models of reacting packed beds capable of self-ignition, are described by employing the direct simulation of PDE-based models, and, when possible, characterised via a parameter continuation approach. This combined approach permits to analyse and accurately describe a period-doubling cascade, and to consider the problem of the determination of different routes to chaos. Multiplicity of dynamic steady-states is observed, with coexistence of torus doubling sequences and of period-adding bifurcation sequences.

Introduction

The problem of spontaneous ignition in packed beds has attracted the attention of the scientific community for many decades. Rather recently, several papers have been devoted to the problem of spontaneous combustion of coal stockpiles [1,2]. Our group has studied and characterised some dynamical models of the self-ignition of coal stockpiles [3-5]. It was observed that, when natural convection is neglected, the system may show complex oscillatory patterns with possible onset of chaos. In [5] we estimated the power spectra, the Lyapunov Characteristic Exponents (LCE) and the dimensions of the attractors. In [3] a first bifurcation diagram was determined by means of parameter continuation. Later on, mixed-mode oscillations were detected for a model incorporating natural convection within the stockpile [6], and for a reaction-diffusion model of a self-igniting packed bed, with no convection, and in association with multiplicity of dynamic steady states and chaos [7]. The so-called complex or mixed-mode oscillations, a peculiar kind of dynamical asymptotic response, have been observed in connection with various chemical processes, particularly in biochemical systems [8,9], electrochemical models [10], non-isothermal catalytic systems [11,12]. A mechanism of bifurcation originating these oscillations has not been identified yet. Rather recently, Abashar and Elnashaie [13], for a non-isothermal fluidised bed catalytic reactor, and Milik et al. [14] for an autocatalytic system, also detected mixed-mode oscillations and explained the period-adding mechanism by analogy with some appropriate one-dimensional maps.

The analysis of bifurcation and onset of chaos for distributed parameter systems is a nontrivial issue. In many cases the problem, governed by a system of partial differential equations, may be usefully recast in terms of a large ODE problem. However, only if the number is not too large an automatic bifurcation analysis can be conducted and significant theoretical results can be obtained. In the others, the analysis can still be done with brute force, that is to use numerical simulations as if they were experiments. Some of the mentioned reduction techniques and examples of application of them, and of numerical simulations when the systems are still too complex, are described and discussed in the present communication, along with some interesting results.

Non-Isothermal Reaction-Diffusion Models

The self-ignition of a packed bed can be described by means of a distributed-parameter model. Such systems can get very complex. For example, coal stockpiles may self-ignite if reaction of coal with oxygen in the air generates heat which is not efficiently removed towards the external ambient. The heat production determines temperature gradients across the pile, so that a driving force for the onset of natural convection exists. The natural convection has a significant effect on the ignition itself, since it pushes air into the pile thus feeding reactant to the system, but, simultaneously, it cools down the coal pile with air at ambient temperature. The dynamic behaviour of a coal pile modelled as a two-dimensional system shows an interesting and complex dynamic evolution [6]. A rich dynamic behavior is however exibited by a simpler reaction-diffusion model where the gaseous reactant diffuses through the reacting medium along the one space coordinate considered. Consumption of the solid reactant is neglected. A first-order one-step exothermic chemical reaction takes place. The reaction rate depends on the temperature through the classic Arrhenius exponential.

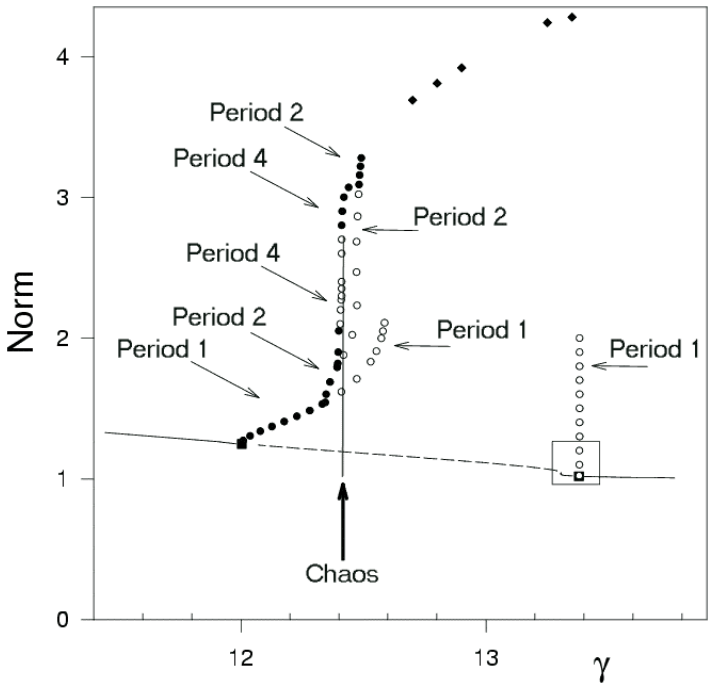

Figure 1 - Solution diagram for the system as determined via continuation with γ as bifurcation parameter. Lines represent static steady states (solid=stable, dashed=unstabe), circles represent periodic solutions (filled=stable, empty=unstable). Diamonds represent stable periodic solutions computed via simulation

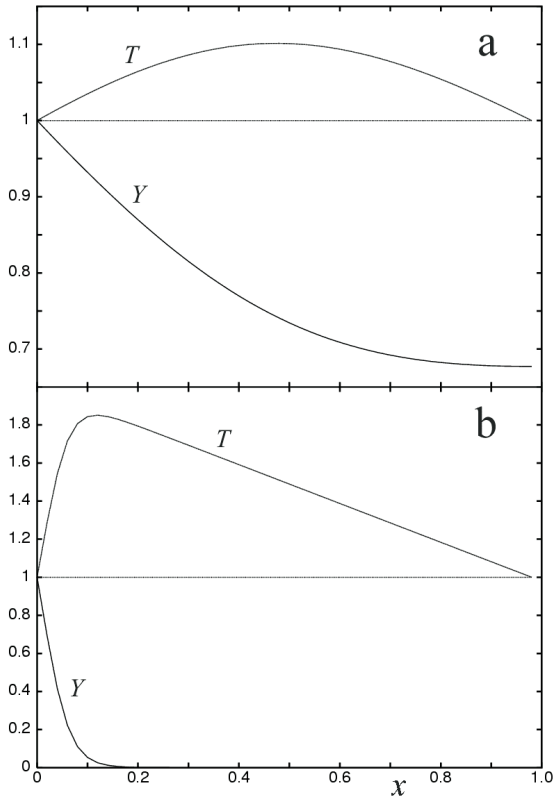

Figure 2 - Temperature (*T*) and concentration (*Y*) profiles along the x coordinate, for the two typical equilibrium solutions, on each of the two sides of the main stable solution branch: **a**) fast reaction regime, γ=10; **b**) slow reaction regime, γ=13.6.

Gas and solid temperature are equal, i.e. only one energy balance equation is written, considering heat conduction and a source term due to the reaction. The model equations, made non dimensional, are:

$$\frac{\partial Y}{\partial t} = \mathrm{Le}\frac{\partial^2 Y}{\partial x^2} - \Phi^2 Y \exp\left(-\frac{\gamma}{T}\right)$$

$$\frac{\partial T}{\partial t} = \frac{\partial^2 T}{\partial x^2} - \beta\Phi^2 Y \exp\left(-\frac{\gamma}{T}\right)$$

with boundary conditions: $T(0,t) = T(1,t) = 1$; $Y(0,t) = 1$; $\partial Y/\partial x\big|_{x=1} = 0$ for $t > 0$. The problem is completed with a set of initial conditions: $T(x,0) = T_0(x)$, $Y(x,0) = Y_0(x)$. Here Y is the gas reactant mass fraction, T is the temperature, t is the time coordinate, x is the space coordinate, Le is the ratio between mass and heat diffusivities, β is a nondimensional heat of reaction, Φ is the Thiele modulus, and γ is a nondimensional activation energy. The model equations were reduced to a set of ODE through orthogonal collocation.All the reported results were computed with 12 collocation points.

Results and discussion

In order to characterise in more detail the global behaviour of the model describing such a phenomenon, an attempt has been made to trace out some solution diagrams, via continuation, as the parameter γ (the non dimensional activation energy) is varied. The diagram in Fig. 1 is obtained by setting the other parameters to the following values: Le=0.233, Φ^2 =70000, β =4.287. It is possible to observe four bifurcation points on the main continuation branch: two static bifurcations, of the saddle-node kind, for γ=13.3047 and γ=13.3056, and two dynamic (Hopf) bifurcations for γ=13.3783 and γ=12.033. An equilibrium point for γ<12.033 corresponds to a fast reaction regime; an equilibrium point for γ>13.3783 corresponds to a slow reaction regime. The distribution of the state variables along the space coordinate are reported for the two cases in Fig. 2. Back to Fig. 1, from left to right, the first bifurcation encountered (γ=12.033) is a supercritical Hopf bifurcation. The system evolves with a period-1 limit cycle of increasing size as γ is increased.

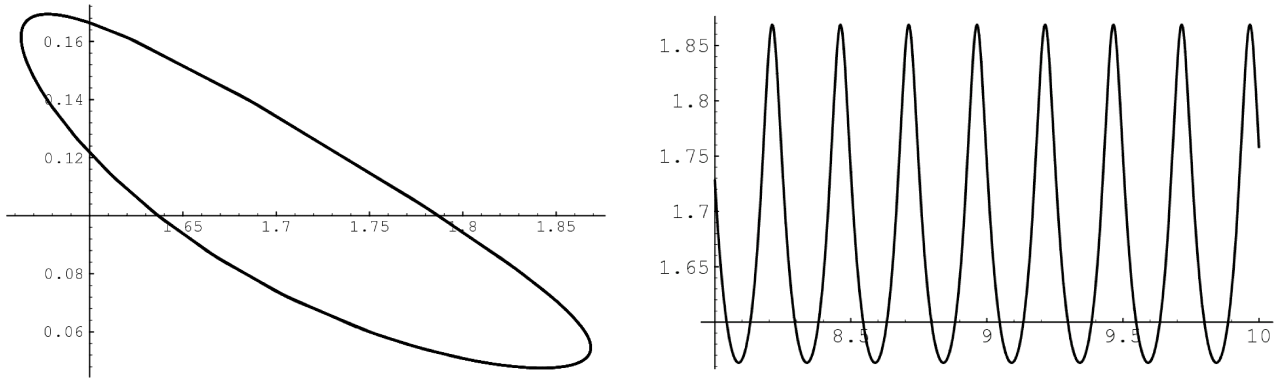

Figure 3 - 'Small' Period-1 limit cycle after the supercritical Hopf bifurcation, γ=12.3

Figure 3 reports a phase-plot and a time series of a typical limit cycle in this range of the bifurcation parameter. The phase-plot was obtained by arbitrarily selecting two variables, namely the mass fraction and the temperature at a selected location of the spatial domain. The time series represents the time evolution of one arbitrary selected scalar variable, namely the temperature at the same selected location. It is seen that the space profiles of mass fraction and temperature oscillate in time around those reported in Fig. 2a. (the fast reaction regime). As γ is increased further, a period-doubling cascade is observed, leading to chaos for γ=12.407 approximately. Chaos generated through the Feigenbaum mechanism exists for a very narrow range of the bifurcation parameter. In fact, a second, reverse period-doubling cascade is found as γ is increased. This chaos window includes many periodic windows, where period-3, period-5 and period-12 cycles were identified.

Diamond symbols reported in Fig.1 represent the periodic solutions obtained via simulations in a field that could not be characterised via continuation. With the aid of simulations, it was possible to conclude that, in this specific case, a cyclic fold bifurcation point is found within a very small interval (less than 10^{-7}) from the Hopf bifurcation value. The rightmost diamond point in Fig. 1 corresponds to the ignition limit for the system, that is when the reactant concentration builds up to a point where the heat generated by the (slow) reaction overcomes the heat losses to the boundaries.

For γ varying from 13.3783 to 12.65 the limit cycle undergoes several bifurcations, detected via simulation, among which a series of period-adding bifurcations that produce complex stable mixed-mode oscillations. These are not the only stable attractors seen for the system in this range of the parameter. For the same values of γ, quasi periodic (tori) and aperiodic attractors exist along with periodic solutions. The basins of attraction have extremely complex shapes, and a large number of simulations are needed to identify such multistability.

If one looks at the numerical results in terms of phase plots, the behaviour of all dynamic attractors seems to be driven by the contrasting tendencies of the system, attracted both to the slow reaction regime (large cycles, long periods, steep gradients) and to the fast reaction regime (small cycles, short periods, moderate gradients). Particularly, chaotic orbits often consist of several large loops followed by few smaller loops This feature is found in other chaotic systems (see for example [15]). Various possible mechanisms could justify the observed dynamic steady states. By looking at the structure of the underlying mathematical model of the system under study, it is seen that the two partial differential equations are only coupled through the nonlinear source term. This means that, in the regions where the source term vanishes or is very small, the equations are locally almost uncoupled. Note that such an occurrence is quite common in combustion, where activation energies are large and reaction zones are small compared to the spatial domain. The system can be at times separated into subsystems which are very loosely coupled only through heat conduction, and this makes it possible, for example, to think of the periodic attractor generated via a period-adding bifurcation as a result of the synchronisation of two dynamic steady-states of two (weakly) coupled oscillators. Analogously, a torus can be thought as generated by the composition of two (almost) independent dynamic steady-states of two (almost) uncoupled oscillators.

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