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Modeling and analysis of local hot spot formation in down-flow adiabatic packed-bed reactors

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Abstract

Down-flow packed-bed reactors used in hydrodesulphurization and hydrogenations have been known to form localized temperature hot spots during their operation. Local hot spots may form due to non-uniform packing or activity of the catalyst bed. We examine a mechanism for hot spot formation which may occur even in perfectly uniform catalyst beds. In a down-flow reactor, the increase in temperature due to heat released by the reaction causes a decrease in the fluid density and hence creates a buoyancy force (in the upward direction), which can destabilize the uniform flow. Any spatial non-uniformity in the bed packing or activity is amplified by the buoyancy effect. In this work, we consider the case of an adiabatic down-flow catalytic packed-bed reactor of uniform porosity and activity and develop a 3D two-phase model consisting of the continuity and momentum equations, fluid and solid phase species and energy balances, and use the Boussinesq approximation for the variation of density and viscosity of the fluid with temperature. Unlike previously published research on this subject, our model also accounts for the variation of local interphase heat and mass transport coefficients with local fluid velocity. This eliminates the existence of non-physical solutions obtained at small residence times. The steady-state behavior of the 1D (transversely uniform) solutions is analyzed in detail. It is found that up to five steady-states can exist for $Le_f \geqslant 1$, while for $Le_f < 1$, there can be as many as eleven 1D uniform flow solutions. The stability of these 1D solutions with respect to two and three-dimensional transverse perturbations is analyzed. We find that the uniform solution can become unstable to small transverse perturbations and lead to transversely non-uniform velocity, concentration and temperature profiles, corresponding to maldistributed flows and localized hot spots. The size of the parameter region in which transverse non-uniformities exist increases monotonically with increasing interphase transport resistances. Spectral methods are used to follow numerically the bifurcating non-uniform solutions and the corresponding velocity, concentration, and temperature fields. The bifurcation from uniform to non-uniform solutions changes from supercritical to subcritical as the interphase transport resistance increases. Finally, the results are summarized in terms of some guidelines for minimizing the occurrence of maldistributed flows and hot spots in down-flow adiabatic packed-bed reactors.

Keywords: Packed-bed; Hot spot; Down-flow; Flow maldistribution; Stability

1. Introduction

Catalytic packed-bed reactors are used extensively to carry out several exothermic catalytic reactions such as partial oxidations and hydrogenations. The design and operation of large diameter commercial adiabatic packed-bed reactors is complicated by the possibility of flow maldistributions and localized hot spot formation. Hot spots can lead to local deactivation of the catalyst, loss in selectivity, or reaction runaway. Hot zones have been reported to form in commercial adiabatic packed-bed reactors. Jaffe (1976) observed steady-state hot spots in a hydrogenation process and simulated them with limited regions of low flow. Barkelew and Gambir (1984) reported that small aggregates of molten catalyst pellets were generated during the hydrodesulphurization of crude oil. Boreskov and Matros (1983), and Matros (1985) observed local hot zones in the bottom of a reactor used to carry out partial oxidation of isobutyl alcohol. Schmitz and Tsotsis (1983), Razon and Schmitz (1987), and Brown and Schmitz (1989) found, using IR imaging, that the surface temperature of catalytic pellets

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were not uniform even for some atmospheric, exothermic reactions.

Several investigators attempted to explain and predict the mechanisms for the formation of hot spots in large diameter packed-bed reactors. We review here the three main mechanisms that have been identified for hot spot formation in commercial reactors (Balakotaiah et al., 1999). It has been shown that hot zones may form due to non-uniform catalyst activity or packing (porosity) in the bed (Matros, 1985). The second mechanism, which is the major focus of this work, arises from the coupling between the heat released (due to reaction) and the temperature dependence of the density. When an exothermic reaction is carried out in a down-flow adiabatic packed-bed reactor, the density of the reacting fluid decreases in the flow direction. The buoyancy force destabilizes uniform down-flow and, beyond a critical point, flow maldistribution and hot spots spontaneously appear in some regions of the reactor (Stroh and Balakotaiah, 1991, 1992; Nguyen and Balakotaiah, 1994, 1995). Stroh and Balakotaiah (1991, 1992) analyzed the pseudohomogeneous model of the packed-bed reactor with various types of boundary conditions and determined the stability boundary of uniform flow. Nguyen and Balakotaiah (1994, 1995) and Christoforatou and Balakotaiah (1997) analyzed a 1D pseudohomogeneous model of the packed-bed reactor and determined the conditions for the onset of flow maldistributions using residence time as the bifurcation parameter. The third mechanism arises from the different rates of diffusion of species and energy in the transverse direction and nonlinear chemical reactions. In this case, a uniform temperature field can become unstable to transverse perturbations even when the physical properties of the reacting fluid are constant and bed porosity and activity are constant. Balakotaiah et al. (1999) investigated a 3D pseudohomogeneous model of a packed-bed with Langmuir–Hinshelwood kinetics and showed that transverse non-uniformities can arise when reactor to catalyst particle diameter exceeds about five. Dommeti et al. (1999) analyzed the criteria that predict the applicability of pseudohomogeneous models of catalytic reactors and concluded that one should use heterogeneous models for most industrial catalytic reactors, though such models display a higher degree of complexity. Marwaha et al. (2004) and Sundarram et al. (2005) have shown experimentally that global coupling can lead to formation of hot zones on the top of a packed-bed reactor. Recently, Viswanathan et al. (2005), and Viswanathan and Luss (2006) showed that a two-phase model of a shallow, adiabatic packed-bed reactor cannot predict a stable patterned solution bifurcating from the uniform solution when the transversal heat dispersion is larger than that of the species. However, this result applies only to the case of constant physical properties (in addition to constant porosity and activity), constant (or velocity independent) values of interphase heat and mass transfer coefficients and a single step first-order exothermic reaction.

The *a priori* prediction of the conditions leading to hot spot formation requires a comprehensive analysis of mathematical models that account for all of the mechanisms previously described. In this work, we present a 3D two-phase model for an adiabatic down-flow catalytic packed-bed reactor that includes two of the above cited mechanisms. Unlike all prior work on this subject, the model accounts for the variation of the local heat and mass transport coefficients with velocity. The model consists of the continuity and momentum equations, fluid and solid phase species and energy balances, and the Boussinesq approximation (for the variation of density and viscosity of the fluid with temperature). [It should be pointed out that a model of this type (that accounts for interphase gradients, fluid physical property variations, and dependence of local heat and mass transfer coefficients on local velocity) has not been developed or analyzed in the prior literature even for the simpler 1D case.] The model is non-dimensionalized such that the fluid velocity (or residence time) enters in only one dimensionless group, namely the Damköhler number (dimensionless residence time). First, the 1D transversely uniform flow solution of this model is analyzed. The stability boundary for uniform flow is determined with respect to two- and three-dimensional transverse perturbations and the influence of various parameters on the stability boundary of uniform down-flow is analyzed. Some bifurcation diagrams and 2D non-uniform solutions are computed for typical parameters. Finally, some guidelines for avoiding flow maldistributions and localized hot spot formation in down-flow adiabatic packed-bed reactors are outlined.

2. Mathematical model

The two-phase mathematical model of an adiabatic packed-bed reactor consists of the continuity, momentum, species and energy balances. We assume that the velocity through the packed-bed is small so that inertial effects are negligible and Darcy's law is valid. The variation of the reaction mixture density and the viscosity are assumed to be linearly dependent on the temperature [$\rho_f = \rho_{fo}(1 - b(T_f - T_{fo}))$ and $\mu_f = \mu_{fo}(1 - a^*(T_f - T_{fo}))$]. In addition, it is further assumed that density and viscosity variations are small and affect only the body force term in the momentum balance (Boussinesq approximation). For simplicity of calculations, the model assumes negligible intraparticle thermal and concentration gradients. In most practical cases, temperature gradients inside the catalyst particles are very small and may be neglected. However, the intraparticle diffusional resistance may not be negligible. Thus, the last assumption gives a more conservative stability limit because diffusional resistance inside the catalyst pellet is neglected. [It is also satisfied when the catalyst activity profile is of egg shell type.]

In accordance with the aforementioned assumptions, the resulting governing dimensionless equations are

$$\nabla \cdot \mathbf{v} = 0, \tag{1}$$

$$\nabla \Pi = -\frac{1}{Da}\mathbf{v} - \left(\frac{1}{Da_N} - \frac{\Lambda}{Da}\right)\theta_f \mathbf{e}_z, \tag{2}$$

$$\varepsilon\frac{\partial w_f}{\partial \tau} + \frac{1}{Da}\mathbf{v}\cdot\nabla w_f = \frac{1}{\phi^2_{m,f}}\nabla^2 w_f - \frac{(w_f - w_s)}{Da_{pm}}, \tag{3}$$

$$\varepsilon\frac{\partial \theta_f}{\partial \tau} + \frac{1}{Da}\mathbf{v}\cdot\nabla \theta_f = \frac{1}{\phi^2_{h,f}}\nabla^2 \theta_f - \frac{(\theta_f - \theta_s)}{Da_{ph}}, \tag{4}$$