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Modeling of trickle-bed reactors with exothermic reactions using cell network approach

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Abstract

One-dimensional (1D) and two-dimensional (2D) cell network models were developed to simulate the steady-state behavior of trickle-bed reactors employed for the highly exothermic hydrotreating of benzene. The multiphase mass transfer-reaction model and novel solution method are discussed in this report. The 1D model was shown to satisfactorily simulate the axial temperature field observed experimentally for multiphase flow with exothermic reactions. The 2D reactor modeling provided valuable information about local hot spot behavior within the multiphase reactor, identifying situations in which hot spots may form. The model took into consideration the heterogeneous nature of liquid distribution, including radial liquid maldistribution and partial external wetting. This approach was proven to be stable and efficient in dealing with the complex interaction of phase vaporization and temperature rise. Through analysis and discussion, this report established the cell network model as a valid representation of the flow environment produced in a trickle bed with exothermic reactions.

Keywords: Catalysis; Evaporation; Trickle bed reactors; Multi-phase flow; Reaction kinetics

1. Introduction

A large group of industrial processes involve exothermic catalytic reactions between liquid and gaseous components, such as hydrogenations and oxidations. One multiphase reactor frequently used in such industrial reactions is the trickle-bed reactor, in which gas and liquid flow downward through packed beds to undergo chemical reactions. During this process, reactor scale maldistribution and incomplete external wetting of particles can occur due to trickle flow. In large industrial units, considerable radial temperature gradients can exist when the reaction heat release and maldistribution of the liquid–gas mixture are significant. Unfortunately, mixing-cup temperature controls at the reactor outlet are not sufficient in preventing large temperature differences between the central zone of the packed bed and the wall (Specchia and Baldi, 1979). As a result, the interaction of heat and mass transfer during strong exothermic reactions inside the reactor can be so serious that under certain conditions, a partial or total phase change can occur. This may include an evaporation of the liquid phase, complicating both the reaction and transport phenomena. The higher reaction rate accelerates the heat production and thus enlarges the hot-spots. The result is the formation of hot zones that may have a deleterious impact on the yield of the desired product. For example, sintering of the catalyst has been observed in industrial trickle-bed reactors (Specchia and Baldi, 1979; Chaudhari et al., 2002; Deans and Lapidus, 1960). When a hot zone exists next to the reactors walls, there may be a decrease in the mechanical strength of the wall. This will eventually cause damage to the reactor vessel by generating a leak that may lead to an explosion. Thus, it is of great practical importance to understand the nature and origin of these hot spots and predict their location and size.

The hot zone evolution in a packed bed is affected by the integration of the reaction rate, transport of heat and species in the radial direction, and the impact on the fluid physical

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properties due to the changes of reactant concentrations and temperature in the flow direction. Considerable capital and operational costs have been invested to resolve rapid and excessive temperature changes. To reduce this cost and elucidate these phenomena, heterogeneous models have been developed. Remediation efforts have been made to provide a detailed analysis of non-isothermal effects on reactor operations, such as the one-dimensional (1D) reactor modeling and experiment verification reported for a trickle-bed reactor under non-isothermal conditions (Chaudhari et al., 2002). Two-dimensional (2D) models, including the cell network model, have been developed to locate the hot spot in the reactor.

The cell network model has come into being since the early 1960s. In its original concept, a packed bed of spheres was approximated as a cylindrically symmetrical network of perfectly stirred tank reactors. The reactants were envisioned to enter any given stirred tank as a single phase from the two preceding tanks. Alternative rows were offset at half a tank to allow for radial mixing. The effluent from the stirred tank was then fed through subsequent stages (Deans and Lapidus, 1960). Jaffe (1976) applied this concept to the heat release of a single phase hydrogenation process, and simulated the occurrence of steady state hot spots due to flow maldistribution. Schnitzlein and Hofmann (1987) developed an alternative cell network model in which the elementary unit consisted of an ideal mixer and a subsequent plug flow unit. These fluid streams were split or merged in infinitesimally small adiabatic mixing cells (without reaction), located between the different layers of the elementary units. Kufner and Hofmann (1990) incorporated the radial porosity distribution into the above cell model, which led to a better agreement of the predicted temperature profile with the experimental data. Despite all the development in the cell network model, one should note that the above-mentioned models were examined only for single phase flow with offset in alternative rows of cells.

In this work, our overall objective is to develop a model capable of handling multiphase flow and reactions, as well as temperature change due to both phase transition and flow maldistribution for a trickle-bed reactor. The model will serve as a guide to understanding the reactor performance and optimization. At first a comprehensive 1D mixing cell model is developed to account for the phase transition in pilot plant reactors devoid of radial flow maldistribution. This model includes the local changes of phase velocities, species concentrations, external wetting efficiency, liquid holdup and mass transfer rate due to phase transition, and their effects on the reaction rates. The 1D model is applied in the reaction system of benzene hydrogenation to cyclohexane in order to predict the temperature profiles and the change of species concentrations along the reactor axis. After the 1D model is validated against the experimental temperature data reported in the literature, it is extended to the 2D model to assess the impact of flow maldistribution on the formation of hot spots. The 2D model is able to take advantage of the validated 1D model and a new solution scheme is designed to expedite the solution process and to enhance the solution stability. The ultimate goal is to develop a reliable model that will enable a rational design and control to avoid the undesirable formation of hot spots.

2. 1D reactor model

2.1. Reactor scale model

The model is designed to incorporate exothermal reactions, vaporization of liquid phase components and the effect of partial external wetting of the catalyst. Plug flow for each phase is assumed in many reported reactor scale models that consist of a set of ordinary differential equations (ODEs) with well-defined initial and boundary conditions. However, when a large amount of heat is released due to chemical reactions, the numerical computation of these ODEs becomes difficult due to the stiffness of the equations. To overcome these difficulties we have implemented a cell network approach. As shown in Fig. 1, the reactor from the inlet to the outlet is divided into a number (N) of well-mixed cells in stacks along the axial direction. Mixing only occurs within each cell and back-mixing is not accounted for between the adjacent cells. The grid independence was obtained after N reached a certain limit. The 1D model is applicable for systems without liquid or gas maldistribution.

Governing equations are presented for the multiphase mass transfer in each cell. The mass balance equations for any species in the reaction system need to be solved in conjunction with hydrodynamic and kinetic models. Mass transfer resistances consist of the resistance between the gas, liquid and solid phase, whereas the intraparticle mass transfer is reflected by the catalyst effectiveness factor. An external wetting efficiency (η_{CE}) was introduced in the cell model to account for the partial external wetting of the catalyst particles in the local cell. The mass balance equation for liquid phase at each cell is given as

$$U_L \frac{(C_{k,L} - \phi_{k,L})}{L_c} = (K_L a_{GL})_k [C_{k,e} - C_{k,L}] - k_{LS,k} a_{LS} [C_{k,L} - C_{k,LS}]. \quad (1)$$

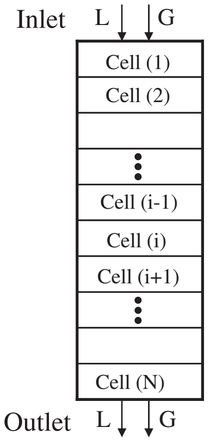

Fig. 1. Schematic representation of flow through the cell stacks.