Design of multi-phase and catalytic chemical reactors: a simulation tool for pollution prevention

Jack R. Hopper, Jamal M. Saleh, Ralph Pike

Abstract A comprehensive chemical reactor analysis tool was required to complete a project to develop an advanced on-line process optimization analysis system for pollution prevention. The advanced process analysis system integrates programs (reactors, on-line optimization, pinch analysis, and process flow-sheeting) to analyze and modify chemical processes for waste minimization. The reactor analysis program is to be used to evaluate and analyze multi-phase and catalytic reactors to suggest to the plant and process engineers the best reactor type and operating conditions. A multi-phase catalytic reactor design and analysis tool, ReaCat, has been developed. ReaCat incorporates models to design the following reactor types: plug flow, CSTR, batch, catalytic fixed-bed, catalytic fluidized-bed, gas–liquid stirred tank, trickle-bed, three-phase fixed bubble-bed, bubble slurry column, CSTR slurry, and three-phase fluidized-bed. This paper gives a summary of the multi-phase and catalytic reactors: classifications, theory and design models, numerical methods, and solution algorithms. A description of the reactor analysis tool including comparison cases with experimental data is presented.

List of symbols

$A$ Heat transfer area (m$^2$)

$C_{G,i}$ Gas bulk concentration of component $i$ (mol/l)

$C_{G,i}^*$ Inlet bulk gas concentration of component $i$ (mol/l)

$C_{G,i}^o$ Outlet bulk gas concentration of component $i$ (mol/l)

$C_{L,i}$ Liquid bulk concentration of component $i$ (mol/l)

$C_{L,i}^*$ Inlet bulk liquid concentration of component $i$ (mol/l)

$C_{L,i}^o$ Outlet bulk liquid concentration of component $i$ (mol/l)

$C_{G,i}$ Concentration of component $i$ at catalyst surface (mol/l)

$C_{pg}$ Specific heat capacity of gas [(BTU/°F lb.) cal/Kg]

$C_{pl}$ Specific heat capacity of liquid [(BTU/°F lb.) cal/K g]

$D_a$ Bulk diffusivity (cm$^2$/s)

$D_{l,i}$ Liquid-phase axial dispersion coefficient (cm$^2$/s)

$E$ Energy of activation (kJ/mol)

$F_{gas}$ Flow rate of gas (m$^3$/s)

$F_i$ Inlet flow rate for component $i$

$\Delta H_i$ Heat of reaction of the component $i$ (kJ/mol)

$H_i$ Henry's constant for component $i$

$K$ First-order reaction constant

$K_{BC}$ Gas-phase (bubble-phase) mass transfer coefficient (1/s)

$K_{CE}$ Intermediate (cloud-wake) phase mass transfer coefficient (1/s)

$K_{L,a_i}$ Liquid–solid mass transfer coefficient (1/s)

$(K_{i,a_g})$ Gas–liquid mass transfer coefficient for component $i$ (1/s)

$(K_{i,a_g})$ Liquid–solid mass transfer coefficient for component $i$ (1/s)

$K_{L,a_g}$ Gas-phase mass transfer coefficient (1/s)

$K_{L,a_g}$ Liquid-phase mass transfer coefficient (1/s)

$k_a$ First-order reaction constant

$N_s$ Number of species in mixture

$P_i$ Inlet bulk gas partial pressure of component $i$ [(psi) bar]

$P_{o,i}$ Outlet bulk gas partial pressure of component $i$ [(psi) bar]

$Q_G$ Gas volumetric flow rate (cm$^3$/s)

$Q_i$ Liquid volumetric flow rate (cm$^3$/s)

$Q_{max}$ Maximum gas flow rate (cm$^3$/s)

$R$ Gas constant (1 atm/mol K)

$(r_i)$ Net rate of disappearance of component $i$

$t$ Time (s)

$T_i$ Inlet temperature (°F K)

$T_o$ Outlet temperature (°F K)

$T_a$ Ambient temperature (°F K)

$U$ Overall heat transfer coefficient [BTU/h ft$^2$ °F (cal/s cm$^2$ K)]

$U_G$ Gas superficial velocity (cm/s)

$U_b$ Bulk gas-phase velocity (cm/s)

$U_L$ Liquid superficial velocity (cm/s)

$V_{R}$ Reactor volume (m$^3$)

$V_T$ Volume of reservoir (m$^3$)
z  Axial position (cm)
ρL  Density of liquid (g/cm³)
τ  Gas-space time (VR/Qc) (s)

**Introduction**

To perform an advanced process system analysis, which is used to evaluate chemical and refinery processes for waste minimization (Telang 1996, Pike et al. 1998), an advanced chemical reactor tool is essential. The reactor design tool is to be used to evaluate and analyze the various types of industrial multi-phase and catalytic reactors. In an effective and time saving manner, plant and process engineers need a reactor design tool to analyze and study the effect of operating conditions on the pollution index. For this purpose, ReaCat, a multi-phase catalytic reactor analysis simulation tool, has been developed with the following features:

1. User-friendly input/output interface
2. Graphical and tabular data output
3. Reactor models included
   i. Homogeneous reactors (plug flow, CSTR, batch)
   ii. Heterogeneous reactors
   I. Catalytic gas fixed-bed
   II. Catalytic liquid fixed-bed
   III. Catalytic gas fluidized-bed
   IV. Catalytic liquid fluidized-bed
   V. Gas–liquid continuous stirred tank
   VI. Three-phase trickle-bed
   VII. Three-phase bubble fixed-bed
   VIII. Three-phase catalytic gas–liquid slurry stirred tank
   XI. Three-phase catalytic gas–liquid slurry bubble-bed
   X. Three-phase catalytic gas–liquid fluidized-bed
4. Power-law reaction rate or the Langmuir–Hinshelwood model to account for the catalytic adsorption effects
5. Equipped with correlation to estimate the external mass transfer effects (gas–liquid, and liquid–solid), and dispersion coefficients
6. Estimation of the catalytic effectiveness factor to account for the intra-particle resistance
7. Isothermal and non-isothermal/non-adiabatic conditions
8. Multi-reaction systems with up to 30 reactions and 36 components
9. Prediction of reactor hydrodynamics such as pressure drop, power consumption, catalyst-wetting factor, and flow regimes.

**Multi-phase and catalytic reactor, definitions and classifications**

In industrial chemical processes, multi-phase reactors have a wide range of applications such as oxidation, hydrogenation, hydro-desulfurization, and the Fischer–Tropsch synthesis. Table 1 lists the definition for the different reactor types included in this study. Multi-phase reactors are defined as reactors with at least two distinct phases in contact. Reactors, in general, may be classified based on the number of phase coexistence into the following category:

i) Homogeneous: one phase such as gas or liquid exists in the reactor. The hydrodynamic flow characteristic of the mixture determines the reactor type such as plug, CSTR, or batch.

ii) Heterogeneous: two distinct phases of reactants (or catalyst) coexist. This category may be classified into the following subcategories:

a. Catalytic reactors: gas or liquid phase (or both) is in contact with a catalyst (mainly solid, but could be another liquid phase). Examples of this category include the catalytic packed-bed (catalytic gas reaction) and the three-phase trickle-bed (catalytic gas–liquid reaction).

b. Non-catalytic reactors: gas–liquid or liquid–liquid reactions are carried in a variety of contact vessels such as the gas–liquid continuously stirred tank reactor.

Each reactor type exhibits certain characteristics and advantages that may make it the best candidate for a specific reaction; the power of a simulation tool becomes very clear for such an analysis. For a complete discussion on examples, classification, definitions, and advantages or disadvantages of multi-phase and catalytic industrial reactors, the reader is referred to Ostergaard (1974), Shah (1979), Tarhan (1979), Ramachandran and Chaudhari (1983), Tsutsumi et al. (1987), and Saleh (1994).

**Table 1. Definitions of reactors**

<table>
<thead>
<tr>
<th>Reactor type</th>
<th>Description</th>
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<tbody>
<tr>
<td>Catalytic packed-bed</td>
<td>Gas or liquid reactants flow over a fixed-bed of catalysts</td>
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<tr>
<td>Catalytic fluidized-bed</td>
<td>The up-flow gas or liquid phase suspends the fine catalyst particles</td>
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<tr>
<td>CSTR gas–liquid</td>
<td>Liquid and gas phases are mechanically agitated</td>
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<tr>
<td>Bubble gas–liquid bed</td>
<td>Liquid phase is agitated by the bubble rise of the gas phase. Liquid phase is continuous while gas phase is continuous</td>
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<tr>
<td>Trickle-bed</td>
<td>Concurrent down-flow of gas and liquid over a fixed-bed of catalyst. Liquid trickles down, while gas phase is continuous</td>
</tr>
<tr>
<td>Bubble fixed-bed</td>
<td>Concurrent up-flow of gas and liquid. Catalyst bed is completely immersed in a continuous liquid flow while gas rises as bubbles</td>
</tr>
<tr>
<td>CSTR slurry</td>
<td>Mechanically agitated gas–liquid catalyst reactor. The Fine catalyst particles are suspended in the liquid phase by means of agitation. (Batch liquid phase may also be used)</td>
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<tr>
<td>Bubble slurry column</td>
<td>Liquid is agitated by means of the dispersed gas bubbles. Gas bubble provides the momentum to suspend the catalyst particles</td>
</tr>
<tr>
<td>Three-phase fluidized-bed</td>
<td>Catalyst particles are fluidized by an upward liquid flow while gas phase rises in a dispersed bubble regime</td>
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