Chemical Engineering Journal 321 (2017) 286–300

Contents lists available at ScienceDirect

Chemical Engineering Journal

journal homepage: www.elsevier.com/locate/cej

Steady-state characteristics of autothermal structures with fluidized-bed catalytic reactors

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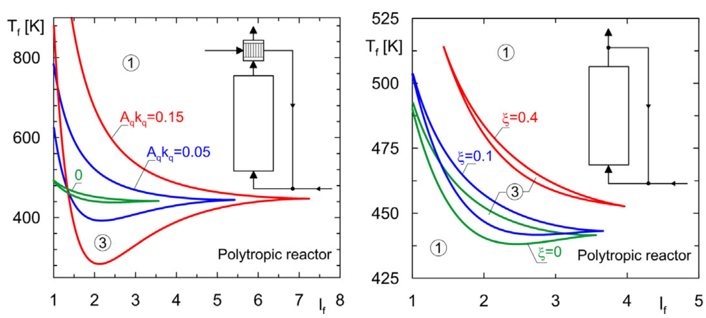
HIGHLIGHTS

- Steady-state properties of various fluidized-bed autothermal structures are analyzed.
- Effect of an external heat exchanger and a flue-gas recycle is evaluated.
- The impact of thermal feedback onto autothermicity and multiplicity is analyzed.
- Feed preheating permits to extend the region of the autothermal operation.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 26 December 2016
Received in revised form 26 March 2017
Accepted 27 March 2017
Available online 29 March 2017

Keywords:
Autothermal structure
Fluidized-bed reactor
Steady states
Multiplicity
Stability
Process yield

ABSTRACT

The paper focuses on the analysis of steady-state properties of autothermal structures with a fluidized bed. The concept of autothermicity is discussed, and typical autothermal structures are presented. A method for the determination of the steady-states of such systems is described. Quantitative analysis of steady-states multiplicity is presented for two autothermal structures, that is a fluidized-bed reactor with an external heat exchanger, and with a partial recirculation of hot gases. A two-phase bubbling bed model is employed to describe quantitatively the behavior of the catalytic fluidized bed. The influence of selected design and operating parameters on steady-state characteristics is analyzed for two simple kinetic models. The effect of these parameter on the desired product yield is evaluated for the multiple-reaction process. The results obtained have both scientific and practical importance.

1. Introduction

An autothermal system is a chemically reacting system which is self-sufficient in terms of its energy requirements [1]. Examples of such systems are flames, single chemical reactors, and systems of chemical reactors coupled together or with heat exchangers. The overall thermal effect of the chemical process taking place in such systems is exothermic. Every autothermal system is characterized by the presence of internal or external thermal feedback.

The problem of the autothermicity of chemical reactors has been a frequent subject of research. Var Heerden [1] was the first to perform the steady-state analysis of an autothermal continuously stirred tank reactor (CSTR) and a catalytic tubular reactor with an external heat exchanger. He demonstrated that the process autothermicity is inherent to the existence of multiple steady states (MSS) in such systems. A comprehensive analysis of autothermicity and steady-state multiplicity for the classical chemical reactors, i.e. a tubular reactor with an internal exchanger and with an external heat exchanger, and a CSTR with an external heat exchanger, was presented in [2]. Due to the possible occurrence of multiple steady states it is necessary to determine the

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http://dx.doi.org/10.1016/j.cej.2017.03.129
1385-8947/

Nomenclature

a_i	model parameter
a_q	heat transfer area per unit volume of a bed, m^{-1}
A_q	heat transfer area in an autothermal heat exchanger, m^2
B_i, B_{ij}	model parameter characterizing interphase mass and heat transfer
c_g, c_z	specific heat of gas and solid particles, $kJ \cdot kg^{-1} \cdot K^{-1}$
C_j	concentration of the j^{th} component, $kmol \cdot m^{-3}$
d_z	diameter of a catalyst pellet, m
D_{ij}	diffusion coefficient of the j^{th} gaseous reactant in the i^{th} phase, $m^2 \cdot s^{-1}$
E_{aj}	activation energy of adsorption of the j^{th} component, $kJ \cdot kmol^{-1}$
E_i	activation energy of the i^{th} chemical reaction, $kJ \cdot kmol^{-1}$
F_M	mass flow rate of gas, $kg \cdot s^{-1}$
h	coordinate of height in a fluidized bed, m
Δh_i	enthalpy of the i^{th} chemical reaction, $kJ \cdot kmol^{-1}$
H	total height of a bed, m
k_{0i}	frequency coefficient in the Arrhenius equation, s^{-1}
k_q	overall heat transfer coefficient, $kJ \cdot m^{-2} \cdot s^{-1} \cdot K^{-1}$
K_{aj}	slope of adsorption isotherm for the j^{th} component, $m^3 \cdot kg^{-1}$
l_f	fluidization ratio
p	total pressure, atm
q_j	amount of the j^{th} component adsorbed on inert support of a catalyst pellet, $kmol \cdot kg^{-1}$
Q_i	model parameters determining intensity of heat transfer
r_i	rate of the i^{th} chemical reaction, $kmol \cdot m^{-3} \cdot s^{-1}$
$\tilde{r}_i$	modified rate of the i^{th} chemical reaction, s^{-1}
R	gas constant, $kJ \cdot kmol^{-1} \cdot K^{-1}$
S	cross section of a fluidized bed, m^2
t	time, s
T	temperature, K
x_E	ratio of activation energies E_2/E_1
$x_{\Delta h}$	ratio of enthalpies $\Delta h_2/\Delta h_1$
x_k	ratio of frequency coefficients in the Arrhenius equation k_{02}/k_{01}
u	gas velocity, $m \cdot s^{-1}$
W_P	yield of a process with respect to product P, $kmol \cdot m^{-2} \cdot s^{-1}$
y_j	molar fraction of the j^{th} component
z	dimensionless height of the bed

Greek symbols

α_i	conversion degree of the j^{th} component
α_q^{ij}	heat exchange coefficient between phases i and j, $kJ \cdot m^{-3} \cdot s^{-1} \cdot K^{-1}$
β_{gk}^{ij}	gas exchange coefficient of component k between phases i and j, s^{-1}
β_z^{ij}	solid particles exchange coefficient between phases i and j, s^{-1}
δ	volume fraction of bubbles in a bed
ε_{mf}	bed porosity at minimum fluidization conditions
ε_z	porosity of a catalyst pellet
ν_j	stoichiometric coefficient of the j^{th} component
η_j	dimensionless concentration of the j^{th} component
ρ_g, ρ_z	density of gas and solid particles, respectively, $kg \cdot m^{-3}$
ξ	recirculation ratio
ζ	refers to cold stream inlet to an autothermal heat exchanger

Subscripts

A, P, R	refers to components A, P and R, respectively
b, *e*	refers to bubble and emulsion phase, respectively
f	refers to feed stream
mf	refers to minimum fluidization conditions
M	refers to mass
p	refers to product
r	refers to recycled stream
ref	refers to reference conditions
q	refers to cooling medium
s	refers to external heat exchanger

Superscripts

b, *c*, *e*	refers to bubble, clouds and wakes, and emulsion phase, respectively

regions of their existence. Information on the process conditions under which MSS occur is required during start-up and for proper process control. The problem of autothermal processes control was studied in [3]. The authors discussed various control strategies applied in fixed-bed autothermal reactors for catalytic combustion. There are also numerous works dealing with the practical aspects of the various configurations of autothermal catalytic reactors. However, they are limited to the chemical processes run in fixed-bed reactors [4–6]. Apart from a few works focusing on selected chemical processes, e.g. hydrogen production [7], there is a lack of literature dealing with steady-state analysis of so-called fluidized-bed autothermal structures (FBAS). Therefore this problem is the subject of the present study.

In a single fluidized-bed reactor the bed hydrodynamics can provide conditions that ensure the autothermicity of the process. It appears that the fluidized bed creates an internal channel with a positive thermal feedback. A cold feed is heated by the contact with the bed of the catalyst. The mechanism of internal thermal feedback reduces with the decreasing thermal effect of the process.

In this work, a fluidized-bed reactor having additional external channels of thermal feedback is referred to as a fluidized-bed autothermal structure (FBAS). The feedback can be implemented in different ways, depending on the reactor's configuration. Fig. 1 shows some typical fluidized-bed autothermal structures. The external autothermal feedback can be implemented by the installation of an external heat exchanger to preheat the feed (Fig. 1a) or by partial recirculation of the hot gases leaving the reactor (Fig. 1b). The choice of the type of thermal feedback depends highly on the catalytic process that is carried out in the fluidized-bed reactor. In some practical situations simultaneous heat and mass feedback implemented by the partial recirculation of exhaust gas can be unsafe or may influence the process yield. Therefore, in practice the selection of the configuration has to be done carefully and depending on the process type.

Other examples of autothermal structures are the configurations with a circulating bed shown in Fig. 1c and d. In the configuration with two fluidized beds shown in Fig. 1c each bed is fed with one of the two reactants required in the process. The structure depicted in Fig. 1d can be used for the integration of endothermic and exothermic processes. Analogical design is applied, among others, in chemical-looping combustion process (CLC) [8]. The configuration of two apparatuses (Fig. 1d) can also be adopted to run