Energy xxx (2016) 1–7

Contents lists available at ScienceDirect

Energy

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

Emergence of traveling wave endothermic reaction in a catalytic fixed bed under microwave heating

Alexander P. Gerasev

Boreskov Institute of Catalysis, Siberian Branch of Russian Academy of Sciences, Prospekt Akademika Lavrentieva 5, Novosibirsk 630090, Russia

ARTICLE INFO

Article history:
Received 19 February 2016
Received in revised form
3 May 2016
Accepted 8 November 2016
Available online xxx

Keywords:
Microwave heating
Traveling waves
Heterogeneous catalysis
Endothermic chemical reaction
Mathematical modeling

ABSTRACT

This paper presents a new phenomenon in a packed bed catalytic reactor under microwave heating - traveling wave (moving reaction zones) endothermic chemical reaction. A two-phase model is developed to simulate the nonlinear dynamic behavior of the packed bed catalytic reactor with an irreversible first-order chemical reaction. The absorbed microwave power was obtained from Lambert's law. The structure of traveling wave endothermic chemical reaction was explored. The effects of the gas velocity and microwave power on performance of the packed bed catalytic reactor were presented. Finally, the effects of the change in the location of the microwave source at the packed bed reactor was demonstrated.

1. Introduction

The phenomenon of traveling wave (heat wave or else autowave) in a heterogeneous medium is a subject of big interest in the theory of unsteady-state processes in fixed bed catalytic reactor. This phenomenon was first discovered by Frank-Kamenetskii in 1947 [1] and was described as ‘migration’ of the reaction zone of isopropyl alcohol oxidation over the static bed of a cooper catalyst. The traveling wave phenomena in heterogeneous media is a spatiotemporal structure, which can form in the layer because of self-organization process and were extensively studied in the 1970s and 1980s [2–5]. Theory of nonlinear-wave propagating in active distributed systems originated from the fundamental works by Fisher [6], Kolmogorov et al. [7], Zel'dovich and Frank-Kamenetskii [8–10]. The phenomenon of traveling wave means a self-sustained nonlinear wave process (including stationary structures) whose characteristics (propagation velocity, period, wave (impulse) length, amplitude, and shape) remain unaltered due to an energy source distributed in the medium [11–13]. The phenomenon of traveling wave of exothermic reaction in packed bed catalytic reactor provides a vivid example of self-organization [14]. One of the most interesting aspects of the theory of nonlinear waves in active distributed systems is the question of traveling waves of endothermic reactions.

Cooking food with microwaves was discovered accidentally in the 1940s. The use of microwave heating is rapidly expanding. Microwave technology has been extensively employed in food and chemical engineering [15–22]. Interest in new nontraditional technologies based on catalytic processes with the use of microwave radiation, in particular, endothermic catalytic reactions, has quickened [23–26]. Heating of sample by microwave radiation is caused by interaction of this radiation with molecules (ions) within the entire volume of the irradiated material.

A quasi-homogeneous (one-temperature) model is one of the simplest (basic) models, which describe catalytic traveling wave in an active distributed media [27]. For practical applications, however, it is of undoubted interest to take into account the processes of heat and mass transfer between the filtered flow of reagents and the motionless catalyst bed, which makes the problem much more complicated. The simplest model of heterogeneous medium is a two-phase model that takes into account processes of heat and mass transfer between the phases [2,28–30]. At the same time, new effects may appear as a result of interaction between microwave heating and endothermic chemical reaction, convection, diffusion, heat conductivity, heat and mass transfer.

The aim of this paper is numerical simulation of new phenomenon - traveling wave of endothermic reaction in a packed bed catalytic reactor under microwave heating. For this purpose, we have developed a two-phase mathematical model of the packed bed catalytic reactor with an irreversible first-order endothermic

E-mail address: a.gerasev@ngs.ru.

http://dx.doi.org/10.1016/j.energy.2016.11.042

Nomenclature

a_V	interphase heat transfer coefficient
c_s, c_p	specific heat capacity of solid and gas mixture
c_i	molar density of component
D, $Đ_{12}$	effective diffusivity and Maxwell-Stefan diffusivity
d	diameter of catalyst particle
E	activation energies of surface and gaseous reactions
I_0	incident microwave power
J	molar diffusion flux
j	interphase mass transfer
k_0	reaction rate coefficient
k_λ	absorption coefficient
k_1, k_2	parameter, Eq. (24)
ℓ	longitudinal coordinate
L	length of the heterogeneous layer
M, M_A	average molecular weight and molecular weight of reactant A
p	pressure
Pr	Prandtl number
q	heat of reaction
Q_{MW}	local volumetric power dissipation due to microwaves
R	Universal gas constant
Re	Reynolds number
S	thickness of the substance layer through which radiation is passed
S_V	total surface area of catalyst particles per unit volume of bed
Sc	Schmidt number
Sh	Sherwood number
t	time
T	absolute temperature
u_s	parameter, Eq. (24)
u	mean-mass velocity of gas mixture
v_s	catalytic reaction rate
V_f	wave velocity
x, y	gas-phase and catalytic mass (mole) fraction of reactant A

Greek symbols

β	interphase mass transfer coefficient
β_1, β_2	parameter, Eq. (24)
γ_i	activity coefficient
ε, ε_s	porosity of the layer and the catalyst grain
Λ_s	parameter, Eq. (24)
λ_{eff}, λ_0	thermal conductivity of porous medium
μ	dynamic viscosity
μ_i	chemical potential
ρ	density
σ_s	Stefan–Boltzmann constant
τ_s	parameter, Eq. (24)
χ_i	stoichiometric coefficient
χ_λ	parameter, Eq. (24)
χ_C, χ_0	parameter, Eq. (24)

Subscripts

A	index for reactant A
i	index for species
s, g	solid, gas
0	boundary condition

chemical reaction under microwave heating. We study the effect of model parameters on changes in the main performance characteristics of the process, and the effect of the change in the location of the microwave source at the packed bed reactor.

2. Mathematical description of the process

Consider a packed bed catalytic reactor of uniform cross-sectional area and constant void fraction. A static catalyst bed is a complex system with inevitable statistical distribution of properties in individual structural elements. Irregularity of dispersity in a heterogeneous medium, turbulization of flows, and other factors make the system more complicated. Construction, selection, and simplification of models depend on a number of requirements imposed on these models.

To describe the unsteady process of chemical conversion of substances in the packed bed catalytic reactor, we use the approximation of the two-phase model. This model takes into account convective flow of reactive gas mixture through a porous medium, thermal conductivity of the solid phase, chemical reactions on the catalyst, which serve as a source of heat, and heat and mass transfer between the phases. Processes of heat and mass transfer inside the catalyst grain are assumed to be so intense that the difference in concentrations of substances and temperatures inside the catalyst grain can be neglected. We also ignore the diffusion, thermal conductivity, and pressure gradient in the gas phase. It is believed that the reaction mixture obeys the ideal gas law. When system is exposed to microwave radiation, two-phase model must be supplemented with the subsystem, which takes into account local heating of the medium. Fig. 1 shows the scheme of installation for microwave heating of system. Then, the one-dimensional equations of material and thermal balance have the following form:

$$(1-\varepsilon)c_s\rho_s\frac{\partial T_s}{\partial t} - \lambda_{eff}\frac{\partial^2 T_s}{\partial \ell^2} = \alpha_V(T_g - T_s) + (1-\varepsilon)qv_s\left(T_s, p_{s,i}\right), \quad (1)$$

$$\varepsilon c_p\rho_g\frac{\partial T_g}{\partial t} + c_p\rho_g u\frac{\partial T_g}{\partial \ell} = \alpha_V(T_s - T_g) + Q_{MW}(\ell), \quad (2)$$

$$\varepsilon\frac{\partial \rho_{g,i}}{\partial t} + \frac{\partial\left(\rho_{g,i}u_i\right)}{\partial \ell} = j_{g,i}, \quad (3)$$

$$(1-\varepsilon)\varepsilon_s\frac{d\rho_{s,i}}{dt} = -j_{g,i} + (1-\varepsilon)\chi_i M_i v_s\left(T_s, p_{s,i}\right), \quad (4)$$

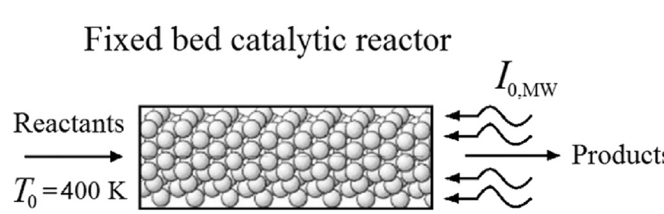

Fig. 1. Schematic representation of the configuration of heating the packed bed catalytic reactor via microwave.

Please cite this article in press as: Gerasev AP, Emergence of traveling wave endothermic reaction in a catalytic fixed bed under microwave heating, Energy (2016), http://dx.doi.org/10.1016/j.energy.2016.11.042