NUMERICAL SIMULATION OF HEAT AND MASS TRANSFER PROCESSES IN LARGE-SCALE FLUIDIZED BED COMPLEX STRUCTURE APPARATUS AS AN EXAMPLE OF THE REACTOR OF ISOPARAFFINS DEHYDROGENATION

Sergei S. Solovev¹, Svetlana R. Egorova², Alexander A. Lamberov², Olga V. Soloveva³

¹ Kazan (Volga region) Federal Univ., Inst. of Mathematics and Mechanics Kremlevskaya St. 35, 420008 Kazan, Russia e-mail: serguei_s349@mail.ru

> ² Kazan (Volga region) Federal Univ., Inst. of Chemistry Kremlevskaya St. 29, 420111 Kazan, Russia segorova@rambler.ru, lamberov@list.ru

³ Kazan State Power Engineering Univ., Inst. of Heat Power Engineering Krasnoselskaya St. 51, 420066 Kazan, Russia rara avis86@mail.ru

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Abstract. In the chemical industry are widely used fluidized bed apparatus. The advantage of them is the high speed of heat and mass transfer between components of the reaction, which are in different aggregation states. Studies of large-scale apparatus are hindered big sizes and plurality of structural elements. Often such apparatus operate at high temperatures (500-900 C), which further complicates the study. In this paper we consider a fluidized bed reactor block intended for the dehydrogenation of isobutane. In numerical simulation of fluidization was extended Eulerian-Eulerian approach. Differential equations that describe the hydrodynamic and thermal processes in the field of computational model of the reactor were solved in ANSYS Fluent CFD for axisymmetric unsteady flow scheme. At full simulation of the unit of the reactor in differential equations for the mass fraction of components of the gas mixture is necessary to consider changes related with chemical reactions. In the model used for this purpose it is necessary to add terms to the equations of mass transfer and absorption of heat depending primarily on the gas temperature and catalyst concentration. In this paper we'll restrict considering the minimum number of components of the reaction (raw materials – isobutane, product - isobutylene). For a given chemical reaction is written User Defined Functions (UDF). The influence of the ambient gas, the catalyst and the time step on the progress of chemical reaction in the volume element is studied. Numerical calculations were carried out, due to them circulating streams in the apparatus, the temperature field distribution of the catalyst and the conversion of the feeding gas-raw were analyzed.

1 INTRODUCTION

In the chemical industry fluidized bed apparatus are widely used. The advantage is the high rate of heat and mass transfer between the reaction participants being in different aggregate states. It is known a large number of works devoted to the experimental results and constructing on their basis simplified models of fluidization and recommendations for the fluidized bed apparatus operations (e.g. [1-5]). Although such apparatus are used in the industry for over fifty years, interest in the study of the properties and possibilities of the fluidized bed use is not dried up due to the advent of new and modernization of existing apparatus and technologies.

In numerical simulation of fluidization Euler-Euler approach was extended, when the carrier (gas, liquid) and discrete (solid) phase are considered as continuous. To account for features of movement in a fluidized bed, by analogy with the kinetic theory of gases, is added the equation describing the change in the kinetic energy of the granules due to their collision (e.g. [3]). Ability of numerical implementation of the equations, describing the fluidized bed, allows calculations of different variations of the fluidization process, that are of interest both in terms of fundamental research and practical application.

Each structural element in particular the feed supplying device to the reactor contributes to the apparatus efficiency. Changes in the character of the movement and concentration of particles along the wall with deflectors are shown in [6]. In works [7, 8] by numerical simulation the effect of ring baffles located on the walls of the cylindrical apparatus on the hydrodynamics of the fluidized bed was analyzed. Experimental studies with internal elements in a liquid-solid fluidized bed are presented in [9]. Investigations of fluidized bed processes in the column with an inner cylinder were experimentally made in [10]. Modelling and comparative analysis of two large fluidized bed units with various feed gas supply devices is carried out in [11]. Investigations of [12] describe a numerical simulation of the fluidized bed and the kinetics process of olefins production from methanol. There are various mathematical models, for example, to the forces of resistance, viscosity and heat transfer, analysis of the applicability of the investigated parameters, for example, in [13, 14], performing numerical simulations.

A mathematical model and a numerical study of the chemical parameters of the fluidized bed reactor unit, intended for the dehydrogenation of isobutane to isobutylene, were made in this paper. A simplified model of the basic chemical reaction was constructed. The fields of the catalyst concentration, temperature fields and fields of dehydrogenation reaction product were obtained. The analysis of the circulation flows was carried out. The effect of slight structural modifications on the efficiency of the reaction product formation was carried out. The change of the circulating gas flow and catalyst due to the changes in apparatus construction was shown. The effect of fractional composition of the catalyst on the efficiency of the reaction product was demonstrated. It was obtained that formation of circulating flow is sensitive to the particle size selection.

2 PROBLEM FORMULATION

Chemical unit of the fluidized bed reactor intended for the dehydrogenation of isobutane is considered. Reactor has a cylindrical form and a height of the operating area is 11 meters and 5.2 meters in diameter. At the bottom of the reactor a feeder of gas feedstock is located. The load on the reactor unit feed is 20-40 tonnes per hour at a temperature of 550 ° C. Used gas feedstock is isobutane fraction of the following composition:

Component	H_2	$i-C_4H_{10}$	n-C ₄ H ₁₀	i-C ₄ H ₈	$n-C_4H_8$	C ₄ H ₆
Mass						0.01-
fraction,.%	0.3-1.0	91-90	0.5-2.0	5.0-7.2	0.2-1.3	0.05

Table 1: Fractional composition of the gas feedstock.

There is aluminum chrome microspheroidal catalyst in the reactor with the particle diameters of 20-200 microns. A vertical pipe feeding regenerated catalyst at a temperature of 650 °C is along the axis of the reactor. The pipe length is of 7 m. The process of catalyst circulation from the reactor to the regenerator and back is continuous and the volume flow rate of the circulating catalyst is 60-100 tons per hour. Channel for discharging the catalyst into the regenerator is at the bottom part of the reactor. In the middle of the reactor in the working area angular falling type lattices with a free cross section about 30% are located. In the upper part of the reactor 6 pairs of cyclones are arranged for separation of exhaust gas from the reactor and returning the catalyst particles getting into the working zone of the reactor. Figure 1 demonstrates a scheme of meridian section of the reactor unit.

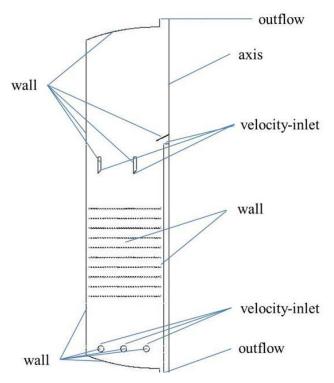


Figure 1: Scheme of meridian section of the reactor unit.

The main chemical reaction in the block is the dehydrogenation of isobutane, accompanied by the hydrogen evolution and heat absorption:

$$C_4 H_{10} \leftrightarrow C_4 H_8 + H_2 + \Delta H. \tag{1}$$

Dehydrogenation of C₃-C₅-paraffins is thermodynamically feasible only at temperatures 300-900 °C [15]. With non-localized energy supply to the hydrocarbon molecule the conversion will follow the path of cracking. Therefore, the dehydrogenation of paraffins can be carry out practically only in the presence of catalysts, that selectively increase the rate of dehydration by changing its activation energy, but not affecting the rate of reaction chain breaking. In

industrial processes the dehydrogenation of C3-C5 paraffins is carried out at temperatures of 520-620 °C and at a pressure close to atmospheric pressure or under vacuum [15]. At a temperature of 550-580 °C in the dehydrogenation of isobutane to aluminum chrome catalyst dehydrogenation reaction yield product (isobutene) is 35-50 % of mass fraction, cracking products is $\sum (C_1-C_3) = 3-7$ % of mass fraction, isomerization (n-butylene) = 0.2-0.5% of mass fraction, the oligomerization product yield (divinyl) is not more than 0.2% of mass fraction. The amount of coke formed on the catalyst is 0.2-2% of mass fraction. To remove high molecular hydrocarbon deposits use periodic catalyst regeneration by burning out the coke from the catalyst surface with fuel gas and air at a temperature of 650-670 °C.

3 MATHEMATICAL MODEL AND NUMERICAL SIMULATION

Considering chemical reactor unit consists of a large number of structural elements. The constructing of a three-dimensional mathematical model with all structural parts would lead to a great cost of calculation time. The cylindrical form of unit, the central pipe and circular arrangement of gas supply nozzles and cyclones allows the use of the model of axial symmetry. It is considered that an arbitrary meridional cross-section of the unit fully describes the processes taking place in the reactor. The geometry used in calculations corresponds to the figure 1. The axisymmetric model assumes that the reactor model is constructed by the geometry (shown in the figure) rotating around the axis of symmetry. The geometrical sizes of the model correspond to the geometric sizes of the test reactor unit.

3.1 Fluidized bed model

In the numerical simulation of a fluidized bed in a chemical reactor the continuous multiphase model was used, supplemented by kinetic theory of gases for the accounting the collisions of solid particles. At the same time several phases may be considered: gas, liquid or solid in any combination. For each of the phases holds the laws of mass conservation, momentum and energy.

The equation of mass conservation for the *i*-th phase is:

$$\frac{\partial \alpha_i \rho_i}{\partial t} + \nabla \cdot (\alpha_i \rho_i \vec{v}_i) = 0, \tag{2}$$

where for the *i*-th phase: α_i is volume fraction, ρ_i is real density, \vec{v}_i is velocity.

The equation of momentum conservation for the i-th phase can be expressed as:

$$\frac{\partial \alpha_i \rho_i \vec{v}_i}{\partial t} + \nabla \cdot (\alpha_i \rho_i \vec{v}_i \vec{v}_i) = -\alpha_i \nabla p + \nabla \cdot \vec{\tau}_i + \alpha_i \rho_i \vec{g} + \sum_i \vec{R}_{ij}, \tag{3}$$

where p is pressure, $\vec{\tau}_i$ is stress tensor in the i-th phase, \vec{R}_{ij} is the interaction force between the i-th and j-th phases. In equation (3) the stress tensor is written as:

$$= \frac{1}{\tau_i} = \alpha_i \mu_i \left(\nabla \vec{v}_i + \nabla \vec{v}_i^T \right) + \alpha_i \left(\lambda_i - \frac{2}{3} \mu_i \right) \nabla \cdot \vec{v}_i = \frac{1}{I}, \tag{4}$$

where μ_i , λ_i are shear and bulk viscosity, \overline{I} is unite tensor.

Interfacial interaction force is written as

$$\vec{R}_{ii} = K_{ii}(\vec{v}_i - \vec{v}_i), \tag{5}$$

where K_{ii} is interfacial interaction coefficient.

The equation of energy conservation for the *i*-th phase is:

$$\frac{\partial \alpha_i \rho_i h_i}{\partial t} + \nabla \cdot (\alpha_i \rho_i \vec{v}_i h_i) = \alpha_i \frac{\partial p_i}{\partial t} + \vec{\tau}_i : \vec{v}_i + \sum_i Q_{ij}, \tag{6}$$

where h_i is enthalpy of the *i*-th phase, Q_{ij} is heat transfer rate between *i*-th and *j*-th phases.

At movement of microspheroidal solid phase, consisting of granules, particles collide. As a result of the collision changes the kinetic energy of the granules. To account for the interaction of solids particles is used equation with the change of energy through a change in temperature of the particles [26]:

$$\frac{3}{2} \left[\frac{\partial \alpha_i \rho_i \theta_i}{\partial t} + \nabla \cdot (\alpha_i \rho_i \vec{v}_i \theta_i) \right] = \left(-p_i \vec{I} + \vec{\tau}_i \right) : \nabla \vec{v}_i + \nabla \cdot \left(k_{\theta i} \nabla \theta_i \right) - \gamma_{\theta i} + \phi_{ij}, \tag{7}$$

where p_i is granules pressure of the *i*-th solid phase, $k_{\theta i}$ is diffusion coefficient of granules energy, θ_i is granules temperature of the *i*-th solid phase, $\gamma_{\theta i}$ is dissipation of energy because of particles collision, ϕ_{ij} is energy exchange between the *i*-th solid phase and the *j*-th liquid (gaseous) phase.

Written system of equations is not closed. To close the system use relationships obtained from experimental studies of the fluidized bed.

In fluidized bed models the force of interaction between phases is written in the form (5), where the coefficient of interfacial interaction K_{ij} obtained in experimental studies, depending on types of the interacting phases. It is considered, that $K_{ij} = K_{ji}$. In the case of the interaction of two phases liquid or gaseous type the resistance is determined by the Schiller and Naumann model [17]. In the case of interaction between two phases, that are liquid (gaseous) phase (the *i*-th phase) and the granular solid phase (*j*-th phase) in (5) used Syamlal and O'Brien model [18]. In the case of interaction between two solid granular phases in equation (5) interfacial interaction coefficient is taken according Syamlal O'Brien model [19].

The term $\gamma_{\theta i}$ in equation (7) to the temperature of solid phase particles granules characterize the rate of energy dissipation in the *i*-th solid phase caused by collisions between phase particles. Energy dissipation due to the particles collision is written in the expression, obtained in Lun et al. model [20]. Coefficient of heat transfer intensity between *i*-th and *j*-th phase is determined for the interaction of the two liquid phases by expression of Ranz and Marshall model [21, 22]. In the case of granular solids for Nusselt number calculation at the heat exchange between the *i*-th solid phase and *j*-th liquid (gaseous) phase use an expression from the work of Gunn model [23]. For the heat exchange intensity balance relations $Q_{ii} = -Q_{ii}$ and $Q_{ii} = 0$ are performed.

The motion considered in the reactor at the accepted loads of gas feedstock and parameters of the used catalyst was turbulent. In the model allowed to calculate solutions used $k - \varepsilon$ turbulence model in which the motions of "secondary" granular solid phases were generated on the background of the turbulent motion of "primary" gas phase.

3.2 Chemical reaction model

Let us consider the basic chemical reaction only. It is described by the relation (1). The equation for the reaction product of can be written as:

$$\frac{\partial m_{prod}}{\partial t} = V_{cat} \cdot \rho_{feedstock} \cdot f(T_{feedstock}), \qquad (8)$$

where m_{prod} is mass of the dehydrogenation (isobutylene) reaction product, V_{cat} is volume fraction of catalyst, ρ_{prod} is the product (isobutylene) density, $f(T_{feedstock})$ is function that determines the conversion depending on the feedstock gas temperature.

The function $f(T_{feedstock})$ is based on the approximation of the experimental data and is:

$$f(T_{feedstock}) = 734.9314 - \frac{19702.791}{\sqrt{T_{feedstock}}}. (9)$$

The dependence of the functions in the range of the temperature change from 550 to 650 °C is presented in Figure 2.

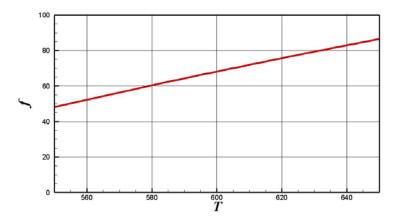


Figure 2: Function of conversion depending on the feedstock gas temperature.

3.3 CFD solver

Differential equations that describe the hydrodynamic and thermal processes in the field of computational model of the reactor were solved in CFD ANSYS Fluent package for axisymmetric unsteady flow scheme. The entire computational domain, shown in Figure 1, was divided into elements of a triangular or rectangular shape in different subregions of the computational domain, the dimensions of which were sufficient to determine the characteristic of the investigated phenomenon factors. For specified initial distribution of the catalyst in the reactor, after a while the flow entered a quasi-stationary mode. Hydrodynamic and thermal characteristic pictures of the reactor work calculated on this mode. Mass transfer due to the chemical reaction is implemented as a plug-in user-defined function (UDF). Time to quasi-stationary mode of one calculation depended on the conditions of the computational experiment and the average value of the order of 60 seconds of real time reactor's unit operation.

In each of the selected items are solved described above equations of the mathematical model of fluidized bed. Accordingly, the partition into a greater number of elements leads to a better accuracy of the calculations, but also leads to increase in the required computer resources. In this work, in calculations carried out the average number of finite elements was 500 thousand. Partitioning into finite elements near the gas feedstock supply models or arrays of nozzles built small enough to maximally correct reflect features of hydrodynamics and heat

and mass transfer relatively to the processes taking place in the operating chemical reactor. The average size of discrete element faces is 5 mm.

Boundary conditions were set on all elements of each of the constructed model of the reactor unit in accordance with the operating mechanism and used the solver. As the reactor model assumes rotationally symmetric, so on the axis of the constructed model was set conditions of axial symmetry. On all impermeable surfaces were conditions as walls. On models of gas feedstock supply nozzles and the outlet of the centrally located pipe of catalyst feed used velocity-inlet conditions (the value of the flow rate). Velocities were set according to the balance of the gas and catalyst flow rate. Temperatures of gas supplied and the catalyst were placed at 550 °C and 650 °C respectively. Outflow conditions (free output stream) were set in the area at the top of the reactor model for a gas outlet and at the bottom zone of the reactor to simulate the flow of catalyst into the regenerator. Also, through the part of free outlet stream at the top of the reactor there is a remove of fine catalyst particles. In the operating reactor particles from the top part return back into the working area over lattices with the help of cyclones. In constructed model on the output channels of cyclones were set velocity-inlet conditions in accordance with a flow rate of catalyst through the outlet channel in the upper part of the reactor.

4 RESULTS AND DISCUSSION

4.1 Results for base case of reactor

First were performed calculations for monodisperse catalyst. Particles size was selected of 100 microns. Operating time of the reactor unit is 90 seconds. Further calculation for two-fractional catalyst was carried out: a coarse fraction is 100 microns (80%), fine fraction is 50 microns (20%). Figure 3 shows the field of catalyst concentration and volume fraction of the reaction product of: \boldsymbol{a} is for monodispersed catalyst, \boldsymbol{b} is for polydisperse catalyst. The blue color corresponds to the absence of the phase under consideration.

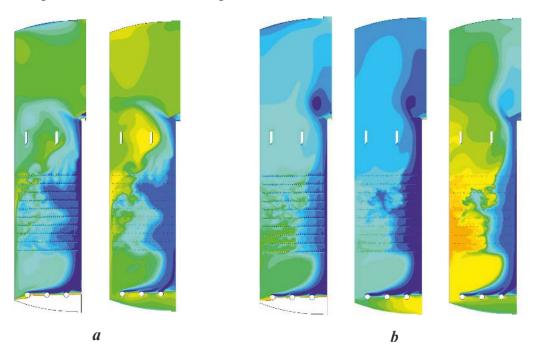


Figure 3: Calculation results.

It is evident that product formation corresponds to the catalyst concentration. In both cases there is an area with a low concentration of catalyst and product around the central tube. Detailed analysis has shown that we can see a downflow along pipe of incoming catalyst at a rate of about 2.5 m/s. In general, the presence of fine fraction contributes to a more intensive circulation and more uniform distribution of the catalyst in the unit. The volume of the reaction product located in unit in the case of monodisperse catalyst is 40% and for the case of polydisperse catalyst is 44%.

4.2 Reactor optimization variants

Two versions of a slight unit modification have been proposed to improve performance. The aim was to redirect flow downward along the tube of incoming hot catalyst into the central zone of the reactor. Following this purpose two variants of modifications were suggested.

- a) The dispenser on pipe outlet, which should increase penetration of the catalyst into the heated reactor zone midway between the central pipe and the outer wall. Considered a deflector on the end of catalyst feed pipe.
- b) Deflectors along the tube in the lattices zone provide catalyst redirection in the inter-lattice space. Number between all the lattices. Slope 45 degrees. Length so that the edge of the deflector cover the edge of the lattice. Position above the lattice, with a small gap between deflector and the lattice.

Figure 4 shows volume fraction of the reaction product fields for monodisperse and polydisperse two-fractional catalyst: a is the base of the reactor configuration; b is the presence of the deflector at the end of the catalyst feed pipe; c is the case with deflectors along the central tube

Table 2 shows calculation results for volume fraction of the reaction product in the unit without modifications and with two variants of slight modifications.

Calculation variant	Basic variant		Modification 1		Modification 2	
	mono- disperse	poly- disperse	mono- disperse	poly- disperse	mono- disperse	poly- disperse
Volume frac-						
tion, %	40	44	49	42	36	41

Table 2: Volume fraction of the reaction product for different variants of the reactor constructions.

It is shown from Figure 4 and Table 2 that the presence of fine particles, zone of reaction product formation and the volume fraction are just about the same for all three cases. This is due to the fact that the presence of fine phase determines circulating flows in the unit, pushing and pulling the larger particles. Since the fine particles are moving fast on the carrier gas background phase, then they are less sensitive to small structural modifications.

A completely different picture is observed while considering in numerical simulation only one averaged fraction of coarse particles. The coarse particles have more inertia and respond to the geometrical features of the reactor unit. In original variant of the apparatus one global circulation flow on the whole volume of the unit is formed. The use of deflectors on the tube (modification 2) forms a main circulating flow in the upper part of the apparatus and a few smaller on the bottom. Such modification degrades the production at the bottom unit, and decreases overall efficiency. Throw the coarse particles in the middle zone of the reactor (modification 1) showed the best result because of breaking a single global circulation flow.

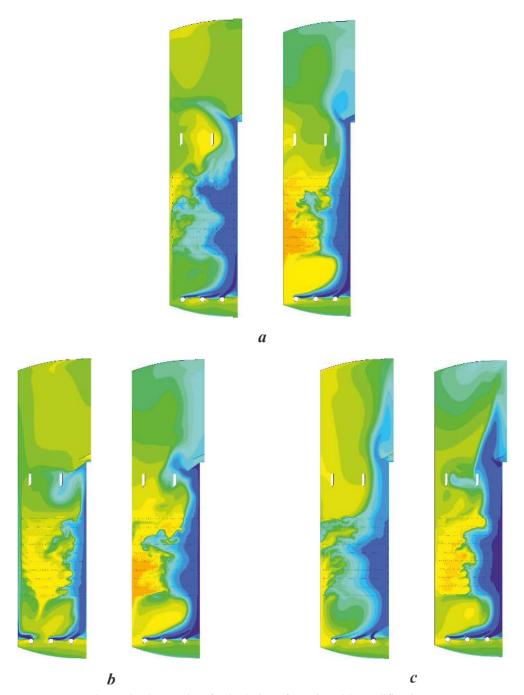


Figure 4: The results of calculations for units with modifications.

Calculations in this work were made for the averaged coarse fraction of catalyst of 100 microns, with an average speed of the cross section of the apparatus of 0.3 m/s. Note that the sensitivity of circulating flows formation of the reaction product does not depend only on the apparatus geometrical characteristics, but also on the fraction of the catalyst composition. The particle size of the main coarse fraction at specified gas flow rate will also determine the apparatus efficiency.

5 CONCLUSIONS

- A model of large-scale industrial fluidized bed reactor is constructed. The mathematical model is based on the Euler approach in describing discrete phase of particles motion as a continuous. Numerical implementation was carried out using Ansys Fluent software.
- A simplified chemical reaction model of isobutane to isobutylene dehydrogenation is constructed. The model is implemented and connected to a user-defined function (UDF).
- The fields of catalyst concentration, temperature field and the field of dehydrogenation reaction product were obtained. The analysis of circulation flows was carried out.
- The effect of slight structural modifications on the efficiency of product formation is investigated. The change of circulating gas flows and catalyst due to the change in apparatus construction is shown.
- The analysis of catalyst fractional composition effect on the efficiency of the reaction product was carried out. The sensitivity to the formation of circulating flow on the range of particle size is shown.

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